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Institute of Advanced Energy
Kyoto University

ANNUAL REPORT

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2007

京都大学エネルギー理工学研究所

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Kyoto University

Gokasho, Uji, Kyoto 611-0011
Japan

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FOREWORD

National universities including Kyoto University was turned into independent administrative entities in 2004. Four years have passed since then. We have continued our efforts toward further progress of the Institute under the new system in the first mid-term period (2004-2009). The efforts have yielded fruitful results in the fields of plasma, bio-, and photo- and quantum energies of advanced energy science and technology. In 2007, the ADMIRE (Application of DuET and MUSTER for industrial research and engineering) project was adopted as the “Project for industrial innovation by using advanced-research institutions” of the MEXT. The successive and extensional activities have been developed after the end of the 21st COE program (Establishment of COE on sustainable energy system, 2002 to 2006). The research activities and the external research funding were sound. Antiseismic reinforcement work for the main building started with the projected period of four years. The work for our institute's area is scheduled in 2008 and 2009, and it may affect our research activities.

We are now in the phase that we make a plan for the second mid-term period starting at 2010 based on our fruitful results and activities in the first period. The situations in energy supply and global worming are becoming worse. Energy issues are urgent priorities to be addressed under the present circumstances. We are going to work hard in research and education to cope with these critical energy issues. We strongly believe that it is our mission to challenge this difficult task and contribute to the benefit of the future generation. The mission of realizing CO_2-independent energy systems is our target of the second mid-term period.

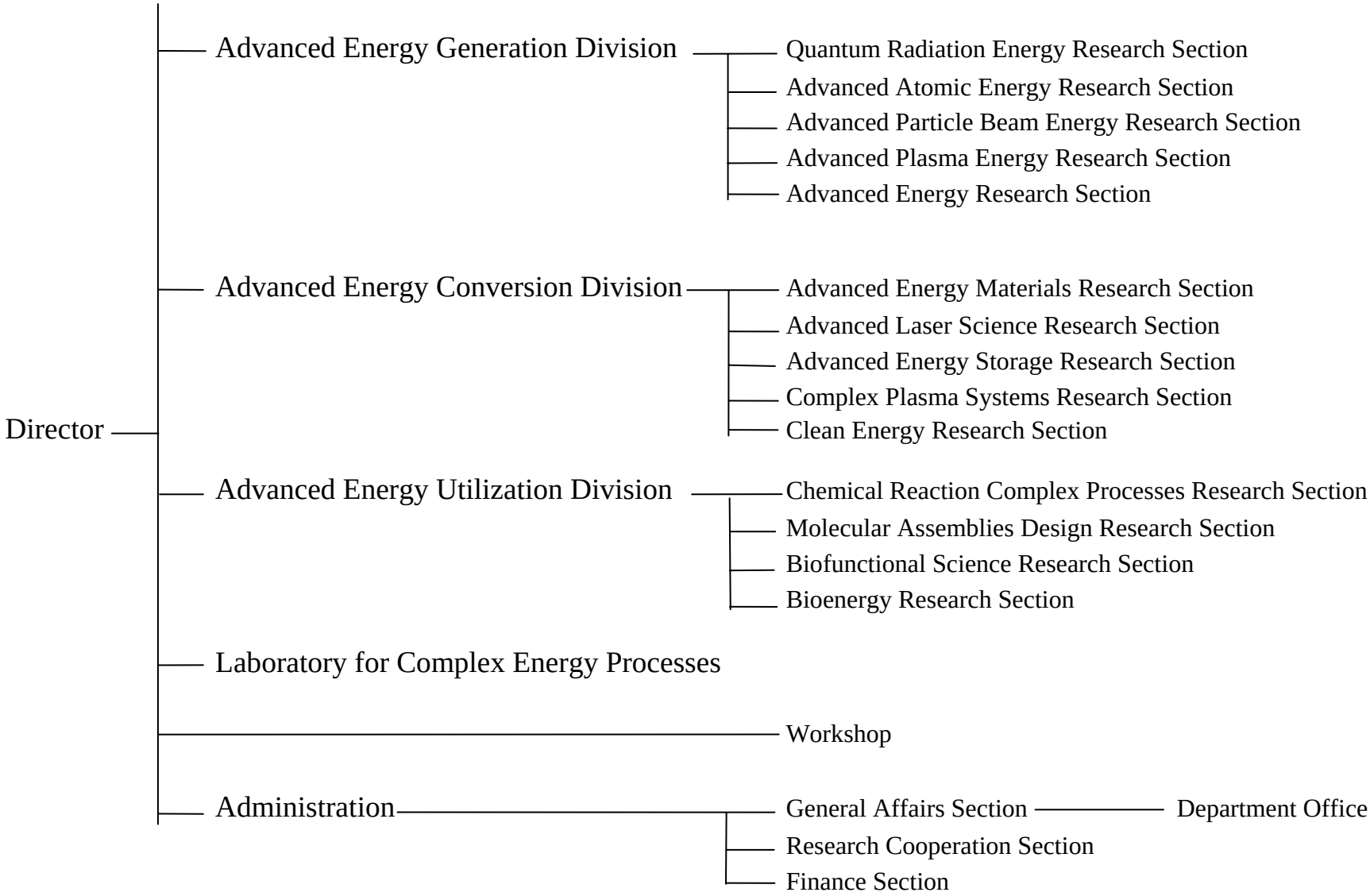
It is our great pleasure that this Annual Report would provide you a better understanding of the current activities of the Institute of Advanced Energy, Kyoto University.

March 2008

（署名）

Yukio H. OGATA
Director
Institute of Advanced Energy
Kyoto University

ORGANIZATION CHART OF THE INSTITUTE OF ADVANCED ENERGY



BRIEF HISTORY OF THE INSTITUTE OF ADVANCED ENERGY

The Institute, established in 1971 as the Institute of Atomic Energy, was renamed on May 11, 1996, to the Institute of Advanced Energy with the consolidation of Plasma Physics Laboratory, to represent its research interests in advanced and socially acceptable energy systems in the entire processes of energy generation, conversion and utilization. The former Institute of Atomic Energy, the predecessor of which was the Engineering Research Institute founded in 1941 as originally seeking to carry out synthetic studies of engineering through the cooperative work of specialists in different fields, has sought since 1971 to assume part of the responsibility for peaceful applications of atomic energy and has performed various kinds of fundamental researches in nuclear engineering.

The recent rapid expansion of the research fields at the Institute of Atomic Energy toward various fields of energy sciences, such as systems engineering for social and environmental energy systems, advanced energy conversion researches in quantum engineering, physico-chemistry and materials science, consequently has brought about the new institute, i.e., the Institute of Advanced Energy in search of advanced and socially acceptable energy systems.

The Institute of Advanced Energy had joined to the Kyoto University 21st Century COE program named "Establishment of COE on Sustainable Energy System" from 2002 with Graduate School of Energy Science and Radio Science Center for Space and Atmosphere, Kyoto University. The 21st Century COE program was successfully completed in FY 2006. At the moment, extension of the COE program, International New Energy Cooperative Organization is going to be established.

In FY 2004, the structural reform of the national universities has started to introduce innovative and reasonable systems, completely different from the old systems.

Professors of the Institute give lectures on their specialized fields to students of the Graduate School of Energy Science. Graduate students in various specialized fields of energy science prepare their theses for Master's or Doctor's degree at the Institute under professorial guidance. The Institute also provides research opportunities to those who specialize in advanced energy science and engineering, and related fields covered by the academic staff. The educational activities of the staff are described in their respective sections.

The institute is located on the Uji campus of the university about 20 km south of Kyoto city. Additional facilities, for researches such as advanced materials, advanced chemistry, magnetoplasma, plasma direct energy conversion, plasma physics, and fusion engineering, are located in the campus with individual buildings.

The publications are renewed into the followings;
(1) Annual Report of the Institute of Advanced Energy, Kyoto University, (in English).
(2) Research Report of the Institute of Advanced Energy, Kyoto University (occasionally, in English).
(3) News Letter of the Institute of Advanced Energy, Kyoto University, (three issues a year, in Japanese).

The Institute consists of the following three main research divisions each containing four research sections, and an attached laboratory, with two visiting staff research sections in addition.

It is strongly encouraged for every research section or division to make joint collaborative researches with other divisions, to cope with, in particular, energy-related interdisciplinary critical issues we are facing at present or in the future to come.

I. ADVANCED ENERGY GENERATION DIVISION
(study publicly acceptable high quality energy generation of advanced energy)
- (a) Quantum Radiation Energy Research Section
- (b) Advanced Atomic Energy Research Section
- (c) Advanced Particle Beam Energy Research Section
- (d) Advanced Plasma Energy Research Section
- (e) Advanced Energy Research Section (foreign visiting professor)

II. ADVANCED ENERGY CONVERSION DIVISION
(study of improvement of efficiency and other performances in the functional energy conversion processes)
- (a) Combines and Varying Conversion Processes Research Section
- (b) Advanced Laser Science Research Section
- (c) Advanced Energy Storage Research Section
- (d) Complex Plasma System Research Section
- (e) Clean Energy Research Section (domestic visiting professor and associate professor)

Ⅲ. ADVANCED ENERGY UTILIZATION DIVISION
(study processes for high performance energy utilization)
- (a) Chemical Reaction Complex Processes Research Section
- (b) Molecular Assemblies Design Research Section
- (c) Biofunctional Science Research Section
- (d) Bioenergetics Research Section

IV. LABORATORY FOR COMPLEX ENERGY PROCESSES
(promote equipment designing, software development, and collaboration with domestic and foreign institutions for high quality energy processes, including generation, conversion, and utilization)

RESEARCH ACTIVITIES

Advanced Energy Generation Division

Quantum Radiation Energy Research Section

H. Ohgaki, Professor
T. Kii, Assistant Professor

1. Introduction

Coherent-radiation energy with wide wavelength tunability, high power and high efficiency is quite promising in the 21st century that is sometimes called the "era of light".

The research in this section aims at developing the technology to generate new quantum-radiation energy and apply the radiation in various fields; atomic energy including plasma heating, energy transportation in the universe, material science, material synthesis, electronic device, medical and biological science, etc.

Free-electron laser (FEL) is one of the powerful candidates for the new quantum radiation, and it is sometimes called the light source of next generation.

2. Construction of an IR-FEL facility KU-FEL

FEL is regarded as a light source of the next generation because of its wide wavelength tenability where the conventional lasers cannot reach, potential high efficiency, and high power. However, the system is usually much larger and the cost is higher than conventional lasers. We are going to overcome these difficulties by exploiting an RF (radio-frequency) gun, energy recovering system, etc.

2.1 KU-FEL

The FEL system consists of a 4.5 cell thermionic RF gun, 3 m accelerator tube, beam transport system, and an undulator. Fig. 1 shows a schematic drawing of the system. The FEL system has been completed in the Laboratory for Photon and Charged Particle Research. Beam commissioning has been also successfully finished.

2.2 First lasing

The first lasing experiment has been made with the electron beam of 25 MeV and the undulator gap of 25.5 mm. Fig. 2 shows a temporal evolution of the electron beam current measured at the upstream of the undulator and the optical output from the optical resonator. As shown in fig. 2, the optical output steeply increased at the end of electron macro beam pulse, and its peak strength was about 50 times larger than the spontaneous radiation. The FEL gain was 16% and the optical loss was 2.8 % respectively.

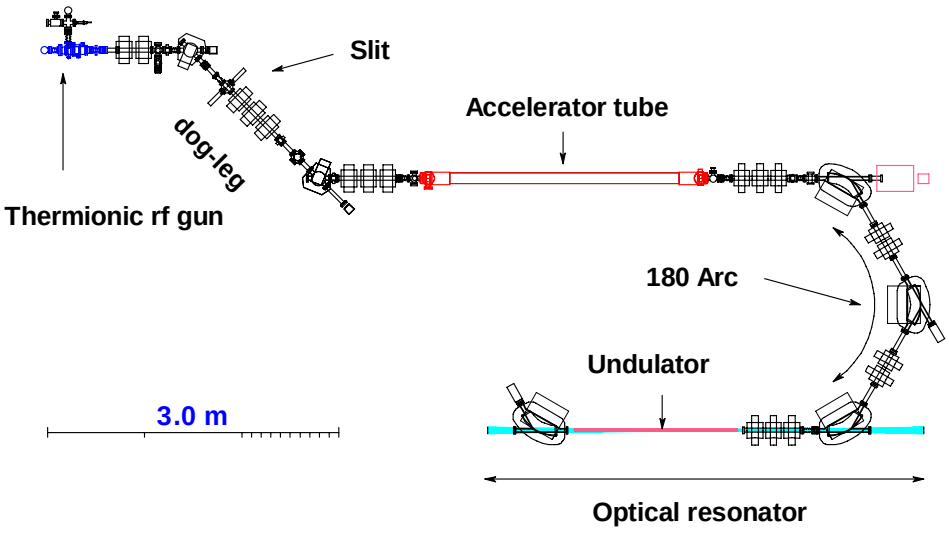


Fig. 1 Schematic drawing of the KU-FEL

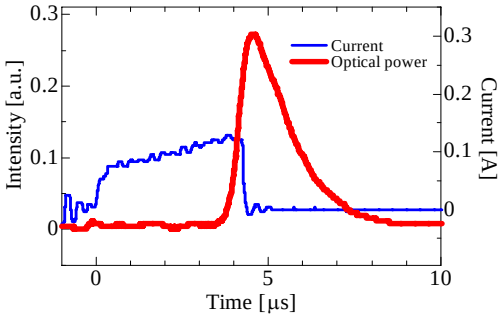


Fig. 2 Temporal evolutions of the electron beam current and the optical output

Spectra measurement was also made to ensure the FEL lasing. Fig. 3 shows an experimental setup for spectrum measurement of the FEL output. An optical resonator, a monochromator: DK240 (Digikrom Inc.) and an InSb IR detector : J15D12-M204-S01M-60 (Judson Inc.) were aligned using a semiconductor laser. The electron beam trajectory was carefully controlled to optimize strength of spontaneous radiation using beam profile monitors and steering magnets.

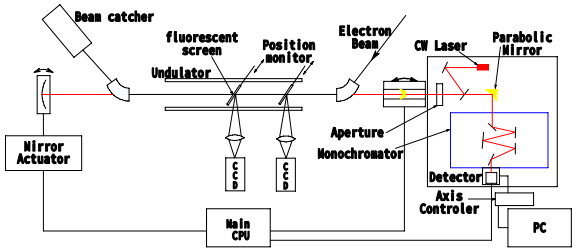


Fig. 3 Experimental setup for measurement of the FEL output

A measured spectrum of the FEL output is shown in fig. 4. As shown in fig. 4, spectrum width is narrower than 100 nm at 12300 nm, where the width of the spontaneous radiation is 470 nm. Therefore exponential growth of the optical output and linewidth narrowing are the specific features of the FEL lasing, the first lasing was successfully observed at the KU-FEL.

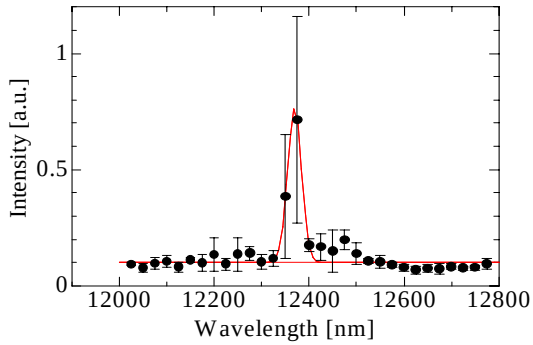


Fig. 4. Spectrum of the FEL output

3. Improvement of thermionic RF gun

A thermionic RF gun is suitable for a compact and economical FEL system. However it is difficult to produce electron beam of pulse width longer than a few μsec with constant energy owing to inherent back-bombardment effect. To overcome the difficulty, we tried to reduce beam energy degradation due to the back-bombardment effect by feeding modulated RF power into the thermionic RF gun using remotely controllable pulse forming network (PFN) of Klystron modulator. In addition, phase drift due to the PFN modulation was compensated using an electric phase shifter.

3.1 Energy compensation

Fig. 5 shows the experimental setup. The cathode is of 2-mm diameter LaB_6 crystal. The RF power fed into the RF gun is controlled by remotely adjusting the reactors in the PFN with stepping motors. The effect of the modulated RF input was evaluated experimentally by comparing the temporal evolution of the energy spectrum of electron beam measured with a bending magnet, a beam slit, and a Faraday cup (FC).

Fig. 5 Experimental setup

The input RF power amplitude modulation was applied to compensate the energy degradation due to the back-bombaredment effect. Time evolutions of the applied voltage to the Klystron and the input and reflected RF pulse and extracted beam current are shown in fig. 6. As increasing the extracted electrons from the RF gun, the RF amplitude is increased by controlling the PFN parameter.

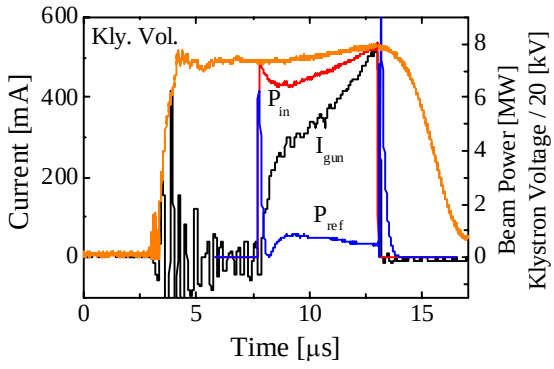
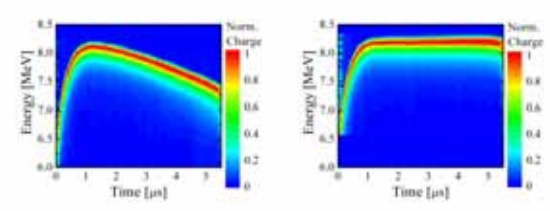


Fig. 6. Time evolution of the Klystron voltage (Kly. Vol.), the input and reflected RF power (P_{in}, P_{ref}), and the extracted beam current from the RF gun (I_{gun})

The temporal evolutions of the extracted electron beam are shown in figs. 7 a) ,b). As shown in figs. 7, peak energy was successfully kept constant during the macro-pulse. As a result, macro-pulse duration effective for FEL lasing was increased from 800 ns to 4000 ns as shown in fig. 8.



a) without amplitude modulation, b) with amplitude modulation

Fig. 7 Temporal evolutions of the energy spectrum of the electron beam

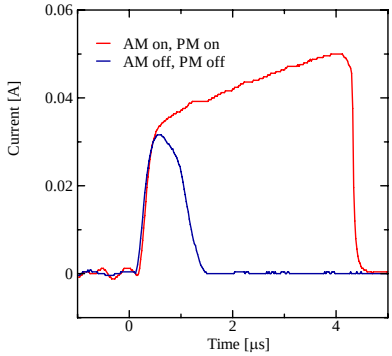
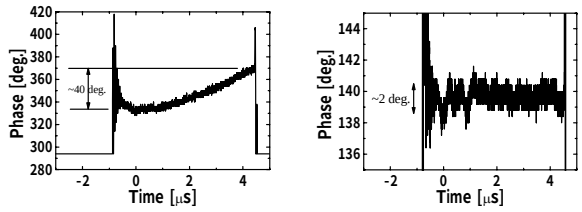


Fig. 8 Wave form of the electron beam injected to the accelerator tube

3.2 Phase compensation

Although the energy compensation by controlling the PFN parameter seems to be quite effective for the FEL lasing, phase advance due to the change of the electron bema velocity in the Klystron tube is introduced. Consequently, phase difference between the RF gun and the accelerator tube arises, and thus the energy spread is increased after the acceleration by the accelerator tube. To reduce the growth of the energy spread, an electric phase shifter was introduced to the low-level RF system for the RF gun. The phase drift during macro-pulse was successfully reduced from 40 to 2 degree as shown in figs 9 (a),(b).



(a) before phase compensation (b) after phase compensation

Figs 9 Temporal evolution of the phase difference between standard RF from signal generator and input RF to the RF gun.

4. Development of photocathode RF gun

To reduce the back-bombardment effect in the 4.5-cell RF gun, several attempts have been made, and the macro pulse duration of 5 μs has been achieved. However, several efforts are needed both to extend the macro pulse duration and to increase the peak current to reach the FEL saturation. Replacing the thermionic RF-gun to a photo-cathode RF-gun is the most promising way to obtain a high peak current electron beam. Therefore, we made a preliminary design study and started to develop a 1.6-cell photo-cathode RF-gun.

Recently, the improved design of the photo-cathode RF-gun has been installed in the linac in AIST and succeeded to generate 100-bunch electron beam. The beam charge of 1.5 nC/bunch has already been achieved. A multi-bunch operation also succeeded with a 100 bunch UV drive laser and a Cs_2-Te cathode. Manufacturing of the photo-cathode RF-gun have been successfully carried out under the collaboration with KEK, AIST and Waseda University. The photo-cathode RF-gun will be installed into the KU-FEL system in the next financial year. The results of low level RF measurement are shown in fig. 10. The measured resonant frequency was 2856.91 MHz and the quality factor Q of the cavity was about 11000. The photograph of the RF gun is shown in fig. 11.

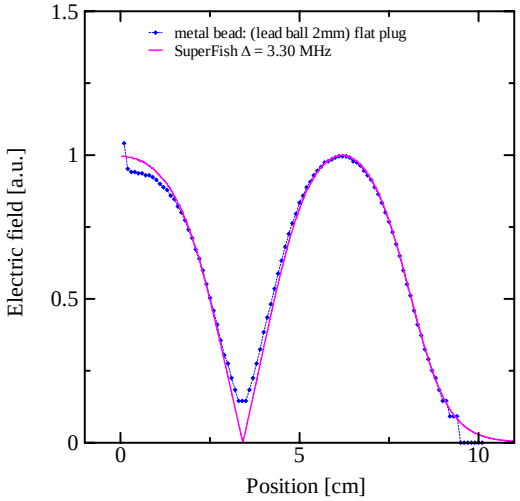


Fig. 10 Electric field distribution in the photocathode RF gun measured using microwave cavity perturbation technique.

Fig. 11 Photograph of the newly manufactured photo -cathode RF gun.

The 1.6-cell photo-cathode RF gun will be located at the upstream of the accelerator tube. Schematic view of the upgrade design of the KU-FEL with the photo-cathode RF-gun is shown in fig. 12. To evaluate the FEL performance with the photo-cathode RF-gun we made a start-to-end calculation by using PARMELA, ELEGANT and GENESIS which has been improved to take an optical cavity into account. The evaluated peak current of the electron beam at the undulator was 443 A when the RF phase was tuned to obtain the minimum energy spread, dE/E=0.054%, of the electron beam. When the RF phase was tuned for energy chirped beam, dE/E=0.86%, the peak current of the electron beam was 2.46 kA. Since the thermionic RF-gun generated 40 A electron beam, 10-50 times large peak current was obtained by the photo-cathode RF-gun. The output power of 12.1 μm FEL, 400 MW, will be saturated within 3 round-trips with the energy chirped beam. The large peak current from the photo-cathode RF-gun is also prospective for expanding the FEL tuning range at the KU-FEL.

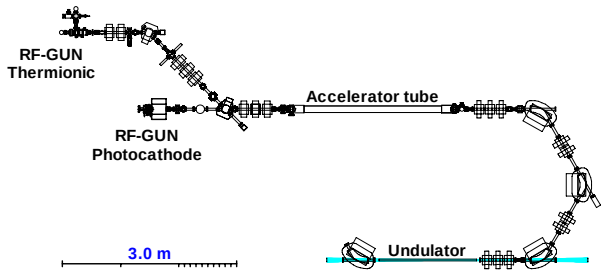


Fig. 12 Upgrade design of the KU-FEL

5 Design study on new type superconducting micro-undulator

A micro-undulator will be a useful device for a compact FEL device and/or a short wavelength FEL. To realize the micro-undulator with undulator parameter K=1 for period of 5 mm, periodic transverse magnetic field B_0 should be almost 2 T. To obtain strong periodic transverse magnetic field in a short period, we have stared design study of high T_c superconducting micro-undulator.

Fundamental properties of the hybrid staggard array structure, shown in fig. 13 were studied using a three-dimensional magnetostatic code RADIA.

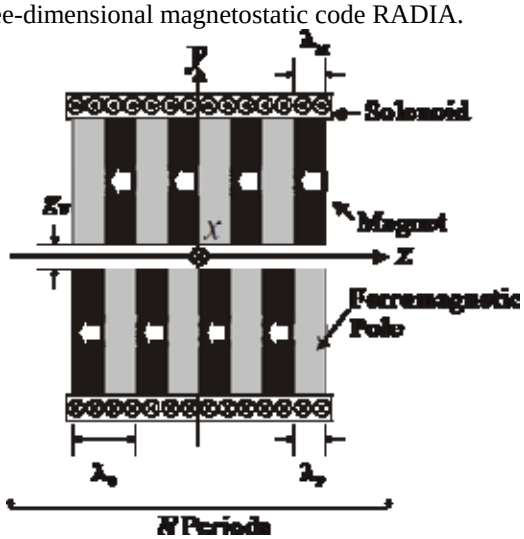


Fig. 13. Conceptual drawing of the high T_C hybrid SC undulator.

It was numerically found that the transverse magnetic field B_0 is variable as strength of the solenoidal magetic field as shown in fig. 14. The maximum undulator field B_0 is 0.93 T for the 10 T bulk superconducting magnet. It was also found that the maximum undulaor field is scalable as the strength of the superconducting magnet. Although there are several difficulties on fabrication of the high T_c hybrid SC undutaor, such as, operating stability, flatness of the magnetic field, etc, it is quite prospective novel undulator for extreme high-field compact insertion device for future light sources.

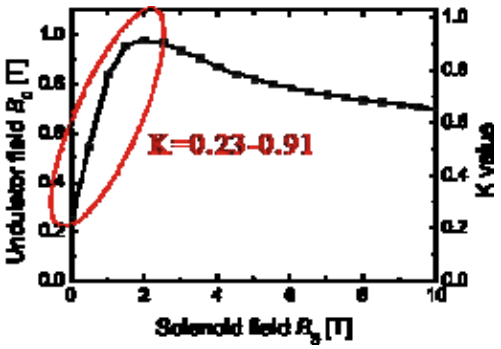


Fig. 14. The undulator field B_0 as the function of the solenoidal field B_s

Advanced Energy Generation Division

Advanced Atomic Energy Research Section

S. Konishi, Professor
Y. Yamamoto, Associate Professor
Y. Takeuchi, Assistant Professor

1. Introduction

The major objective of the study in this field is to research advanced energy technology for sustainable growth under global environmental constraints. The studies described below are featured by the consideration between technical possibility of better suitable energy generation and conversion systems with advanced technology, and socio-economic analysis of future society and markets that actually requires and utilizes such energy. The activity of advanced atomic energy research section covers wide areas of science and engineering based on plasma physics, atomic physics, and nuclear physics. Presently, following topics are studied in this research section:

(1) Assessment of fusion energy and energy system design
(2) Study of the compact neutron source using newly developed cylindrical discharge device for neutron beam generation.
(3) Study of advanced fusion reactor blanket with liquid LiPb and SiC composite for early realization and high temperature output.
(4) Development of Intermediate Heat Exchanger for advanced nuclear energy with SiC composite.
(5) Hydrogen production from biomass using high temperature heat from fusion reactor

In the fiscal year 2007, various interesting results were obtained in the above activities. The followings report some of the topics.

2. Assessment of fusion energy

This study continued to analyze the future fusion electricity market in developing countries, where electrical characteristics of the grid is anticipated to be rather different from the ones in developed countries such as Japan. Activity in this year is expanded to the collaboration with the work of the KSI (Kyoto Sustainability Initiative) for the consideration from the aspect of sustainability and global environmental problem. Discussions with the Institute of Economic Research, Kyoto University and other KSI researchers yielded the analysis of the effect of fusion energy as the reduction of the carbon dioxide emission in the future energy system. It is obvious that in order to contribute the global warming problem, expected role of the fusion energy is to provide carbon- free energy source in the future to replace fossil fuels. To maximize its potential, it should be emphasized that the capability of fusion to generate hydrogen and other synthetic fuels are important. Total sale of fusion in the global market is also significant, while an impact on Japanese domestic market will be rather limited. International trade mechanism under the environmental constraint such as the CDM will be a possible business model for fusion to be deployed.

3. Study of the compact neutron source using newly developed cylindrical discharge device for neutron beam generation.

Cylindrical deuterium discharge fusion neutron source is studied. We proved that surface reaction on the electrode is the dominant process of fusion reaction by both numerical and experimental study as illustrated in the fig.1, where beam-beam, beam-background and beam-adsorbed particle reactions are quantitatively evaluated. Significant increase of neutron production

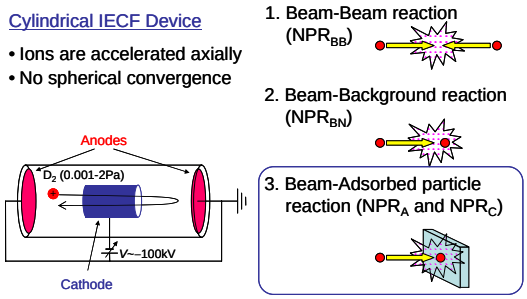


Fig.1 Collision processes in a discharge device.

was achieved by Ti coating on the electrode. Therefore this device is no longer IECF(Inertial Electrostatic Confinement Fusion) device but has a better performance with simple configuration as its modification. In this year we conducted a neutronics study to design a better configuration of reflector and moderator so that desired characteristics as a thermal neutron beam, that is expected in various practical applications. This is one of the major improvement from the traditional IECF that radiates neutrons

isotropically and difficult to form a beams. The numerical calculation that simulates atomic and molecular processes in this device describes all the collision reactions between ions, charge exchange neutrals. Simplified models are shown in the fig.2., the upper describes reaction on the titanium-coated electrodes and the lower illustrates the neutronics model used for the MCNP calculation. In this calculation, the surface of the electrodes are assumed to be covered with monolayer of deuterium atom. Axisymmetrical discharge device is contained in the cylindrical reflector made of various materials that collimates neutrons , and then slow them down at the moderator when lower energy beam is needed.

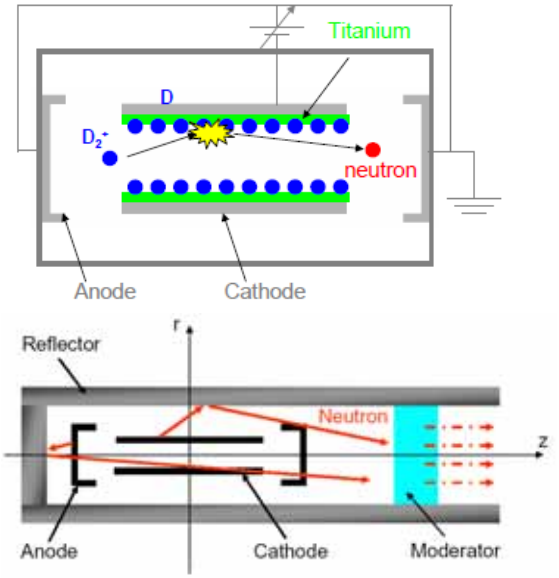


Fig.2 Models of the beam neutron source; the upper is the Ti coated discharge chamber, the lower is for the neutronics calculation.

Figures 3 show the typical results of the neutronics calculation that demonstrates the capability of this device to generate a beam. Upper figure 3(a) shows the capability of the reflector that focuses the neutrons to form a beam into longitudinal direction by gathering approximately 1 order of magnitude higher flux. In the figure 3 (b) the neutron fluxes calculated by the MCNP are shown in the two dimensional map. While neutron beam along the axis is focused, flux in other directions are reflected and shielded.

Energy spectrum of the generated neutron beam is another major characteristics to be considered. Figures 4 are the comparison of the effects of materials used for reflectors. As shown in the figure 4(a), with the metallic reflectors fast neutrons are kept as the majority. For lower energy, particularly thermal neutrons, light element materials such as water or graphite were found to be effective as reflectors as shown in the fig 4(b).

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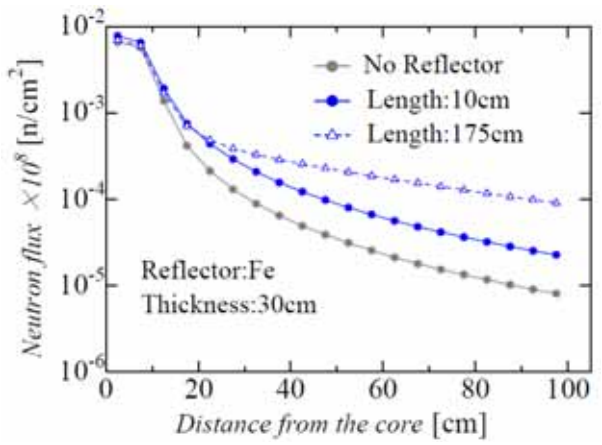


Fig.3 (a) Neutron flux along the axis with various reflectors.

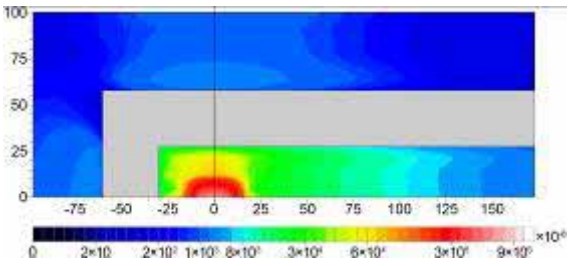


Fig. 3(b). Two dimensional neutron flux generated with the cylindrical reflector configuration..

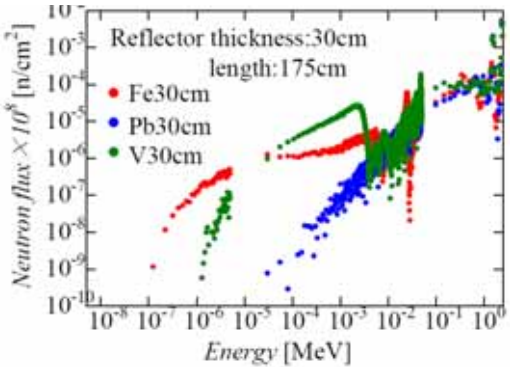


Fig.4(a) Energy spectrum with metal reflectors.

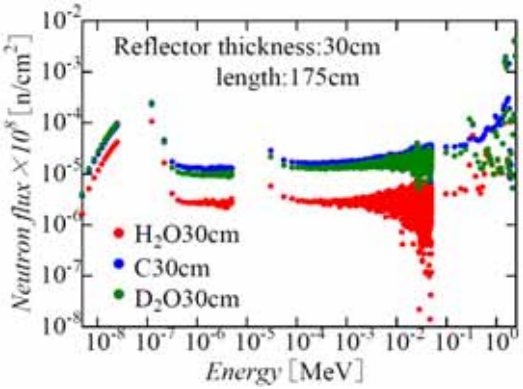


Fig.4 (b) Energy spectrum with light element reflectors.

The study proved the basic possibility to generate a neutron beam with a simple table top device based on fusion reaction. Further study is expected to achieve the capability required for various applications.

4. Production of Hydrogen using high temperature heat from future fusion reactor.

In order to realize hydrogen energy system that is free from fossil consumption and carbon- dioxide emission, it is essential to develop a large scale production method of hydrogen. We proposed a hydrogen production process from fusion energy and biomass as renewable resources, and perform a proof-of principle experiment as well as a system design study. In the previous years we have successfully proved that cellulose can be converted to hydrogen at high temperature. Endothermic reaction utilizes the fusion heat to be converted for chemical energy. This year we measured the effect of catalysts that converts biproducts of tar from the decpomposition of biomass to CO and H_2, as well as conversion of lignin, that is another major element of woody biomass. In the experiment shown in the fig.5, no catalyst was used for the biomass gasification part, and no solid residue was left after the reaction completed.

Maximum production of hydrogen and heat absorption by the reaction is also measured to be compared with the ideal reaction,

$$(C_6H_{10}O_5)+H_2O+814kJ = 6CO+6H_2 \quad .$$

The results are summarized in the figure 6, where gaseous composition of the reaction products with Ni, Co catalysts and the control without catalysts are shown as the function of temperature. Although cellulose decomposes almost completely above 900 degree where theoretical equilibrium calculation suggests, analyzed gaseous compounds such as CH_4, CO and CO_2 do not agree well when catalysts are not used, indicating residual tar or other larger molecules. Both Ni and Co metal converts carbon compounds almost completely. Production of hydrogen is also shown in the fig.7. Indeed very high yield of 80% are obtained with catalysts, there still is missing hydrogen.

Figure 8 shows the result of the same experiment with lignin, that has a nominal composition of $CH_{1.4}O_{0.3}$ and suspected to be more difficult to convert to light molecules, particularly H_2 and CO. As shown in the figure, high conversion ratio over 80 % to C1 molecules were achieved. Although no residual solid was observed, it was suggested larger molecules such as tar still remains as the byproduct. The value of 80% may be satisfactory for proof or principle experiments, further improvement is required for practical applications.

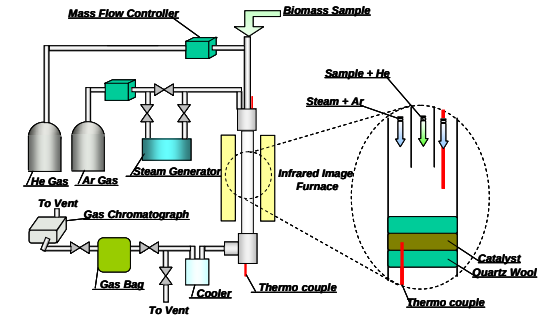


Fig.5 Experimental apparatus for biomass conversion.

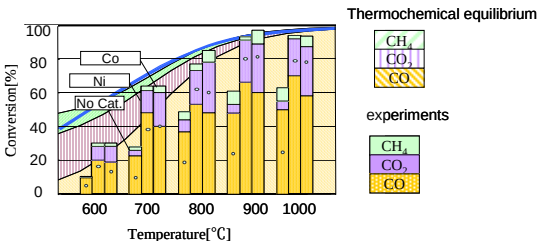


Fig.6 Gaseous products of Carbon compounds generated by the thermal decomposition of cellulose followed by various catalysts.

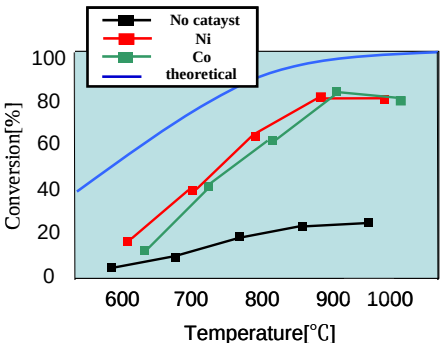


Fig.7 Hydrogen production as the results of the catalytic reaction

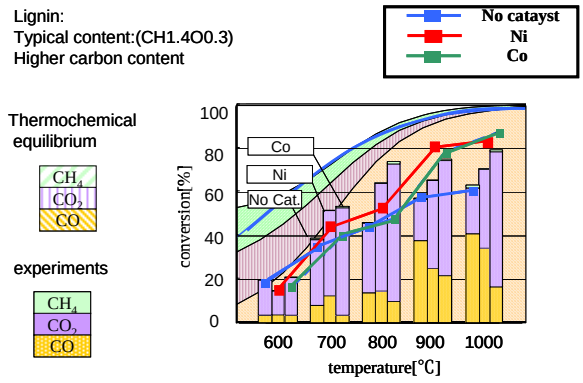


Fig.8 Gaseous products of Carbon compounds generated by the thermal decomposition of lignin followed by various catalysts

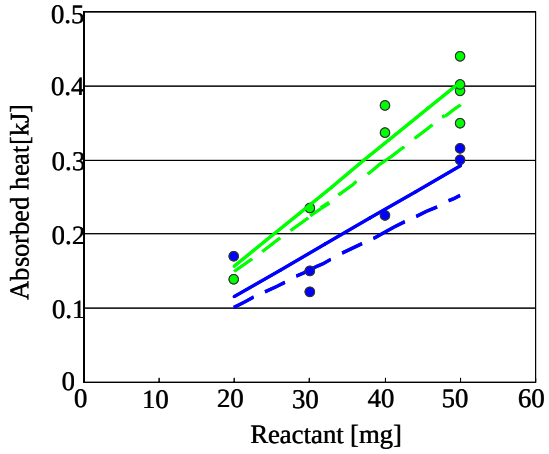


Fig.9 Heat adsorption of the biomass conversion using endothermic reaction.

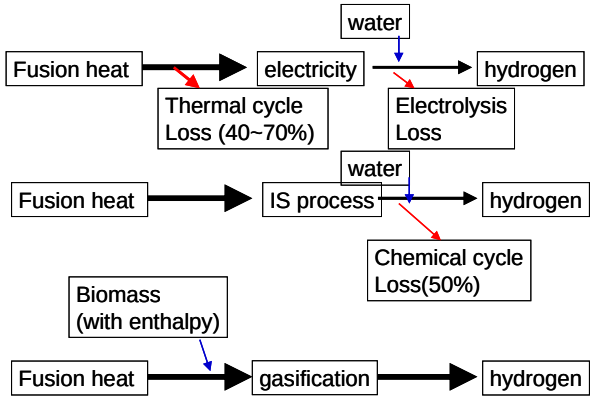


Fig. 10 comparison of fusion energy utilization with various conversion processes.

Heat adsorption was also measured with these experiments by comparison with known endothermic reaction of the decomposition of $SrCO_3$. The apparent heat efficiency was above 80% of the theoretical values required for the conversion of biomass to hydrogen.

Figure 10 illustrates the valious energy conversion processes for fusion based on electricity generation, thermo-chemical cycle, and the biomass conversion. The comparison supported by these experiments suggests hydrogen production by this process is potentially more efficient than that via electricity generation followed by conventional electrolysis, that is limited by the Carnot ' s efficiency and electrolysis loss. This is due to utilization of chemical energy of cellulose that is wasted by oxidation or combustion. This energy is supplied as a form of heat at the temperature approaching 1000 degree C that is only possible with advanced nuclear reactors and fusion.

5. Development of high temperature compact intermediate heat exchanger with advanced composite material

From 2005, we have performed the research project under a contract with the MEXT and JST on the development of compact intermediate heat exchanger with advanced SiC composite material for high temperature use, with subcontractors of Japan Science and Technology Agency and Mitsubishi Heavy Industries. Three years of the project have successfully completed, and the results of the first 2 years are opened for the public

This project has technical tasks of 1) Loop experiments to demonstrate the heat exchanger, 2)development of the heat exchanger scale model, 3)system design and analysis, 4) development of inspection technique.

Figure 11 shows the test loop of liquid LiPb metal to supply high temperature heat, and the test specimen of the heat exchanger model made of SiC composites. With the loop and the sample, compatibility, hydrogen permeability, and heat transfer are measured and evaluated.

Fig.11 Test loop of the liquid LiPb, and test specimen of heat exchanger structure made with SiC composites.

Advanced Particle Beam Energy Research Section

K. Nagasaki, Professor
K. Masuda, Associate Professor

1. Introduction

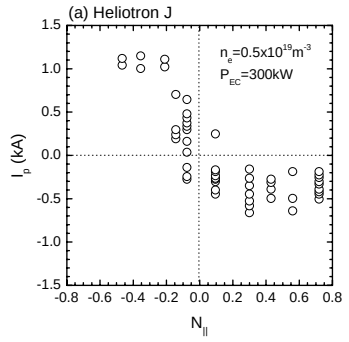
For the 21st century's advanced sciences and technologies to bring about enormous contributions to the human beings, advanced and innovative control methods for the collective behavior of charged particles are being developed. Studies of nonlinear interactions between charged particles and electromagnetic fields are particularly emphasized. The educational function specializes in energy conversion science and electrical engineering in the graduate and undergraduate schools.

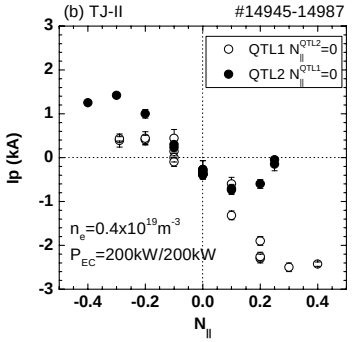
Main research subjects are now strongly focused on the followings; improvement and understanding of confinement and transport in fusion plasma produced and heated by electromagnetic waves, development of heating and current drive systems using high power millimeter waves, development and application of compact and portable neutron/proton sources driven by fusion plasmas, production and diagnostics of highly brilliant relativistic electron beams for advanced light sources such as free electron lasers.

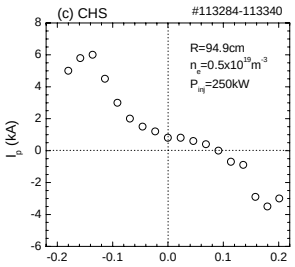
2. ECCD Experiments in Helical Systems

Electron cyclotron current drive (ECCD) is recognized as a useful scheme for stabilizing MHD instabilities and analyzing heat and particle transport. For example, in large tokamaks such as JT-60U, neoclassical tearing mode has been stabilized by localized ECCD, leading to the improvement of normalized beta. In S/H systems, on the other hand, the ECCD is expected to avoid dangerous rational surface by cancelling the bootstrap current particularly in low shear S/H devices. From the viewpoint of diagnostics, on the other hand, the S/H systems have the advantage of precise measurement of the EC current. The estimate of EC current is not so simple in tokamaks since a large amount of Ohmic current flows, and the effects of toroidal electric field and plasma resistivity have to be taken into account. In the S/H systems, we are able to measure the EC current with the accuracy of the order of less than 1 kA by using a conventional Rogowskii coil because of no Ohmic current.

As predicted from the ECCD theory, the amplitude and driven direction depends on the parallel refractive index, $N_{\|}$. Figure 1 shows the dependence of the toroidal angle on the EC injection angle. It can be seen that I_p increases with increasing $N_{\|}$, and saturates at a certain $N_{\|}$. ECCD is a main contribution to the total current since the bootstrap current is small due to the low pressure at $n_e = 0.5\times10^{19}$ m^{-3}. The flow direction is the one expected from the Fisch-Boozer effect opposite to $\mathbf{N}_{\|}$. The same tendency is observed in LHD when the injection angle is changed from the clockwise to counter-clockwise direction. The EC current amplitude is the same order, a few kA, up to now in all the devices, although the magnetic field structure is different. As discussed later, the toroidal direction of EC current strongly depends on the magnetic ripple structure. Under the condition for toroidal injection







scan, the Fisch-Boozer effect may be stronger than the Ohkawa effect. In the injection angle scan with the magnetic field fixed, the electron cyclotron resonance is Doppler-shifted due to finite $N_{\parallel}$, resulting that the decrease in electron temperature and/or the change in ripple structure possibly affects the EC current. The $N_{\parallel}$ dependence considering the Doppler shift effect is left for future.

The ratio of driven current to injection power, I_{EC}/P_{EC}, and the figure of merit, $\gamma=n_e I_{EC}R/P_{EC}$, are conventionally used for the estimation of ECCD efficiency. The drawback of these functions is that they have dimension, and they do not reflect the Te dependence. A figure of merit describing dimensionless ECCD efficiency including the Te dependence is proposed in the following form

$$\varsigma = \frac{e^3}{\varepsilon_0^2}\frac{n_e I_{EC} R}{P_{EC} T_e} = 32.7\frac{n_e I_{EC} R}{P_{EC} T_e}$$

where parameters have a unit of n_e in 10^{20} m^{-3}, I_{EC} in A, R in m, P_{EC} in W, and T_e in keV. This dimensionless figure of merit includes important parameters such as n_e and T_e. If ζ changes under the same plasma conditions, it means that ζ reflects the effect of electron thermal velocity and trapping. Table 1 summarizes the ECCD efficiency on Heliotron J, TJ-II and CHS. No result from LHD is included since it is not estimated yet due to short ECH pulse length compared to the current evolution time. It should be noted that these efficiencies are typical values ever obtained, not optimized ones. Although the magnetic field structure is different among the devices, the EC current amount is a few kA in all the devices, and the ECCD efficiency is similar within a factor of 2 when the magnetic field ripple ratio, B_{min}/B_{max}, ranges from 0.93 to 1.0. Rather low efficiency compared to tokamaks may be due to the strong Ohkawa effect enhanced by the magnetic ripple. The ray tracing calculation code is under development, which would clarify the role of trapped electrons by comparing between experiment and theory.

While the ECCD efficiency is not so high, the EC current is comparable to the bootstrap current, meaning that we are able to control the total toroidal current. Zero net current state has been demonstrated in Heliotron J. The total current is suppressed below 0.4 kA during the discharge by compensating the bootstrap current of 1.5 kA with ECCD. Such a state has also been demonstrated in CHS. In TJ-II, the EC current driven by each launcher is cancelled and the low current of I_{BS}=-0.5 kA is kept. The multi ECCD systems are useful for extending the current control range.

Table 1: Electron cyclotron current drive efficiency in Heliotron J, TJ-II and CHS.

	Heliotron J	TJ-II	CHS
Maximum I_{EC}	4.6 kA	2 kA	6 kA
$\eta=I_{EC}/P_{EC}$	14 A/kW	10-15 A/kW	35 A/kW
$\gamma=n_e I_{EC}R/P_{EC}$	~ 8x10^{16} A/Wm^2	~ 9x10^{16} A/Wm^2	~16x10^{16} A/Wm^2
$\zeta=32.7 n_{20} I_A R_m / P_W T_{keV}$	~0.05	~0.03	~0.04

3. Production and Control of Highly Brilliant Electron Beam for Free Electron Laser

An electron beam with high brightness, high peak current, and long macro-pulse duration is preferred for high power free electron lasers. In the Kyoto University Free Electron Laser (KU-FEL) facility, a 4.5-cell thermionic rf gun is being studied intensively to meet these requirements in an extremely compact and low-cost system using microwave resonant cavities.

For a higher peak current of the electron beam on which the FEL gain depends nonlinearly, the operational conditions of the rf gun and the successive bunch compressor magnets were optimized. Firstly, the dependence on the operational electric field strength in the gun was surveyed numerically. The result indicates that the highest peak current is around 23 A with the average field strength of 24 MV/m. Under this condition, the bunch lengths of the electron beams were then measured by using a streak camera. Figure 2 shows a typical measurement result, showing the temporal bunch shape. The optimal condition for the bunch compressor was surveyed experimentally as shown in Fig. 3. The measured minimum bunch length was ~2 ps, which was found as short as the temporal resolution of the measurement system. Since the bunch charge was 14 pC, the highest peak current was thus estimated to be more than 7 A, which is found still

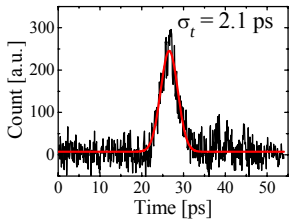
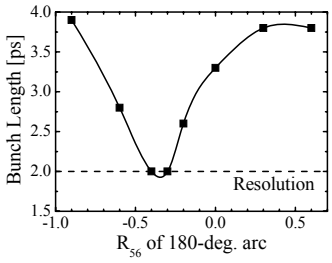


Fig. 2: Typical result of bunch length measurement by using a streak camera.



lower than the numerical prediction and than the target peak current of ~ 40 A for FEL lasing. Improvement in the bunch charge is thus mandatory for a higher peak current.

Thermionic rf guns in general suffer from the back-bombardment of electrons onto the cathodes. Due to this effect, the bunch charge and the macro-pulse duration show a trade-off relationship. For the KU-FEL rf gun, the bunch charge is limited to ~30 pC for a macro-pulse of ~3 μs, which are far below our targets for FEL lasing ranging 4-13 μm wavelength. An innovative method is thus needed to mitigate this problem.

For this purpose we have proposed a 'triode rf gun' with an additional short-gap cavity providing the extraction phase at the cathode independent of the main rf cavity fields. Numerical design studies have shown very encouraging predictions summarized in Table 2. Furthermore, the triode system can be driven by a moderate rf power of tens kW, which preserves the advantages of a linac based on a thermionic rf gun, namely extreme compactness and low cost.

The prototype design of the triode rf gun system has been completed (see Fig. 4), for the first proof-of-principle experiments in the KU-FEL gun. The rf power feed and phase control system of up to 100 kW capabilities has been prepared and tested. Also the triode cavity parameters, namely the quality factor, the shunt impedance, and the coupling coefficient with the rf feed coaxial cable were designed by taking into account both the available maximum field on the cathode surface and amplitude stability against the expected variations of the beam loading and cavity temperature. The triode cavity and the rf coupler are being fabricated, which will replace the demountable thermionic cathode set in the KU-FEL rf gun.

Table 2: Performance comparisons between the existing conventional rf gun and the newly designed triode rf gun (numerical prediction).

	conventional	triode
current density on cathode [A/cm^2]	30	80
bunch charge [pC]	29	59
peak current [A]	83	170
norm. emittance [π mm mrad]	2.0	1.6
macro-pulse [μs]	5	12

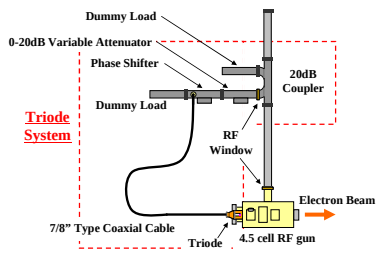


Fig. 4: Schematics of the triode rf gun system.

4. Inertial Electrostatic Confinement Fusion (IECF)

An inertial electrostatic confinement fusion (IECF) device consists of a spherical vacuum chamber as an anode with a D_2 or D_2-^{3}He fuel gases, and a highly transparent central cathode grid at a negative potential of ~100 kV. A glow discharge then takes place between them, thereby, ions produced and accelerated toward the center through the gridded cathode undergo D-D and D-^{3}He fusion reactions through beam-background gas collisions. The following equations show the fusion reaction channels for D_2-^{3}He fuels,

$$D + D \rightarrow {}^3He\ (0.82\ MeV) + n\ (2.45\ MeV),$$
$$D + D \rightarrow T\ (1.01\ MeV) + p\ (3.03\ MeV),$$
$$D + {}^3He \rightarrow \alpha\ (3.52\ MeV) + p\ (14.7\ MeV).$$

Of particular interest, the protons from D-^{3}He reactions are highly energetic and can potentially be used to produce radioisotopes for medical use and to generate mono-energetic γ-rays for versatile uses such as security inspection. As a neutron generator, the IECF device also shows advantages over conventional ones; long lifetime, long-term dc operation capability, easy operation not requiring an expert operator, and safety without radioactive isotope, all of which are essential for practical applications.

For landmine detection application, we have developed an extremely compact IECF neutron generator of 200 mm in diameter. We have also developed a BGO/ NaI-combined scintillation detector for the directional detection of the resultant γ-rays from explosives.

Performance tests of the developed landmine detector prototype were conducted for two kinds of wax-diluted explosives; TNT 240 g, TNT 100 g, RDX 100 g, RDX 29 g buried in the depths of 5, 10 and 15 cm, and soil moisture of 2, 10 and 18.5 wt%. Tests were performed in 7 trials each under several mixed conditions for 20 minutes measurement with the D-D neutron yield of ~1×10^7 n/s. As an example, the γ-ray energy spectra are shown in Fig. 5. The neutron-captured γ-rays of 10.8 MeV emitted from nitrogen in RDX can be clearly detected above the background, which will ensure that our landmine detection system is effective. In total under the various test conditions, the reliabilities of the landmine detection are found to be 0.77 for less than 18.5 wt% soil moisture, and 0.83, for arid soil (<10 wt%), respectively.

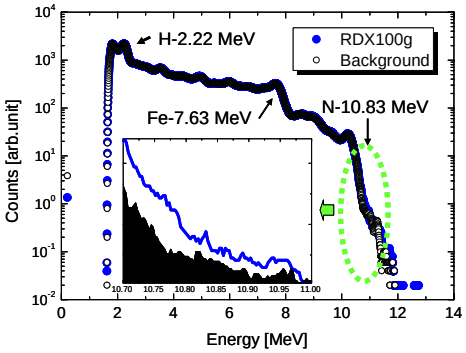


Fig. 5: BGO spectra for RDX 100g buried 15 cm deep in soil moisture of 10 wt%, and without RDX, i.e. the background.

These results reveal that the present neutron-based explosive detection system is basically effective to humanitarian landmine detection. Further improvements, particularly, in the judgment algorithm, will be needed for the practical outdoor demining.

In IECF researches aiming at drastically enhanced neutron/proton yields, understanding the intensity distribution of fusion reactions is one of the most intensive interests. For this purpose we adopted collimated proton measurements by the use of Si diode detector (SSD) and reconstruction scheme based on Maximum Likelihood Expectation Maximization method which reconstructed D-^{3}He reaction distribution successfully, while this diagnostic method has suffered from poor S/N ratio for D-D owing to the lower energy of protons.

Against the severe noise due to X-rays through the SSD, we improved the proton counting system (Fig. 6) by considering the collimation geometry, the choice of

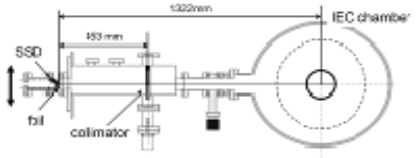


Fig. 6: Layout of the proton measurements with a movable SSD and collimator.

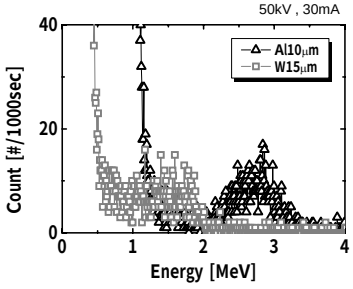


Fig. 7: D-D proton spectra by the use of two different [illegible]

the shielding foil in front of the SSD, and the SSD thickness. As the result, the D-D protons were successfully observed with a significantly enhanced S/N ratio as shown in Fig. 7. Preliminary experiments on the determination of the D-D fusion reaction distribution have shown a clear difference from the distribution of D-^{3}He reactions. Further details will be studied, e.g. dependences on operational voltage, pressure and current.

In the IECF device, electrons escaping from the plasma within the gridded cathode are accelerated, and they reach the chamber with a high energy according to the applied negative bias, which causes undesirable hard X-ray emission and also limits the efficiency, i.e. the fusion reaction rate over the input power.

In order to resolve these problems, we proposed a new IECF-based concept, i.e. the double-grid IECF device schematically shown in Fig. 8. It consists of a spherical anode grid at a potential of V_a in addition to the cathode at $-V_c$ and the grounded chamber. Most of the electrons are now expected to go through the anode and reach the chamber surface, and secondary electrons emitted from the chamber will then hit the anode. The electron energies contributing to the X-ray emission are thus supposed to be V_c and V_a, which are less than the energy of $V_c + V_a$ in the conventional IECF. Because of the strong dependence of X-ray emission on the incident electron energy, we thus expect that the X-ray flux can be greatly reduced.

We carried out preliminary experiments of the double-grid IECF with a deuterium gas. The diameters of the cathode, the anode and the chamber are 65, 250 and 340 mm, respectively. The bias voltages were V_c ~20 kV and V_a ~30 kV, and the dose equivalent rate of the emitted X-rays was measured. As the result, the X-ray flux was found to reduce as expected. The X-ray flux also shows a strong dependence on V_a and little dependence on $V_c + V_a$, despite the X-ray emission rate is known to be roughly proportional to V^4 in the energy range of tens kV. These results thus indicate that the electrons from the cathode mostly pass through the anode successfully.

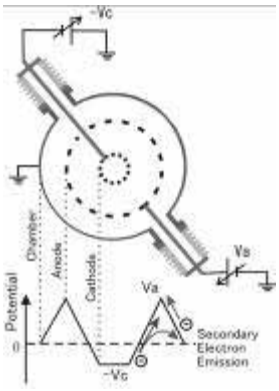


Fig. 8: Schematics of the double-grid IECF.

Advanced Plasma Energy Research Section

T. Mizuuchi, Professor
S. Kitajima, Lecturer
S. Kobayashi, Assistant Professor

1. Introduction

The current subjects of this research section are to study the properties of high temperature plasmas in order to control and improve the plasma energy confinement from the physical viewpoint of nuclear fusion research. The experimental and theoretical researches for the optimization of the helical-axis heliotron configuration are in progress under the collaboration with other groups of the institute and also the groups of other universities/institutes under the auspices of the Collaboration Program of the Lab. Complex Energy Processes, IAE, and the NIFS Collaborative Research Program,.

In this report, we describe some results obtained in the Heliotron J experiment in FY2007 focusing on (1) the global energy confinement in NBI plasmas with regard to the effect of the bumpy magnetic field component, and (2) condition of spontaneous transition in NBI plasmas of Heliotron J.

1. Effect of Bumpiness on Energy Confinement for NBI plasmas

The control of the magnetic field by adjusting its Fourier components is one of the key issues in the heliotron/stellarator configurations to achieve good energy and particle confinement. In Heliotron J, a helical-axis heliotron device, the importance of the bumpiness $\varepsilon_b = B_{04}/B_{00}$ on the collision-less transport has been investigated theoretically and experimentally. Here, B_{mn} is the Fourier component of the field strength with m/n mode numbers in Boozer co-ordinate system. The bumpiness has a role to reduce the $\nabla \mathbf{B}$ drift by aligning the mod-B_{min} contour with the flux surface. On the other hand, the collision-less ion orbit calculation predicts the bumpiness also has an effect to widen the loss cone angle. The bumpiness effects on the plasma performance has been studied by comparing the experiments with three different conditions of $\varepsilon_b(\rho = 2/3)$; 0.15 (high ε_b), 0.06 (medium ε_b) and 0.01 (low ε_b), The standard configuration of Heliotron J corresponds to the medium ε_b. The magnetic axis position ($<R_{ax}>$ = 1.2 m), the plasma volume (V_P = 0.7 m^3), the edge rotational transform ($\iota(a)/2\pi$ = 0.56) of three configurations are kept constant.

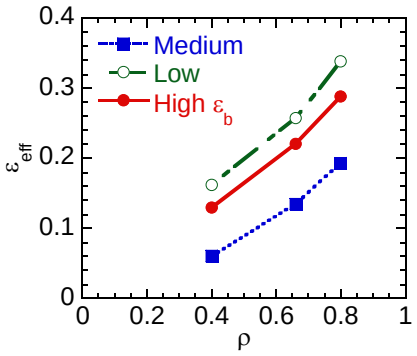


Fig. 1. Radial profile of effective helical ripple deduced by DCOM in high, medium and low ε_b configurations.

For ECH plasmas, the dependence of the energy confinement on bumpiness has been discussed from the viewpoint of the effective helical ripple ε_{eff}, which is used as an index of the particle diffusion coefficient in the collision-less regime. Figure 1 shows the radial profile of the effective helical ripple ε_{eff} in the three configurations deduced by DCOM coded. In the medium ε_b configuration, the lowest ε_{eff} is obtained, that is, the effective helical ripple at ρ = 2/3 in the high, medium and low ε_b cases are 0.22, 0.13 and 0.26, respectively. A favorable energy confinement for the ECH plasmas has been observed in the medium bumpiness configuration where the effective helical ripple was low.

Figure 2(a) shows the stored energy as a function of the NBI power (P_{NBI}) in the three configurations. The hydrogen neutral beam is injected tangentially into the deuterium plasmas after the initial plasma production by 70 GHz ECH. These data were obtained at the averaged electron density of ~ 2×10^{19} m^{-3}. The numerical calculation of the beam absorption profile by FIT-code predicts that the orbit-loss fractions of beam ions in high, medium and low ε_b cases are 25%, 26% and 29%, respectively, indicating a deterioration of the energetic particle confinement in the low ε_b configuration. The beam component in the stored energy is estimated to be less than 7%. In the high and medium ε_b cases, relatively high stored energy was observed than that in

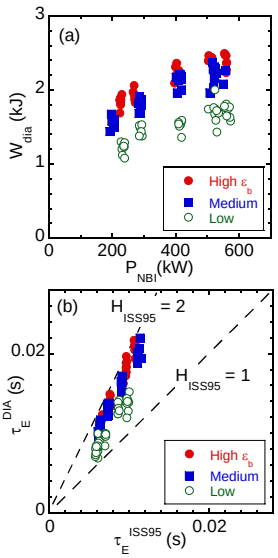


Fig. 2. (a) Stored energy obtained in high, medium and low ε_b configurations as a function of NBI power and (b) comparison between τ_E^{DIA} and τ_E^{ISS95} in NBI sustained plasmas.

the low ε_b case. Figure 2(b) shows the comparison of the energy confinement time τ_E^{DIA} with the International Stellarator Scaling (τ_E^{ISS95}). The enhancement factor of energy confinement to the scaling ($H^{ISS95} = \tau_E^{DIA}/\tau_E^{ISS95}$) was about 1.8, 1.7 and 1.4 in the high, medium and low ε_b configurations, respectively. In the ECH plasmas, H^{ISS95} around 2 has been obtained in the medium ε_b case and it has been about 1.4-1.6 for the high and low ε_b configurations

Figure 3(a) shows the bulk (deuterium) ion temperature T_i evaqluated by CX-NPA as a function of the NBI absorption power P_{abs} in the three configurations. An increase in T_i as increasing ε_b was observed, that is, the ion temperature in the high, medium and low ε_b configurations were 0.23, 0.20 and 0.18 keV at P_{abs} ~ 200 kW, respectively. The electron cyclotron emission measurement indicates increase in the electron temperature both in high and medium ε_b configurations as shown in figure 3(b). The electron cyclotron emission intensity (I_{ECE}) at the core region increased with P_{abs} in high and medium ε_b cases, while a weak dependence of the ECE intensity on P_{abs} was obtained in the low ε_b configuration. The observed improvement in the energy confinement in the two configurations can be explained qualitatively in terms of the increase in the ion and electron temperatures.

The difference in the energy confinement between

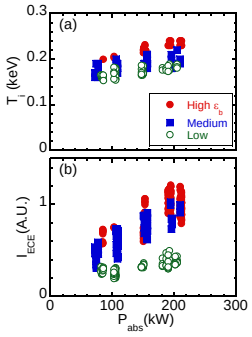


Fig. 3 (a) Bulk (deuterium) ion temperature and (b) ECE intensity as a function of P_{abs} in high, medium and low ε_b configurations.

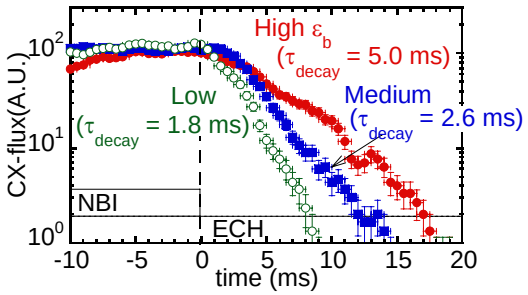


Fig. 4 Time evolution of CX flux (E = 18 kV) after turning-off of NBI. The acceleration voltage of NBI was 28kV and CX-NPA observed the slowing down of beam ions. The base plasma was sustained by 70 GHz ECH.

NBI and ECH plasmas would be interpreted by the increase in the temperature due to the improvement in the energetic ion confinement by bumpiness. As shown in figure 4, the 1/e decay time (τ_{decay}) of the high energy CX flux (E = 18 keV) after the turning-off of NBI increases with increasing ε_b. These results suggest that the control of the bumpiness is effective not only in the energetic ion but also in the global energy confinement in the NBI plasmas of Heliotron J.

2. Spontaneous Shift of Divertor Plasma Footprints during a discharge of Heliotron J [a]

The value of rotational transform has great importance in the core plasma confinement. The effects of rational surfaces in the core region have been studied in many tokamaks and helical devices from the

[a] This work was also supported by Grants-in-Aid for Scientific Research (19560827).

viewpoint of MHD activities. Recently the effects of low-order rational surfaces have been discussed from the viewpoint of the appearance of internal transport barrier or enhanced confinement modes in helical devices. In a low magnetic-shear device, the value of the edge rotational transform $\iota(a)/2\pi$ is essential for the good plasma confinement including L-H transition. In addition, it is closely related to the edge field topology, which is to be used for a 'built-in' divertor in helical systems. In the standard configuration of Heliotron J ($\iota(a)/2\pi \approx 0.56$), for example, the last closed flux surface (LCFS) is surrounded by "ergodic" field lines and some parts of the "whisker" field lines outside LCFS cross the vacuum chamber, forming "divertor traces" on the wall.

Even in non-Ohmic heating plasmas in a helical device, where no net plasma current is necessary to obtain the magnetic surfaces, the plasma stored energy W_p and non-inductive plasma current I_p, which can be driven through plasma heating schemes (e.g. Ohkawa current for NBI heating or electron cyclotron current drive) and/or the plasma pressure gradient (bootstrap current), can affect the rotational transform. Such a modification of the rotational transform can create new rational surfaces in the core region. In the edge region, the change in the rotational transform can modify the divertor plasma distribution in a low-shear device as mentioned above. Therefore, to use the intrinsic edge field topology for a divertor, it is important to experimentally study the dynamics of the divertor plasma distribution caused by the plasma discharge and its controllability. In this section we describe the experimental evidence of the modification of the divertor plasma distribution during a discharge and discuss the mechanism of such modifications.

The position (or spatial profile) of plasma flux outside LCFS was monitored by using two methods at different toroidal and poloidal positions. One is Langmuir probe measurements of the plasma flux coming to the wall along the intrinsic divertor field bundle using poloidal probe-arrays installed near the wall (divertor probe array (DPA) figure 5(a)) at two toroidal sections (ϕ = 67.5° and 112.5° sections), where the field topology is up-down symmetric to each other. The distance between adjacent electrodes in each array is 10mm on the array surface (this distance corresponds to about 7.7mm in the major radius direction). To compensate for this disadvantage of the divertor probe measurement, the second method was additionally introduced in this study; two dimensional measurement of the visible light image near a movable rail-limiter type carbon target (figure 5(b)), which was inserted from a bottom port of the device (ϕ= 157.5° section) and its position adjusted at a proper position for each discharge condition to contact the 'whisker' of the edge field structure. An eight-bit digital video camera system,

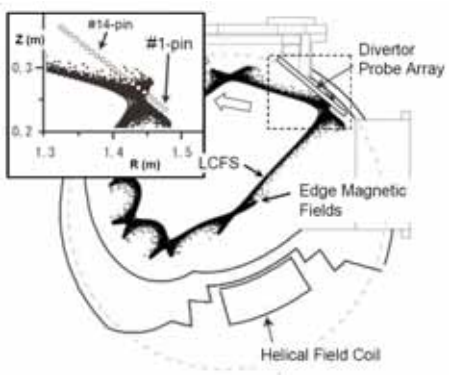


Fig. 5(a) Poincaré plot of the edge field lines at the divertor probe array section (ϕ = 67.5°). The inset shows the probe-pin positions.

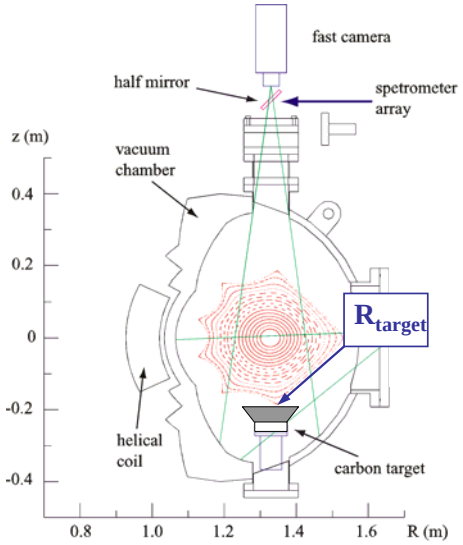


Fig. 5(b) Schematic view of the rail-limiter type carbon target and the fast-camera image monitoring system.

which was installed at the top port facing the target, was used for this measurement. The peak position of the ion-saturation current profile on DPA (R_{DPA}) and the peak brightness position at the carbon target (R_{target}) are estimated to evaluate the divertor plasma footprints.

Figure 6 shows an example of time traces of the line-density of the core plasma ($n_e l$), intensity of Hα -emission (Hα), plasma stored energy (W_p), toroidal plasma current (I_p) and divertor plasma density measured by using one electrode of DPA (ϕ = 112.5°) at R ~ 1.41m for a discharge maintained by the combination of ECH (P_{ECH} ~ 0.3MW) and NBI (P_{NBI} ~ 0.7MW, co-injection) in the standard configuration. An increase in the growth rate of W_p and a sudden drop in Hα at ~270 ms following a slight decrease in Hα intensity from ~255 ms indicate the appearance of an L-H transition. (In this discharge, the target for the video camera measurement was not installed yet.) In this section, we define the direction of the 'positive' toroidal current as the direction in which the plasma

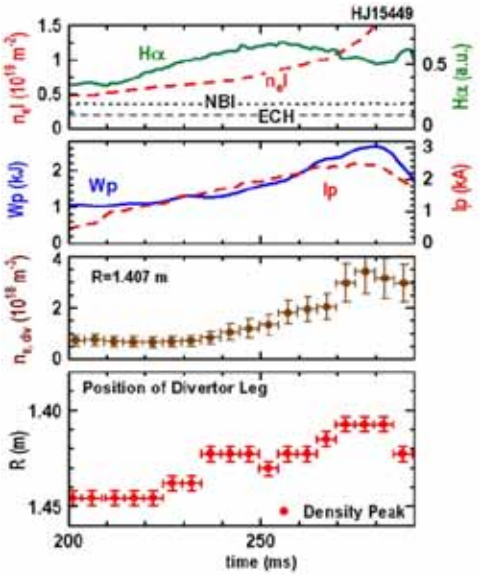


Fig. 6. Time traces of several signals for an ECH and NBI combination heating discharge in the STD configuration (B_0 = 1.25 T).

current increases the poloidal field by the coil system (co-direction). The observed toroidal plasma current gradually increases with increase in the stored energy. Since the plasma was produced during the quasi-steady state of the confinement field and one-turn voltage due to the temporal drift of the external magnetic field strength was negligible, this current is a non-inductive one. In the experimental condition at present, the non-inductive toroidal current can be driven by three different mechanisms; bootstrap current, NB induced current (Ohkawa current) and EC driven current. In the density range shown in figure 6, the observed plasma current is considered to mainly consist of the bootstrap current and the NB induced current. The profiles of the diverted plasma density along the divertor probe array are shown in figure 7 as a function of the major radius at early (211 ms) and later (277 ms) timings of the discharge. The density distribution at 211 ms is almost consistent with that expected from the vacuum field topology if we take into account the ∇B-drift effects during the excursion of ions along the edge field lines to the probe, similar to that observed in low density ECH plasmas. At t = 277 ms, the density-peak-position along DPA, R_{DPA}, clearly shifts inwards by about 4 cm compared with the position at 211 ms. The time trace of the shift of the density-peak-position on DPA is shown at the bottom of figure 6. The overall trend of the density-peak-position shift seems to be well synchronized with the change in W_p or I_p. At the timing when the shift became its maximum, the value of the plasma current was about 2.7 kA.

To estimate the plasma current effects on the field topology, the modification of the vacuum field topology caused by the toroidal current was calculated assuming a filament current on the original vacuum magnetic axis.

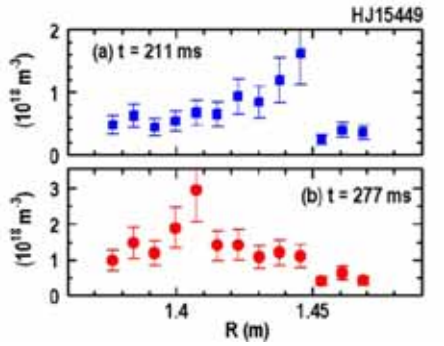


Fig. 7. Density profiles along the divertor probe array at two timings of a single discharge. (The same discharge as in figure 4.)

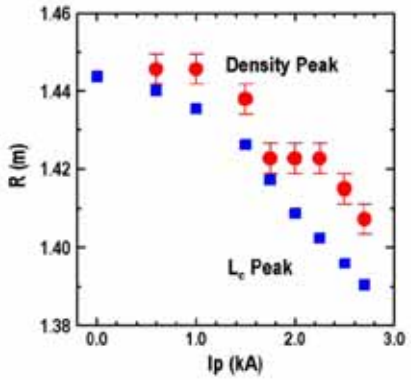


Fig. 8. I_p-dependence of R_{DPA} for ECH + NBI plasmas in the STD configuration. The dataset is the same as that shown in Fig. 6. The position of L_c-peak from the calculation under the filament current assumption for DPA is also plotted as a function of I_p.

Here we ignore the change in the axis position due to this current although the position of the vacuum magnetic axis should also be changed because of the existence of the current. In this calculation, we evaluated the connection length L_c of the field line starting from the probe array after traveling around the confinement region. The position of the L_c-peak on DPA for the discharge shown in fig. 6 is also plotted in fig. 8 as a function of the plasma current. Although there is a systematic discrepancy between the experimentally observed density-peak position and the L_c-peak position, it is observed that there is a similar dependence and almost the same span of shift for both the L_c-peak and R_{DPA}. Except for this discrepancy, the I_p-dependence of R_{DPA} agrees qualitatively with that of the L_c-peak position.

In summary, for discharges with a non-inductive small plasma current (< 3 kA) and the plasma stored energy (< 3 kJ), the observed shift was of the order of a few cm. The observed shift is related to I_p more closely than W_p. The most plausible mechanism for the observed shift is the change in the edge field topology caused by I_p.

Advanced Energy Research Section

Yashige Kotake, Foreign Visiting Professor
(Oklahoma Medical Research Foundation and University of Oklahoma Health Sciences Center, Oklahoma City, OK, USA)

1. Introduction

First of all, I would like to thank Professor Keisuke Makino and the Institute of Advanced Energy, Kyoto University for sponsoring the visiting professorship. Professor Makino and I have been in the same field of science, i.e., chemistry and biology of oxidative stress, for many years. Although it was short three months stay I was inspired by the knowledge and ideas of the members of Professor Makino's laboratory.

Recent progress in biological science indicated that oxygen-derived free radicals which are produced as side products of normal metabolism are injurious to biological molecules, causing protein malfunction and diseases. Our body is equipped with natural defense mechanism against oxygen radical attack, however, people with weakened immune defense becomes vulnerable to the attack. Foods and supplements rich in antioxidants help quenching oxygen radicals, reinforcing the defense. Many evaluation methods have been developed to determine antioxidant's free radical quenching capacity. We found that presently used methods seem to be problematic in many aspects, thus our project concerns the development of a new evaluation method of antioxidant's free radical scavenging capacity.

2. Development of a new Oxygen Radical Absorbance Capacity (ORAC) Assay.

Free radical scavenging capacity of natural substance and its relationship to the heal benefits has attracted considerable attention in food and nutritional science. Recently, fluorescence-based oxygen radical absorbance capacity (ORAC) assay is gaining popularity, and in the USA some foods and drinks list the ORAC values in order to justify their health benefit claims. US department of agriculture (USDA) lists the ORAC database in its website (www.usda.gov) without evaluation. Fluorescence based ORAC assay is to monitor the decay of fluorescence intensity form fluorescent material such as fluorescein when it is exposed to free radical generating systems (e.g. thermal decomposition of azo-compounds). If antioxidant is present in the system, it can protect fluorescent material from free radical damage, thus delays the decay of fluorescence intensity. Such protection ability is converted into numbers using an appropriate standard material. However, this method is an indirect method and is not able to specify the kind of free radical against which antioxidant reacts.

Professor Makino and I have been using electron spin resonance (ESR) spin trapping technique for many years. This method can identify and quantify the free radical at the same time. Our idea was to combine ESR spin trapping technique with ORAC approach. Fluorescence-based ORAC assay (ORAC-FL) uses thermal decomposition of water soluble azo compound AAPH to generate oxygen radicals. But, the free radical production rate in this method is very slow and does not match with ESR spin trapping method.

Our approach was to produce a micromol/liter level of free radicals by illuminating UV light for short period (typically 5 seconds) to the AAPH solution which is located in the ESR spectrometer. Kinetic formulation of the competitive reaction between the spin trap and the antioxidant against AAPH radical (RO) has been published elsewhere. Briefly, in the presence of the spin trap (ST) and the antioxidant (AOx), AAPH radical scavenging reactions that should occur are:

$$RO + ST \longrightarrow \text{RO-adduct} \quad \text{rate constant } k_1 \quad (1)$$

$$RO + AOx \longrightarrow \text{product} \quad \text{rate constant } k_2 \quad (2)$$

When the ESR peak intensity of RO radical adduct in the presence of ST alone and both ST and AOx is I_0 and I, respectively, the amount of product must be $I_0 - I$. Thus,

$$(I_0 - I)/I = k_2[RO][AOx] / k_1[OH][ST] \quad (3)$$

Because almost negligible portions of ST and AOx will be consumed in this reaction, we can safely replace [ST] and [AOx] with $[ST]_0$ and $[AOx]_0$, respectively, where $[\ ]_0$ denotes the initial concentration of the component. Thus,

$$(I_0 - I)/I = k_2/ k_1 \bullet [AOx]_0/[ST]_0 \quad (4)$$

In conclusion, the plot of $(I_0 - I)/I$ against $[AOx]_0/[ST]_0$ will give a zero-crossing line with the slope of k_2/ k_1. When the same spin trap and the free radical-generating condition are used, k_1 becomes common for all antioxidants. As a result, k_2 can be compared each other using k_1 as a reference.

A typical experimental procedure goes as follows: 1) record control ESR signal intensity from the solution containing 1mM AAPH and 10mM CYPMPO (ST) (Fig.1 Top); 2) record ESR signal intensity I in the presence of a

known amount of the antioxidant (AOx) (Fig.1 Bottom); 3) plot $(I_0 - I)/I$ against $[AOx]_0/[ST]_0$; and 4) calculate k_2/k_1 (k_1 : CYPMPO's OH radical trapping rate constant , k_2 : AOx's RO radical scavenging rate constant) for each AOx concentration, yielding k_2/k_1.

The proposed method of ESR-based ORAC has merits as compared to ORAC-FL: This method: 1) readily identifies free radical species to be scavenged by antioxidants; 2) directly monitors the amount of free radical scavenged by the antioxidant; and 3) is quick and the time required for the measurement is considerably shorter (typically 3min per one point) than ORAC-FL (typically 70min for one decay curve). Presently, we are collecting data for various antioxidants. The problem to be resolved is that the ORAC-FL standard Trolox cannot be used because it is light sensitive. Nevertheless, this method appears to be promising. We are planning to publish these results very soon.

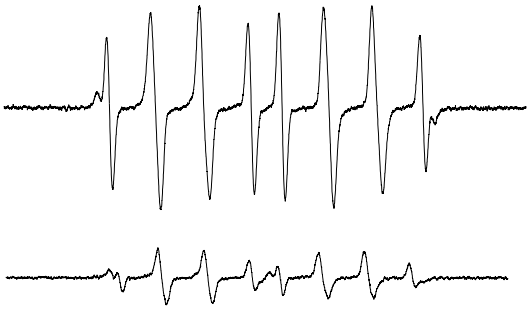


Fig. 1 ESR spectra of spin-trapped AAPH-derived free radical from UV illuminated AAPH (1mM) solution in 50mM phosphate buffer. Top: Blank sample in the absence of antioxidant; Bottom ESR signal in the presence of 5mM N-acetyl cysteine (NAC).

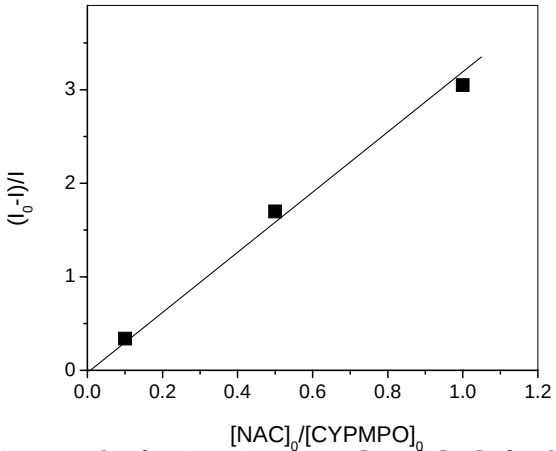


Fig. 2 A plot for $(I_0 - I)/I$ against $[AOx]_0/[ST]_0$ for three different concentrations of AOx. AOx=NAC, ST=CYPMPO. Average slope of this line is 3.2±0.1, which represents that free radical scavenging rate constant of NAC is 3.2 time larger than that of CYPMPO.

3. Spin trapping study of superoxide generation from 6-FP derivatives

Newly developed spin trap CYPMPO was used to detect superoxide generation from the mixture of NADPH and 6-FP derivatives. More water soluble 6-FP derivatives were synthesized by Dr. Nonogawa, but existing spin trap was not able to detect superoxide radical form these compounds. The following paper has resulted form this study: Nonogawa M, Pack SP, Arai T, Endo N, Sommani P, Kodaki T, Kotake Y, Makino K. Synthesis of 6-formylpterin nucleoside analogs and their ROS generation activities in the presence of NADH in the dark. Org Biomol Chem. 2007 21;3314-3319.

Acknowledgements: ESR measurements were performed by Dr. Endo at Wakasawan Energy Research Center, Dr. Oowada at St Marianna Medical University, and Dr. Fujii, at Sapporo Medical University. Mr. Shimmei Radical Research Inc, kindly provided the spin trap CYPMPO.

Advanced Energy Research Section

Mark S. Tillack, Foreign Visiting Professor
(Associate Director, Center for Energy Research, University of California San Diego, USA)

1. Introduction

During the period September through November of 2007, I was hosted by Prof. Satoshi Konishi at the Institute for Advanced Energy. My activities included weekly lectures for the students and staff in Prof. Konishi's group, seminars at Kyoto University and other locations throughout Japan, attendance at conferences, and collaborative research on fusion energy technology.

2. Lectures on fusion power plant design and technology

A series of lectures was presented on magnetic fusion energy technology. The lectures covered four main topics: (1) requirements for an attractive fusion energy source from the perspective of the US electric power industry, (2) features and components of a magnetic fusion energy (MFE) power plant, (3) functions and characteristics of the tritium-breeding blanket, and (4) details of the design and operation of a dual-coolant (He and PbLi) liquid metal blanket.

3. Seminars

Seminars were presented at Kyoto University (Uji campus), Osaka University (Institute for Laser Engineering), and Tokyo University (Kashiwa campus). The seminars spanned a range of topics, including:

a. An overview of inertial fusion energy (IFE) chamber and target technology. Researchers at UC San Diego study several of the key issues associated with inertial fusion energy technology. Research related to IFE targets includes scalable fabrication technologies, electrostatic steering, and target injection, tracking and engagement. Research related to IFE chambers includes chamber design studies, armor thermo-mechanical response under cyclic loading, magnetic intervention of high-energy ions, chamber gas dynamics and clearing, and processes involved in surface ablation, ejecta formation and vapor condensation. Research on final optics includes mirror fabrication, high-cycle testing and system integration.

b. Grazing incidence metal mirrors for direct-drive IFE. UC San Diego leads the development and testing of final optics for the US High Average Power Laser (HAPL) program. Solid solution alloys of aluminum have been shown to meet the requirements on reflectivity at 248 nm and offer damage resistance up to 5 J/cm^2 over long- term exposures.

c. Laser-produced plasma light source research for extreme ultraviolet (EUV) lithography. Researchers at UCSD collaborate with companies in both the US and Japan to develop the light source for next-generation semiconductor lithography. The leading candidate at present is a laser-produced soft x-ray source based on Sn emissions. Our research attempts to optimize in-band light emission around 13.5 nm while avoiding the damaging effects of high kinetic energy ions on nearby EUV optics.

In addition to seminars, a presentation was given at the 5th International Conference on Inertial Fusion Science and Applications in Kobe, on the subject "Geometric effects on EUV emissions in spherical and planar targets". In this work, we reported differences in radiation hydrodynamic behavior of laser-irradiated Sn targets, supported by both experimental and numerical simulation studies.

4. Collaborative research on fusion energy technology

The Institute for Advanced Energy at Kyoto University and the Center for Energy Research at UC San Diego share a common interest in fusion power plant design studies, socio-economic studies and fusion nuclear technology R&D.

UC San Diego leads a US government-sponsored multi-institutional activity on fusion power plant studies called the ARIES (Advanced Reactor Innovation and Evaluation Studies) program. The ARIES Team developed an innovative "dual coolant" blanket concept as a part of the ARIES-ST spherical torus power plant study in 1996 (M. S. Tillack and S. Malang, "High-Performance PbLi Blanket," 17th IEEE/NPSS Symposium on Fusion Engineering, San Diego, CA, Oct. 1997.) This design concept has evolved to become one of the leading power plant blanket candidates under investigation in R&D programs worldwide. Further refinements and evolution of the concept continue to be made, and R&D programs are underway to develop components and to validate the key blanket features.

One of the critical issues associated with the use of PbLi and SiC relates to materials compatibility at high temperature. For an electricity-generating blanket, high temperature operation permits high thermal conversion efficiency, which not only improves the economic attractiveness of fusion, but also helps to ease constraints on other parts of the system (such as plasma confinement and power density). If sufficiently high temperatures can be achieved in the dual coolant blanket, then hydrogen production becomes a viable option; this requires temperatures in excess of 800˚C.

Experimental research on materials compatibility requires high-temperature flow loops using prototypical materials. Due to the exceptionally high temperature desired, combined with the relatively narrow database on these materials, several key enabling technologies require further development in order to build and operate a corrosion loop. These include heat sources, heat exchangers, high-temperature liquid metal pumps and diagnostics.

A survey of existing corrosion loops was performed and component options and R&D needs were evaluated. Analysis was performed to determine the parameters and requirements on corrosion loops at different scales, including low-flow natural convection, small-scale and large-scale forced convection loops. The technology challenges were characterized as a function of the peak temperature and temperature gradient in the loop.

In addition to fusion blanket design and R&D, the pathway from ITER to a power plant is a subject of common interest between Kyoto University and UC San Diego. Although a general consensus exists on the issues and R&D needs for fusion energy, strategies for the commercialization of fusion differ significantly between the different parties to ITER. In the US, a program-wide review is underway to assess the research gaps and facility needs from the present until fusion energy can be transferred to the private sector.

In order to evaluate our current state of readiness and remaining R&D needs, we are developing a methodology using "Technology Readiness Levels" (TRL's). This approach has become increasingly popular in large US government sponsored R&D programs in NASA, DOE and DOD. TRL's provide an objective, integrated, self-consistent and design-independent procedure that can be understood and utilized by the full range of stakeholders in fusion, including governments, R&D providers and private sector developers and end-users.

A detailed set of readiness levels that encompass the major technology challenges for fusion energy development have been defined and applied to evaluate our current level of advancement.

5. Plans for further collaborative research

Due to the strong overlap in interest on fusion nuclear technology between Kyoto University and UC San Diego, we expect continuing collaborations on both power plant studies and fusion nuclear technology R&D. These include:

- Further refinements and improvements to the dual coolant blanket, with an emphasis on higher outlet temperature.
- Technology R&D planning and pathway development.
- Fusion nuclear technology R&D, with an emphasis on high temperature operation and materials compatibility.

The level of collaboration on FNT R&D will depend on funding and program plans within the DOE-sponsored US fusion program.

Advanced Energy Research Section

Detecting Nanoscale Dissolution Events by Microelectrochemical Techniques

S. Virtanen, Foreign Visiting Professor

(Department of Materials Science, University of Erlangen-Nuremberg, Erlangen, Germany)

1. Introduction

Electrochemical dissolution (corrosion) of technical metals and alloys seldom proceeds in a uniform manner over the surface; instead many different types of localized corrosion can be observed (e.g., pitting corrosion, intergranular corrosion). The origin of such localized corrosion processes is typically related to a heterogeneous surface, for instance to the presence of inclusions or precipitates on the surface, or to a multiphase alloy system. For homogeneous single-phase alloys, grain-boundaries also can act as preferential dissolution sites. As the localized corrosion phenomena often are difficult to predict, and show a non-linear time-dependence of propagation, they can lead to catastrophic non-expected materials failure. Generally, localized corrosion phenomena determine the life-time of many otherwise highly corrosion resistant materials in important practical applications (high-corrosion resistance of these alloys is provided by “passivity”, meaning self-protection of the metal by thin metal oxide layers on the surface). In the visiting scientist’s home laboratory, these passivity and localized corrosion are studied for materials used for instance in biomedical, aerospace, automobile, or energy technology applications. The materials studied include stainless steels, Al alloys, Mg alloys, Ti alloys (and others). Moreover, novel material systems such as glassy metals or nanocrystalline materials are also of interest.

Due to the high practical significance of localized corrosion phenomena, the critical factors of many of these processes are well recognized today. However, mechanistic understanding of the nucleation and initiation of localized dissolution is still missing. Partially this is of course due to the fact that a study of the earliest stages of localized dissolution (nm-scale dissolution events) is a very challenging task. This short summary presents some recent findings on how this challenge can be tackled.

2. Electrochemical study of nm-scale dissolution events

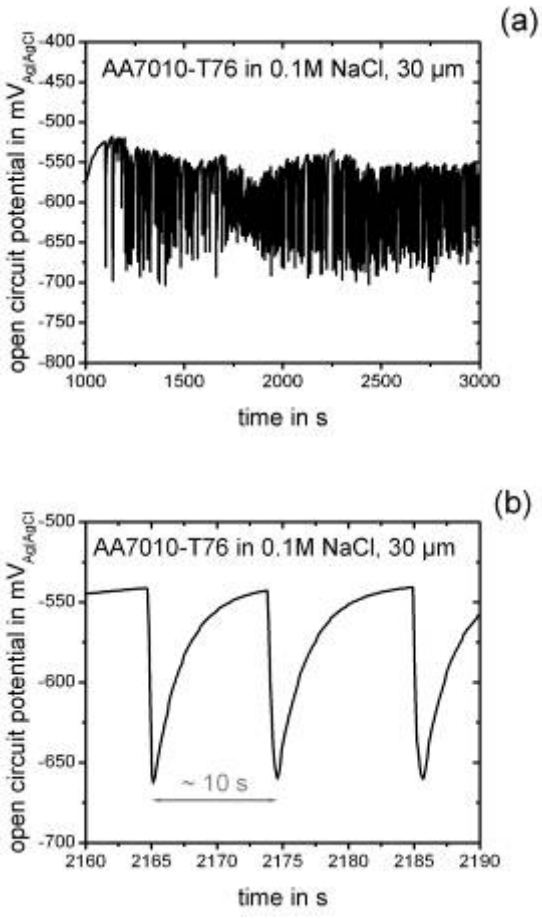
Nucleation and initiation events of localized breakdown of passivity typically can be observed as electrochemical noise or in shape of transients of potential or current. Current transients in the passive region are typically interpreted as localized breakdown and repassivation events. On a passive metal surface, a huge number of such small nucleation and initiation events take place. Therefore, in conventional electrochemical experiments, many of the smallest dissolution events are hidden in the background current and cannot be resolved as single events. To be able to detect nm-scale dissolution events electrochemically, one must be able to carry out measurements with a high current resolution (high signal-to-noise ratio). This can be accomplished by using microelectrodes. Different type of experimental set-ups have been used, including embedded microwires or using a microcapillary as an electrochemical cell. In our laboratory, we carry out experiments with the glass capillary cell [see e.g., 1], as this enables to study all materials and not only those available in the shape of microwire (in addition, the microstructure of the material in the shape of wire is different from the microstructure of for instance plate material).

Figure 1 shows transients in the open-circuit potential of an aluminium alloy in a 0.1 M NaCl solution measured with a glass capillary diameter of 30 μm [2]. After a short incubation time ocp transients can be observed (1a). The frequency of these transients increases slightly with the contact time but remains mainly in the order of 0.1 Hz. As it can be seen from the magnification shown in Fig. 1b, the drop in ocp is very rapid within fractions of seconds, whereas the recovery can not be completed because the next transient occurs. Nevertheless, a rather stable upper value of –550 mV and a lower value of –670 mV can be observed. By carrying out the ocp measurements at different sites of the surface it was found that these ocp fluctuations only take place when the surface investigated contains the Al alloy matrix as well as noble intermetallic particles. The origin of the potential transients was therefore suggested to be related to local galvanic coupling of different phases present on the alloy surface. The ocp fluctuations can then be explained by considering the different anodic and cathodic reactions, which are balanced at the open-circuit potential. More details can be found in Ref. [2]. By using different capillary sizes it can be demonstrated that the transients as single events are only detectable for small contact diameters, whereas for larger diameters the high number of single transients lead to a noisy ocp meas-

urement. Interestingly, the occurrence of ocp transients in such systems was predicted already in 1971 as it was claimed that in the ocp "regular potential fluctuations occur which, owing to the small dimensions of the area considered, are practically impossible to determine" [3]. With the microcapillary technique it is now possible to decrease the surface area to be studied. In the present case, diminishing the area to <1000 μm^2, is already sufficient to resolve the "regular potential fluctuations" as ocp transients.

Fig. 1 (a) Open-circuit potential of an aluminium alloy AA7010-T76 in 0.1 M NaCl measured with the microcapillary cell, capillary diameter 30 µm. (b) Magnification of the ocp-behavior.

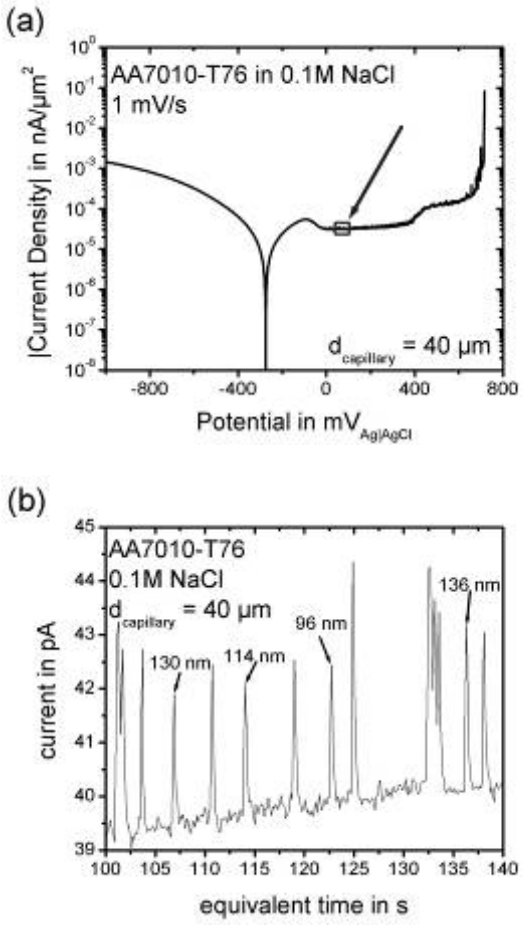
For the same alloys, while recording the current under



potential control, current transients can be observed in the passive region (Fig. 2) [2]. Assuming that the transients are single events associated with the dissolution of small anodic intermetallic particles present in the alloy microstructure it is possible to estimate the particle size from the current during the transient. By integrating the transient current and applying a simple model, one can assign a particle size to each transient (shown in Fig. 2b). The hence determined size of the dissolution events corresponds very well to grain boundary precipitates present in the alloy, as observed by transmission electron microscope (TEM). Further work is required, however, to directly correlate the current transients to specific dissolution events. Nevertheless, the results demonstrate that decreasing the surface area probed it is indeed possible to electrochemically detect single nanoscale dissolution events.

Fig. 2 (a) Potentiodynamic polarization curve of an aluminium alloy AA7010-T76 in 0.1 M NaCl; (b) Magnification of the passive current density showing the occurrence of small current transients.

In addition to the results shown above, we are interested



in using the microcapillary technique to study the local electrochemical behavior of specific surface sites of heterogeneous substrates, such as different intermetallic particles, the particle/matrix-interface, pure matrix, or grain boundaries. These studies can help to identify "weak sites" of the surface, or enable to understand formation of internal galvanic couples on alloy surfaces. Examples of such investigations can be found in Refs. [4-6].

Moreover, the locally resolved electrochemical techniques can be complemented by carrying out high-resolution surface analytical examination of the

materials. In this manner, it may be possible to determine, if detected changes in the local electrochemical reactivity are due to variations in the surface chemical composition, surface strains, or surface structure. Examples of high-resolution surface analytical studies of heterogeneous substrates are given in Refs. [7,8].

3. Metals corrosion and electrochemical patterning of Si surfaces?

During the research stay, in discussions and in seminars, it was highly interesting to compare the electrochemical dissolution behavior of homogeneous and patterned Si surfaces with the knowledge on metallic corrosion. Porosification of semiconductors in many aspects resembles localized corrosion, albeit the fact that the semiconductor substrate is a far more ideal system than many of the materials studied by corrosion scientists. However, the general electrochemical concepts of course hold both for semiconductors and for metals. Many technically important alloys are self-protected from corrosion by thin oxide films on their surface (called passive films). These oxide films typically show a semiconducting nature, but are very non-ideal semiconductors as they are quite highly doped. However, understanding of semiconductor electrochemistry is required, to fully understand the corrosion behavior of passive metals and alloys.

The visiting researcher had also the opportunity to other universities and research institutes in Japan including the University of Osaka, University of Hokkaido, Kogakuin University in Tokyo, and the National Institute of Materials Science in Tsukuba. During these visits, existing collaborations were strengthened and new contacts were established.

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Advanced Energy Materials Research Section

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1. Introduction

The importance of the materials development for advanced energy systems including nuclear fusion and fission reactors has been rapidly growing in these years and expected to be emphasized in the coming years and the upcoming century. The mission of the Advanced Energy Materials Research Section at the Advanced Energy Conversion Division is to develop advanced energy materials to be used in advanced energy systems with the emphasis on advanced energy conversion systems. The research section is unfolding unique and extensive researches in the fields of functional and structural materials development as well as playing important roles in national and international programs for R & D of energy materials.

The current emphasis of the research section is on materials studies for nuclear fusion and nuclear fission reactors and for aerospace applications where resistance of materials under severe environments, such as high temperature, high heat flux, high energy particle irradiation, complex and variable stresses and others, are required. The ongoing research themes include (1) research and development of steels including both advanced reduced activation ferritic steels and conventional austenitic steels for fusion and fission applications, (2) research and development of silicon carbide fiber-reinforced silicon carbide matrix composites (SiC/SiC composites) for fusion and other advanced energy systems, (3) theoretical modeling and computational studies on the material behavior under high energy particle irradiation conditions, (4) development of sub-sized or miniature specimen techniques for mechanical property evaluation of structural and functional materials, (5) joining of the materials for extremely severe environment, (6) fundamental studies on the physical processes responsible for the creep and fatigue phenomena in metallic materials, and (7) experimental and theoretical studies on the heat transfer through liquid coolants in the high-efficiency energy conversion systems.

The research section is functioning as one of the central organizing bodies of fusion materials research in universities. The current JAPAN/USA Collaboration program, “Dynamic behavior of fusion materials and their response to varying and complex irradiation conditions”, so called JUPITER program, is one of the most important activities and the program has been strongly related with scientific, engineering and technological studies on interactions of materials with energetic particle bombardments, such as neutron irradiation and charged particle irradiations. JUPITER program finishes in March, 2007. The new collaboration program, so called TITAN program, is going to start in 2007.

2. On-going and up-coming projects

Under the title of “Fundamental R&D on Advanced Material System for (High Efficiency Environment-conscious) Very High Temperature Gas-cooled Fast Reactor Core Structures,” a new R & D activity to establish core structure for gas-cooled fast reactor (GFR) using SiC/SiC composites has been established for the research period in December, 2002 through March, 2006, as one of the programs of Development of Innovative Nuclear Energy System Technique supported by Ministry of Education, Culture, Sports, Science and Technology (MEXT).

The novel processing called Nano-powder Infiltration and Transient Eutectoidic (NITE) Processing has been developed based on the liquid phase sintering (LPS) process modification in the program. The NITE processing can achieve both the excellent material quality and the low processing cost. The productivity of the processing is also excellent, and various kinds of shape and size of SiC/SiC composites can be produced by the NITE processing.

Based on the fruitage, the other major two projects related to GFR started in December, 2005, as one of the programs of Development of Innovative Nuclear Energy System Technique supported by MEXT. One is “Development of High Burn-up Composite Fuel for GFR,” through March, 2008. Basic fabrication technique of fuel for GFR will be established using high dense SiC and porous SiC.

Another project is just up-coming. New program having a title of “Industrial User Support Program for Research of Invention and Maintenance of Materials for Energy Applications” started in April, 2007. This project promotes sharing our excellent equipments and research accumulation of energy materials with industry for the benefit of public. The project named “ADMIRE”, which is short for “Application of DuET and MUSTER

for Industrial Research Engineering", is going to continue for 5 years since 2007.

3. Development of advanced SiC/SiC composites for nuclear energy systems

Advanced nuclear energy systems, such as gas cooled fast reactor (GFR), very high temperature reactor (VHTR) and fusion reactor are potential candidates for sustainable energy systems in the future. In order to realize these attractive energy systems, structural materials must be responsible to keep their performance under very severe environment including high-temperature, high energy neutron bombardment and surrounding coolants and fuels. Today a major thrust is by the development of fiber-reinforced ceramic matrix composites (CMCs) in general and silicon carbide fiber-reinforced silicon carbide matrix (SiC/SiC) composites in particular. Because of fiber-reinforcement, SiC/SiC composites are more damage tolerant to mechanical and thermal loading (thermal shock) and have the capability for larger components than their SiC monolithic form. Also in comparison to the best high-temperature metallic alloys, SiC/SiC composites are lower density and thermal expansion, and have the potential for displaying excellent high-temperature thermo-mechanical properties under high energy neutron bombardment.

Nano-Infiltration and Transient Eutectic-phase (NITE) process is the first successful application of liquid phase sintering (LPS) for matrix densification of SiC/SiC composites. The matrix in NITE-SiC/SiC consists of well-crystallized SiC grains with small remnants of the metal oxide sintering additives. Such polycrystalline SiC matrix suggests excellent radiation resistance of the NITE SiC/SiC composites similar to that of chemically vaporized SiC/SiC composites (CVI-SiC/SiC). The differences of properties between NITE and CVI composites are mainly caused by their matrix porosities, the NITE composites are less porosity than the CVI composites. A heat flux capacity of NITE-SiC/SiC is superior in the candidates of first wall materials for fusion reactor, so that it is expected that NITE composite has an excellent figure of merit against the thermal stresses. Satisfactory results for reducing the leakage of helium gas as a coolant gas in the reactor are also reported. The development of SiC fiber (Tyranno^{TM}-SA) has remarkably progressed. Various processes should be conducted to receive a well-crystallized fiber which was better than SiC fibers in existence.

4. Development of evaluation methods of inter-laminar shear/tensile strengths for Advanced SiC/SiC Composites at elevated temperatures

For the application and design of SiC/SiC composites, various testing methods for different fracture mode have been required. Our research group has made efforts to develop various evaluation methods such as monotonic tensile and inter-laminar shear/tensile strengths. Double-notched specimen (DNS) test and diametral compression tests were conducted in order to clarify the inter-laminar shear/tensile properties of NITE SiC/SiC composites at high temperatures, which have highly crystalline fibers and high dense matrix.

Double-notched specimen (DNS) and diametral compression tests were conducted at 298-1573K, in air, Ar and Ar+O2 atmospheres. Fracture surfaces and crack propagation paths were observed by optical microscopy and scanning electron microscopy.

The credibility of diametral compression test was confirmed at ambient temperature by comparing trans-thickness tensile test normalized in ASTM C1468. Inter-laminar shear/tensile strengths increased at 1573K. This reason may be due to the relaxation of residual stress by mismatch of thermal expansion coefficient. At an elevated temperature in Ar+O2, PyC interface was deteriorated and inter-laminar shear/tensile strengths decreased.

5. Development of porous SiC for nuclear energy systems

Recently, there has been an increasing interest in the applications of porous ceramics as hot-gas or molten-metal filters, catalyst supports, battery electrodes, heat insulators, ion exchangers, gas sensors, and water cleaners. In particular, porous SiC ceramics are considered as functional materials of advanced energy systems, such as perforated containment wall or flow channel inserts (FCIs) for blanket module of fusion reactor, and inner/outer tube of a coated particle type fuel compartment for horizontal flow cooling concept with directly cooling system on Gas-Cooled Fast Reactor, because of their low thermal-expansion coefficient and thermal conductivity, good thermal-shock resistance as well as excellent physical and chemical stability at elevated temperature. A number of manufacturing approaches have been applied to fabricate porous SiC including polymer pyrolysis, oxidation bonding, and reaction bonding. However, their processes are complicated and conventional porous SiC shows insufficient physco-chemical stability under high temperature environment. Therefore, from the view point of safety and stability, it is necessary to develop an uncomplicated manufacturing method and to investigate mechanical and thermal properties of porous SiC ceramics.

In our group, porous SiC ceramics have been manufactured based on the Nano Infiltration Transient Eutectic (NITE) process, which is developed as a processing technique for high performance a SiC fiber reinforced SiC matrix composite. The NITE-porous SiC ceramics exhibited a substantially high strength in

comparison with other conventional porous SiC ceramics, due to its robust microstructure consisted of spherical pores. In addition, porosity of the NITE-porous SiC ceramics could be controlled with a high degree of accuracy. Also, the NITE-porous SiC ceramics had low thermal conductivity from room temperature to elevated temperature. Therefore, from the excellent physical and chemical stability at elevated temperature, NITE-porous SiC ceramics could be expected as high performance multi-functional materials for advanced energy systems

6. Engineering research on joining of materials for nuclear energy systems

The important issues to use SiC/SiC composites for industry are the developments of joining and coating techniques. Several kinds of joining techniques have been developed to join SiC and SiC/SiC composites using polymer, glass-ceramics and reaction bonding. One of the key for the development is the stability of the joining at application temperature. For SiC or SiC/SiC composites joining, our new joint technique using SiC has an advantage at the high temperature due to the very limited CTE mismatch. Monolithic SiC and NITE-SiC/SiC composites are successfully jointed applying NITE process. These joined SiC materials by the applied NITE process show stronger strength than the SiC materials joined by other conventional jointing processes.

Additionally, a two step joining method had been developed to stack complex components. The SiC ceramics were successfully joined by two step joining method for multi joined component. The protruded powder or uncompleted joint layer was not observed because the coated joint layer was solid-state after first step. The joining strength of joined material by two step joining method was higher than that of direct joining method.

7. Joining and coating technologies for SiC with dissimilar materials

Joining and coating technologies are necessary for the construction of fusion reactor by SiC/SiC composites. Fundamental study of interface of dissimilar joints or metal coated SiC materials are ongoing in parallel with the engineering developments. Tungsten is an appropriate material for the dissimilar joint and coating with SiC because of the similar thermal expansion coefficient with SiC. Recent developed newly SiC/SiC composite, NITE SiC/SiC composite has excellent resistance against high temperature and high pressure, thus, the hot-press joining is being developed. The hot-pressing is performed over 1000 °C in Ar gas flow environment. A tungsten plate or tungsten powder are put on a SiC plate and consolidated by hot-pressing. The consolidated specimens are investigated by SEM and mechanical testing. SEM investigation show that the reaction zone between tungsten and SiC grew up with the hot-pressing temperature and time. The reaction zone is investigated W-Si, this zone seems to determine the share strength of joined materials. More precise investigation and research of interface are progressing.

The techniques to bond SiC and SiC/SiC composite to general purpose construction materials, stainless steel, are also developing for the expansion of practical applications. Two step joining method is being developed for SiC and steel. The first step is joining of SiC to W or W alloy by diffusion bonding based on the joining technique previously developed for SiC and W, followed by joining of SiC/W to stainless steel (SUS430) with intermediate materials which inserted for reduce the residual stress in the joints. The preliminary results indicate that the joining of SiC to steel is possible by the proposed procedure. More detail evaluation and optimization are under investigation.

8. Modeling of microstructural evolution in β-SiC under irradiation

Lattice defects produced and accumulated in a material under irradiation cause the microstructural changes and affect the material properties. In order to analyze the atomistic behavior and understand the kinetics of defects, some energetic parameters such as defect formation energies and migration energies are necessary. However, even basic properties of SiC materials have been unknown.

In our laboratory, formation energies and stable configurations of self-interstitial atom (SIA) clusters in β-SiC are calculated using a classical molecular dynamics (MD) method using Gao-Weber potential based on the Brenner potential formalism. The properties of SIA clusters with various size and composition of silicon (Si) and carbon (C) interstitials are studied in the MD simulations of 1000 unit cells. Temperature condition is started at elevated temperature and reduced to 0 K during the calculation to obtain the total energy.

MD calculations show that the most stable isolated silicon interstitial is the substitution type in the tetrahedral position surrounded by four regular carbon lattice sites. On the other hands, the most stable isolated carbon interstitial is the dumbbell type with a <100> direction on a regular carbon lattice site. These configurations of each interstitial atom are still stable even when interstitial atoms form a cluster. The size dependence of formation energy of SIA-clusters as a function of cluster size n is an increasing function. The cluster size n shows the total number of SIAs in a cluster. The function is roughly in proportion to n=1 and n=1/2. The formation energy of SIA-clusters consisted of both silicon and carbon interstitials is higher than that

of SIA-clusters of only silicon or carbon interstitials. For example, the formation energy E (Si2C2) is 7.83 eV in contrast to E (Si4C0) = 6.98 eV and E (Si0C4) = 7.69 eV, where SixCy is a SIA-cluster composed of x Si interstitials and y C interstitials.

From these formation energies of SIA-clusters, binding energy of an SIA to SIA-clusters can be obtained. Defect energies such as the binding energies are very important to investigate formation kinetics of defect clusters. In the future, the mechanism of formation of SIA-clusters in β-SiC under irradiation will be investigated using the kinetic rate theory based model employed defect energies evaluated in the present work.

9. Neutron irradiation effects for mechanical properties of monolithic SiC

Flexural properties of CVD- and NITE-SiC were characterized statistically after neutron irradiation. Bar samples were irradiated in target position capsules in the Japan Materials Testing Reactor (JMTR, Oarai, Ibaraki) to nominal fluence levels up to 6.4×10^{24}n/m^2 (E>0.1MeV) at temperatures of 400, 600, and 750^{o}C.

In all conditions, Young's modulus and Weibull modulus reduced after irradiation. Irradiation also caused enhancement of flexural strength at all irradiation temperatures for CVD-SiC. On the other hand, NITE-SiC exhibited no significant degradation in flexural strength. Weibull modulus of NITE-SiC was influenced by the data fractured at low strength. Therefore, identification of fracture mechanism of these materials by SEM observation is strongly required.

10. Irradiation effects on tensile and interfacial properties of advanced SiC/SiC composites

In order to identify the effects of neutron irradiation on tensile and interfacial properties of advanced SiC/SiC composites, cyclic tensile tests were conducted and the hysteresis loop analysis method was applied for the investigation. Neutron irradiation was performed at JOYO (Oarai, Ibaraki). Nominal fluence and irradiation temperature were 3.1×10^{25}n/m^2 at 740^{o}C and 1.2×10^{26}n/m^2 at 750^{o}C. The ultimate tensile strength and proportional limit stress of composites in both conditions showd excellent irradiation resistance. The hysteresis loop analysis indicated that the sliding stress at fiber/matrix interfaces was not changed after irradiation to 3.1×10^{25}n/m^2 at 740^{o}C, whereas it was significantly reduced by 1.2×10^{26} n/m^2 at 750 ^{o}C for both composites. These behaviors might be correlated with the swelling behavior of pyrolytic carbon coating of SiC fibers.

Additionally, irradiation effects for tensile properties of unidirectional and cross-ply NITE-SiC/SiC composites were also studied. Neutron irradiation was conducted at JMTR. Irradiation temperature and neutron fluence were 600 ^{o}C and 5.2×10^{24}n/m^2, respectively. NITE-SiC/SiC composites showed excellent mechanical properties after the neutron irradiation because of the closed swelling of SiC fiber and SiC matrix, and sufficiently high strength of NITE-matrix and fiber SiC.

11. Development of Nondestructive test method for NITE-SiC and SiC/SiC composite materials

The practical use of SiC ceramic materials has been limited, because of the lack of nondestructive investigation method under pre- and in- service environments. It is required to develop a higher reliability nondestructive test method for SiC and SiC/SiC composite materials.

In our group, the defects detection capability of ultrasonic test methods (C-Scan method and Pulse-echo reflection method) on NITE-SiC ceramics has been investigated. Monolithic NITE-SiC specimens with various sizes of artificial defects (cylindrical pores) were prepared and examined by ultrasonic C-Scan method and pulse echo reflection method with the frequency condition of 10, 25 and 50 MHz. Also, the detection limit of 25 and 50MHz was investigated about 100 and 200 um, respectively. It was determined from the wavelengths.

12. The influence of inclusions on the low cycle fatigue properties of reduced activation ferritic/martensitic steels

Reduced activation ferritic/martensitic steels (RAFs), such as F82H, are promising structure materials for ITER test blanket modules (TBMs) and a fusion DEMO reactor. The fatigue properties of RAFs depend on various material factors such as the distributed inclusion, surface morphology and so on. Therefore, the investigation of type of inclusion and these effects on low cycle fatigue (LCF) properties would be essential. The LCF lifetime was increased with decreasing the number density of total inclusions. Two types of inclusions, the complex inclusion consisted of Al_2O_3 and TaO_x, and the simple inclusion of TaO_x, were observed by SEM. It was found that a crack initiation was caused by the separation of the relative weak interface between the matrix and an inclusion. The crack initiation from the Ta-oxide site in a Al_2O_3-TaOx complex inclusion is much extensive than that in the simple inclusions. It can be surmised from these results that the primary reason for the reduction of the LCF property of F82H-IEA heat steel is the existence of complex inclusion with Al_2O_3-TaOx complex inclusions.

Advanced Laser Science Research Section

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1. Introduction

Our research interests are focused on the development and applications of advanced lasers, aiming at the goal of demonstrating the potential ability of coherent radiation to create new bases of future science and technology. The laser development is concerned with the generation of high-intensity ultrashort laser pulses, compressing the laser pulses into a few-cycle or shorter-pulse region, and extending the ultrashort pulse radiation into the extreme ultraviolet region of spectrum. Applications of the advanced laser technology are associated with the study of ultrafast, strong-field interactions with matter to develop new technology of materials control and processing on atomic and molecular levels in the strong field regime, where special attention is focused on nonlinear optical processes such as tunneling ionization of atoms and molecules, Coulomb explosion of molecules, molecular alignment, and nano-structuring on solid surfaces.

2. High-intensity ultrashort pulse lasers

A high-intensity 40-fs Ti:sapphire laser system is working as one of the principal experimental apparatus in our research section. The laser system using the chirped-pulse amplification (CPA) technique consists of a mode-locked Ti:sapphire laser oscillator pumped by the second harmonic output of an all solid-state Nd:YVO_4 laser, an all-reflective pulse stretcher using a pair of grating, a regenerative Ti:sapphire laser amplifier, the second and third power amplifiers, and a grating pulse compressor. The laser system produces a peak power of 1 TW (40 mJ in 40 fs) at a repetition frequency of 10 Hz, and the center wavelength is 800 nm. This system includes a frequency conversion apparatus to extend useful high-intensity fs pulses to the blue and ultraviolet (UV) regions of spectrum. The frequency conversion apparatus employs a simple optical configuration to achieve efficient conversion in a nonlinear optical crystal. The peak power of blue fs pulses at 400 nm is 0.2 TW with the pulse width of 60 fs, and the UV power at 267 nm is 20 GW in 140 fs.

The second Ti:sapphire laser system using the CPA technique was developed for the purposes of advanced material processing with fs laser pulses. This laser system produces 100-fs, 800-nm pulses at 10 Hz with a well-defined intensity distribution and good temporal characteristics. The output pulse duration is designed to be relatively long for the target experiment in which various kinds of optical component are used. The system configuration is almost the same as the 40-fs laser system, consisting of a fs laser oscillator, a pulse stretcher, a regenerative Ti:sapphire laser amplifier, a power amplifier and a pulse compressor. This laser has been used for the study of nano-scale ablation of hard thin films.

A new laser system is under development, of which configuration is shown in Fig.1. The system designed to produce ~ 10 fs pulses at a high intensity uses a Ti:sapphire laser oscillator producing 8 fs pulses and the CPA technique. This laser system will be applied to the study of ultrafast interactions of a few-cycle optical field with atoms, molecules and solid surfaces.

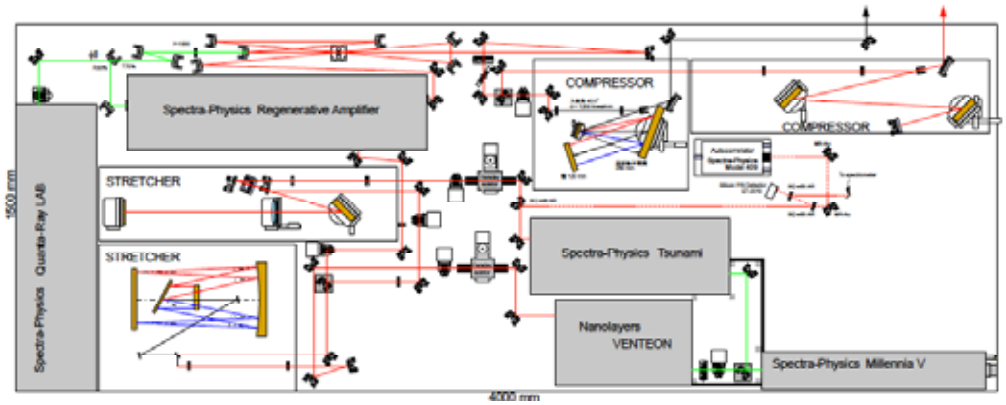


Fig.1. 10-fs Ti:sapphire CPA laser system under development.

3. Molecular alignment with fs laser pulses

Strong-field interaction with atoms and molecules is the subject under investigation for applications of high-intensity fs lasers to the development of new materials-control technology. The fs laser-induced molecular alignment is one of promising approaches to control of nonlinear processes in molecular gases. Recently we have demonstrated that high-order harmonic generation (HHG) provides a sensitive way to probe the dynamic alignment of molecules, and the characteristic harmonic signal in time and frequency domains clearly reveals coherence in the rotational wave packet.

In this study we are collaborating with Prof. F.H.M.Faisal's group of Bielefeld University and have successfully developed a new theory to describe the HHG from aligned molecules such as N_2, O_2 and CO_2. We consider a pump-probe experiment using nonresonant, linearly polarized fs laser pulses for the linear molecules, where the pump pulse forms a wave-packet, and the delayed probe pulse generates high harmonic radiation from the wave packet. We define the orientation of a molecular axis by a polar angle θ and an azimuth angle ϕ. In this scheme, the pump field $\boldsymbol{E}_1$ is fixed along the z direction, and the probe field $\boldsymbol{E}_2$ makes an angle α with respect to the z axis and the relative angle θ' to the molecular axis.

Using the wave function $|\Phi_{J_0M_0}(t)\rangle$ of a rotational wave packet evolving from the initial state $|J_0M_0\rangle$, we solve the Schrödinger equation for the nuclear motion, where the expectation value of the dipole moment is evaluated using the highest occupied molecular orbital (HOMO) and the Volkov wave functions. Then the n-th harmonic signal from aligned molecules is given in a form

$$S^{(n)}(t;\alpha)=\sum_{J_0M_0}\rho(J_0)\left|\langle\Phi_{J_0M_0}(t)|T^{(n)}(\theta,\phi,\alpha)|\Phi_{J_0M_0}(t)\rangle\right|^2,$$

where $T^{(n)}(\theta, \phi, \alpha)$ is an operator to describe the HHG, and $\rho(J_0)$ is the Boltzmann average over the rotational temperature $T_{\rm rot}$.

In the experiment, the linearly polarized output from the Ti:sapphire laser was split into two beams to produce a variable time delay t between the pump and probe pulses. The two beams were recombined collinearly and focused with a 50-cm focal-length lens into a pulsed molecular beam jetted from a 1-mm-diameter nozzle. The harmonic radiation was detected by an electron multiplier mounted on a VUV monochromator, and the signal processed by a boxcar averager was stored on a personal computer.

In what follows, we briefly present the experimental results of the polarization angle dependence of HHG, where the pump polarization was rotated by an angle α to the probe pulse polarization of which direction was fixed to the direction along the monochromator slit.

Figure 2 shows the time-dependent 19th harmonic signals (black lines) observed for N_2 as functions of t for α = 0˚, 45˚and 90˚, together with the 19th harmonic signals (gray lines) calculated by the developed theory under the same condition as the experiment. In the calculation, the rotational temperature of $T_{\rm rot}$ = 100 K is assumed. The time-dependent harmonic signals observed for O_2 were also compared with the theoretical results, and the experimental results have been reproduced well with the theory.

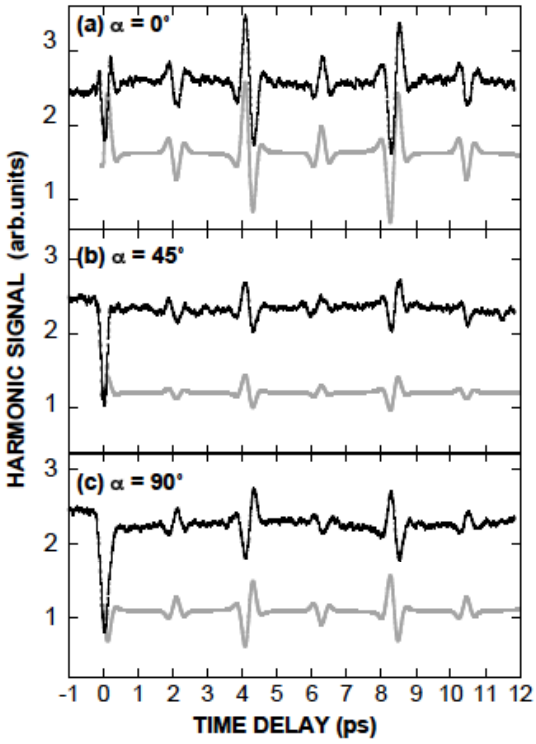


Fig.2. The 19th harmonic signals observed for N_2 as a function of the time delay (black line) and the calculated harmonic signals (gray line) at (a) α = 0°, (b) 45°, and (c) 90°.

The present theory was also applied to reproduce the harmonic signals observed as a function of the angle α between the pump and probe polarizations. The calculated results are in excellent agreement with the experimental. For example, the observed time-dependent harmonic signal was reversed in its amplitude phase at $\alpha \sim 55$˚. This characteristic behavior of the α-dependent harmonic signal was perfectly reproduced with the theory for N_2 and O_2.

It is well known that the angle-dependent HHG from a single molecule is dominated with the HOMO. The π_g symmetry of HOMO and resulting electron distribution in O_2 are quite different from those in N_2 with the σ_g symmetry. Nevertheless, the present experiment has shown that the α-dependent HHG signal for aligned O_2 molecules is very similar to that for N_2. To see the origin of this similarity, we calculated the squared asymptotic wavefunctions of HOMO and the θ-dependent HHG signal from a single N_2 and O_2 molecule, where $\alpha = \theta$ for a single molecule. The

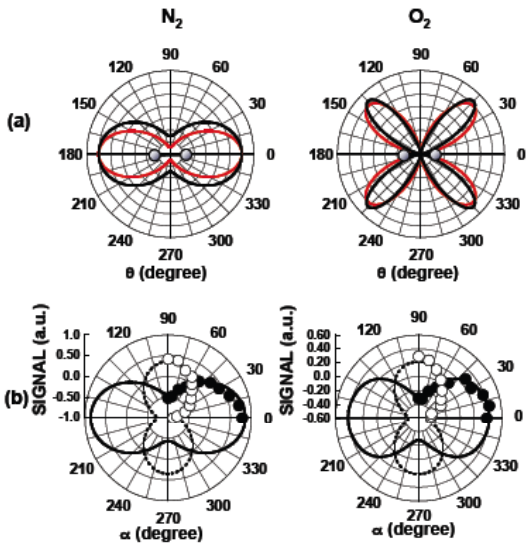


Fig.3. The squared asymptotic wavefunction of HOMO of N_2 and O_2 (red) and the angle-dependent HHG signals for a molecule (black), and (b) the measured and calculated α-dependent signal modulations S_1-S_0 and S_2- S_0.

results are shown in Fig.3, together with the measured and calculated α-dependent signal modulations $S_1 - S_0$ and $S_2 - S_0$ in polar coordinates, where S_1 and S_2 are the maximum and the minimum of the harmonic signal around the half revival time $T_{rev}/2$, and S_0 is the mean value of the background signal.

As seen in Fig.3(a), the electron density in a single N_2 and O_2 molecule is peaked in the direction of $\theta = 0^\circ$ and $\theta \sim 45^\circ$, respectively, and the θ-dependent harmonic signal is subject to the electron density distribution in a molecule. For N_2, as shown in Fig.3(b), S_1–S_0 and S_2–S_0 are peaked and minimized at $\alpha \sim 0^\circ$. The estimated value of $\eta = 0.55$ for N_2 suggests that the degree of alignment is not so large. Nevertheless, the experimental results shown in Fig.3(b) indicate that the α-dependent HHG from the partially-aligned N_2 is strongly dominated with the HOMO of a single molecule. For O_2, the estimated value of $\eta = 0.48$ is comparable with that for N_2. However, the α-dependent HHG observed for O_2 molecules is significantly different from that for a single molecule, as seen in Figs.3(a) and (b), and rather similar to that of N_2 with the maximum to the direction of $\alpha = 0^\circ \sim 20^\circ$. The observed α-dependence shows that the HOMO of *partially aligned* O_2 molecules contributes to the HHG in the direction of $\alpha = 0^\circ \sim 20^\circ$. Then the node at $\theta \sim 0^\circ$ in the electron density distribution of an O_2 molecule is greatly screened in the harmonic signal by the convolution with the alignment distribution.

The excellent agreement between the experimental and theoretical results suggests that the electron density distribution in a single molecule can be deduced by taking into account the alignment distribution in the α-dependent HHG. The characteristic features of HHG

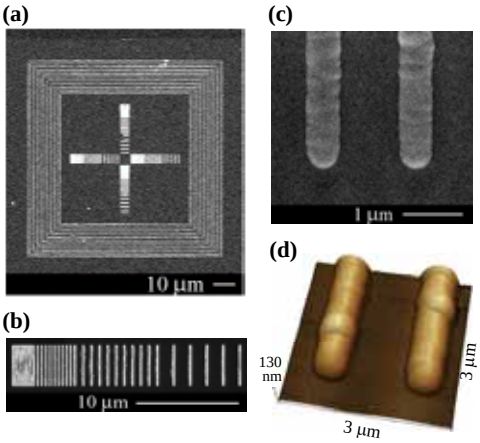


Fig.4. SEM images of (a) the patterned Si substrate, (b) an array of stripes in the central area, (c) the DLC-coated stripes at 800-nm period, and (d) SPM image of the same stripes as in (c).

from aligned molecules were demonstrated for the first time in the present study.

4. Nano-processing with fs laser pulses

When femtosecond (fs) laser pulses are focused on solid surfaces, periodic fine structures are often observed that are much smaller than the laser wavelength λ used. We have found that the structure size as small as ~ λ /10 or 30 nm could be produced on hard thin films such as titanium nitride (TiN) and diamond-like carbon (DLC) films with superimposed fs pulses at fluence less than the single-pulse ablation threshold. Similar fine periodic structures produced with fs laser pulses have been reported for various kinds of materials. The nanostructuring induced by fs laser pulses has attracted much attention as a promising approach to overcome the diffraction limit of light and realize the first *nano-processing with lasers*. For further development of this fs-laser induced nano-structuring, the most important subject to be investigated is the interaction process responsible for the formation of nanosturucture.

In the recent experimental studies we have shown that the nanostructure formation on DLC films is certainly preceded by the change in bonding structure from DLC to glassy carbon (GC). The GC layer was observed to swell on the surface, and the nanostructuring started on the swelled GC surface. Based on the results of reflectivity measurements for the DLC, we have proposed nanoscale ablation induced with a localized field that is enhanced on the film surface. This idea was confirmed by the experiment using DLC and TiN films of which surfaces were patterned with parallel stripes of submicrometer size, where we demonstrated that the nanoscale ablation is preferentially initiated and developed on and/or around the stripe surface with the high surface curvature.

For the experiment, we prepared Si substrates on which Si stripes were fabricated with electron-beam

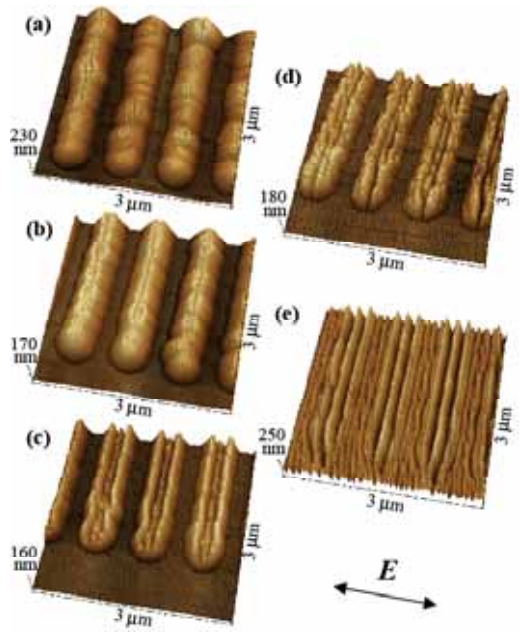


Fig.5. SPM images of the DLC stripes irradiated with (a) N = 100, (b) N = 200, (c) N = 300, (d) N = 500 and (e) N = 1000 at F = 70 mJ/cm^2. The arrow denotes the polarization direction.

lithography and liftoff process. Figures 4(a) and (b) show the scanning electron microscope (SEM) images of the patterned Si substrate and an array of stripes in the central part, respectively. The width, length, and height of a single Si stripe are 0.1 μm, 4 μm, and 50 nm, respectively, and an array consists of the 36 parallel stripes drawn at four different periods of 200, 400, 800, and 1600 nm, as seen in Fig.4(b). The four arrays in the central target area of 50 × 50 μm^2 are set in a cross shape so that the stripes are oriented to both directions parallel and perpendicular to the linearly polarized E-field of incident laser pulses. Figures 4(c) and (d) are the SEM and scanning probe microscope (SPM) images of two stripes coated with DLC, respectively. The DLC of 900 nm in thickness was deposited on the patterned Si substrate with a plasma-based ion implantation system, and then the DLC-coated stripe was ~ 500 nm in width and ~ 100 nm in height, as shown in Fig.4(d). The coated stripes were well isolated at 800- and 1600-nm periods, whereas those at 200 and 400 nm periods were partially overlapped with each other to have a reduced height of the stripes.

Figure 5 shows the SPM images of DLC surface including the 800-nm-period stripes irradiated at different N for F = 70 mJ/cm^2. Comparing with the SPM image of stripe surface shown in Fig.4(d), Fig.5(a) represents the narrow line-like ablation traces that start to be formed in a small area along the crest of stripes. The mean spacing l between the line-like traces or the periodicity of nanostructure on the stripe is measured to be l ~ 60 nm. The nanostructure is certainly formed along the direction perpendicular to the laser polarization or parallel to the stripe. For N = 200, the small structure grows up on the ridge of stripe, as seen in Fig.5(b). With a further increase in N to 300, the mean spacing increases to l ~ 110 nm, while the small ablation traces of l ~ 40 nm start to be formed on the non-patterned surface, as seen in the lower part of Fig.5(c). With increasing N up to 500 – 1000, as shown in Figs.5(d) and (e), the ablation traces on the stripe ridge become deeper with the increased spacing of l ~ 180 nm, while the smaller structure with l ~ 90 nm is created on the non-patterned surface. It is noted that in the area between two stripes the nanostructure grows up almost together with that on the non-patterned surface. The results shown in Fig.5 strongly suggest that the electric field intensity is locally enhanced to initiate the nanometer-size ablation on the stripe ridge, while the laser E-field is kept below the single-pulse ablation threshold.

Based on the results obtained, we can illustrate the interaction process of nanostructuring on the DLC surface. The volume swelling in the change from DLC to GC creates nanometer-size roughness even on a flat DLC surface. The GC layer is able to more efficiently produce free electrons than the DLC in the surface, and then the free-electron density produced would not be uniform in the film surface on the nanometer level. In the laser field, the free electrons coherently oscillate and localize along the E-field direction in the area much smaller than the laser wavelength. The localized electron-hole pairs or the excited surface structure can lead to an enhancement of the evanescent field in the material surface so as to initiate the local ablation, while the laser fluence is less than the ablation threshold. The enhancement of local field intensity should be efficient in the direction of high surface curvature along the laser E-field.

5. Theoretical study of ultrafast laser-atom interactions

There are two main achievements in this year. The first one is that we have theoretically demonstrated that the effects of the carrier-envelope phase can be seen even for chirped pulses in the low intensity regime. The second one is that we have theoretically proposed a new scheme to realize ultrafast "nuclear-spin" polarization which would be of great interest in the broad area of science.

6. Transient critical heat fluxes on inner surface of vertical cylinder

Transient critical heat fluxes and heat transfer characteristics on vertical cylinders of SUS304 and platinum internally cooled by forced flow of pressurized water are studied for a wide range of parameters to clarify the effect of the parameters on the transient critical heat fluxes and heat transfer characteristics and to present the database to realize the high flux heat removal from a divertor of a fusion test facility.

Advanced Energy Storage Research Section

A. Kimura, Professor
K. Morishita, Associate Professor
R. Kasada, Assistant Professor

1. Introduction

Efficient energy conversion and storage are great concerns for sound human life in the near future. This section takes up a mission of materials R & D for advanced nuclear energy conversion and storage, such as development of fusion blanket structural materials and fuel clad materials for high burn-up operation of light water reactors.

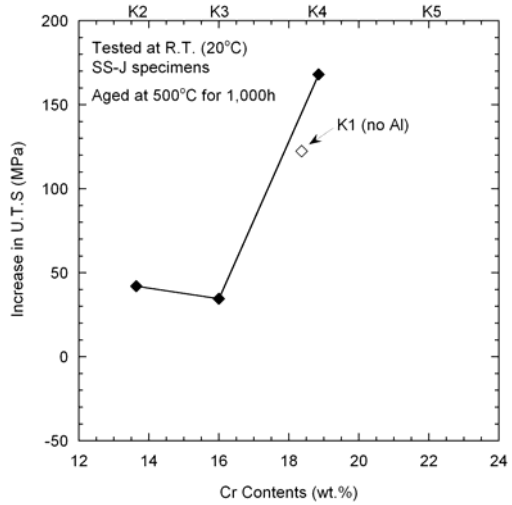
(1) ODS steels for the advanced nuclear energy systems: Oxide dispersion strengthening (ODS) steels have been considered to be very promising for application to advanced nuclear plant as structural materials, because they are highly resistant to thermal recovery of the material structure as well as to neutron irradiation embrittlement. Although the other critical issue of the ODS steels has been considered to be corrosion resistance in the super-critical pressurized water (SCPW) and Pb-Bi (LBE), no data was obtained so far. From fiscal year 2005, our research group has begun a MEXT project **"R&D of Super ODS steels for the Advanced Nuclear Energy Systems"**.

(2) Multi-scale modeling of fusion blanket structural materials: Reduced activation ferritic steels (RAFS) and SiC/SiC composite are the promising candidates for fusion structural materials for future fusion reactors where structural materials are expected to suffer sever irradiation embrittlement. The multiscale modeling approach is very useful to understand and reduce the embrittlement.

(3) Lifetime evaluation of nuclear structural materials: For the sake of the highly efficient and safe operation of nuclear fission reactors, the mechanisms of irradiation embrittlement and stress corrosion cracking have been investigated.

2. R&D of High-Cr ODS steels for the Advanced Nuclear Energy Systems

For high burn-up operation of light water reactor (LWR), realization of Gen IV reactor concepts, such as super critical pressurized water reactor (SCPWR) and lead-bismuth eutectic fast reactor (LBE-FR), and fusion reactor, a series of high-Cr oxide dispersion strengthened (ODS) ferritic steels (named as "K" series) have been developed in Kyoto University under the collaboration with JNC and KOBELCO Research Institute Inc. as fuel cladding materials. Up to now, it has been confirmed that the addition of chromium over 14 wt% and 4 wt% aluminum were very effective in suppressing corrosion of the previous 9Cr-ODS steel in supercritical pressurized water and liquid lead-bismuth. The high-Cr ODS ferritic steels also have excellent



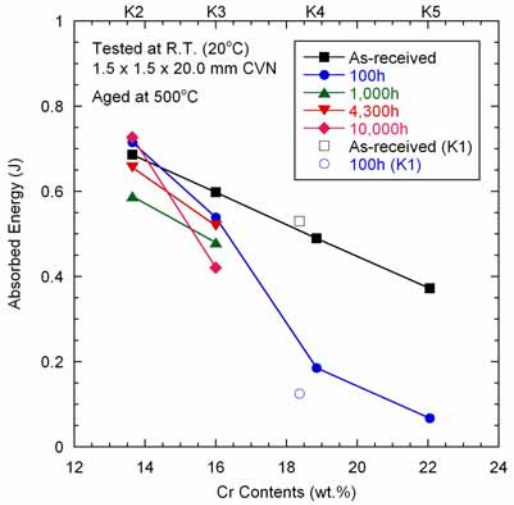


Fig. 1 (left): Cr-dependence of increases in UTS obtained form tensile tests after thermal aging at 500 °C for 1,000 h.
Fig. 2 (right): Cr-dependence of the absorbed energy of Charpy impact test at room temperature after thermal aging at 500°C.

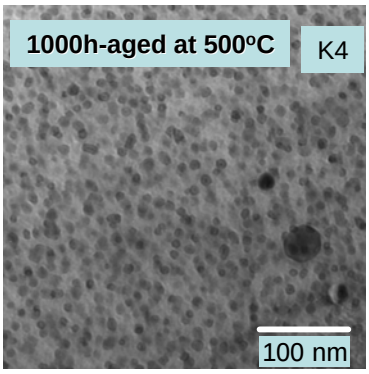


Fig. 3 TEM micrograpgh of K4 ODS steel after thermal aging at 500°C.

mechanical properties at elevated temperatures because of ultra-fine nano-scale oxide particles dispersed in the ferrite matrix. However, at medium-temperatures, high-Cr ferritic stainless steels will suffer aging embrittlement (475 ºC embrittlement), while they will show excellent corrosion resistance. In this study, effects of neutron irradiation and thermal aging on fracture behavior and mechanical properties of the high-Cr ODS ferritic steels are presented. Cr-dependence of increase in the ultimate tensile stress (UTS) obtained from tensile tests at room temperature (20 °C) and the absorbed energy of Charpy impact tests at room temperature after thermal aging at 500 °C are shown in Fig. 1 and Fig. 2, respectively. The K4 (19Cr-4Al) ODS steel showed a significant hardening and reduction of absorbed energy at room temperature. These hardening and embrittlement during aging has been thought to be due to the Cr-rich phase formation observed in the K4 ODS steel aged at 500 °C for 1,000 h as shown in Fig. 3.

Fig. 4 shows the Cr-dependence of increases in a) UTS and b) total elongation after neutron irradiations in JMTR at 290, 400 and 600 °C. After the irradiation at 290 °C, the irradiation hardening appears to be independent of Cr contents, although the hardening is significant. On the other hand, after the irradiation at 400 °C, the irradiation hardening increases with Cr contents. It should be noticed that less irradiation effect on the total elongation was observed. Concerning the effects of the neutron irradiations on the Charpy impact properties, as shown in Fig. 5, remarkable reduction of the absorbed energy at room temperature was observed after the irradiation at 400 °C except for the K2 (14Cr-4Al) ODS steel. Fig. 6 shows the TEM image of the K4 (19Cr-4Al) ODS steel after irradiation at 400 °C. A number of spherical precipitates, which are thought to be Cr-rich phase, were observed as well as observed in the K4 ODS steel aged at 500 °C. After the irradiation at 290 °C, many dislocation loops were observed in the K3 (16Cr-4Al) ODS steel, as shown in Fig. 7. The average diameter and number density of the dislocation loops were 12 nm and 6.1 × 1021 m-3. By applying the Orowan equation [5], it can be concluded that irradiation hardening at 290 °C is mostly due to the formation of dislocation loops. Less change after the irradiation at 600 °C was observed for tensile properties, Charpy impact properties and microstructure of the high-Cr ODS steels.

3. Lifetime evaluation of nuclear structural materials

In this fiscal year, our research group has begun a new METI project **"FY 2007 educational program (Nuclear Energy Fundamental Technology Field Reinforcement Program) to develop human resources in the nuclear energy field"**.

Irradiation hardening is one of the essential degradation issues of Fe-based ferritic alloys for fusion reactor blanket structural materials as well as reactor pressure vessel (RPV) steels of light water reactors. While many studies have agreed that the irradiation hardening of the ferritic alloys should be due to irradiation-induced/enhanced precipitates and matrix defects like dislocation loops and nano-voids, knowledge concerning effects of alloying element on the irradiation hardening of the ferritic alloys is significantly limited to some of elements. It is well

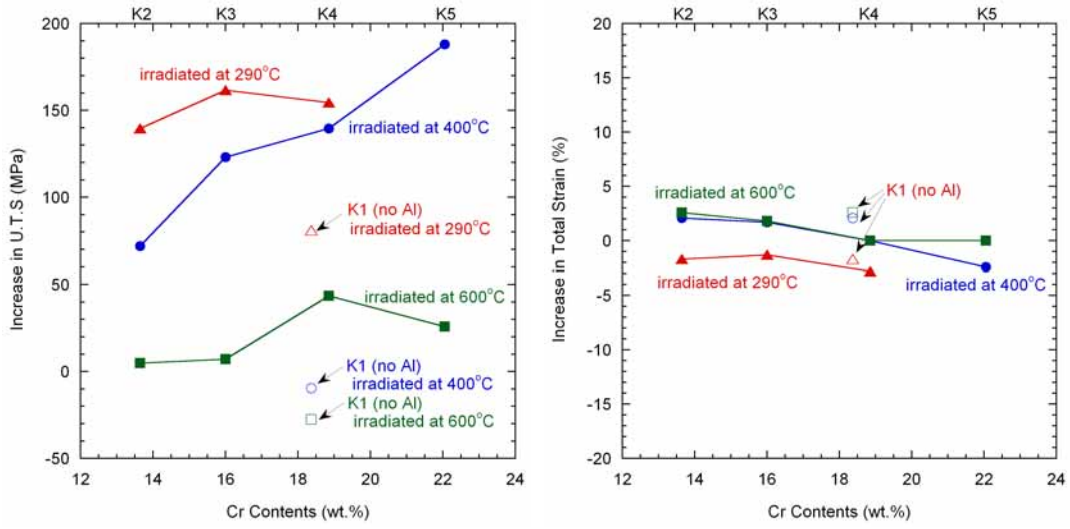


Fig. 4 Tensile test results, a) UTS, b) total elongation, after neutron irradiations at 290, 400 and 600°C.

known that Cu-rich precipitates were responsible to the irradiation hardening and embitterment for the aged RPV steels containing impurity Cu. As for the Cu-rich precipitates, recent advanced techniques, such as three-dimensional atom-probe (3DAP) and positron annihilation spectrometry (PAS), elucidated the microstructural development of the Cu-rich precipitates in irradiated RPV steels, and model alloys. While irradiation hardening due to dislocation loops and nano-voids should be affected by alloying elements, less information is available. Fe-Cr ferritic alloys have been investigated because Cr is one of the major alloying elements in the reduced-activation ferritic steels for fusion reactor blanket structural materials. Suganuma and Kayano revealed that the irradiation hardening of Fe-Cr binary alloys increased with Cr contents. While the irradiation hardening behavior of the ferritic "steels" containing many elements and complex microstructural features, such as precipitates and dislocations, has been investigated and summarized as substantial databases, the mechanistic understanding is necessary to use them for designing fusion reactors.

Self-ion (Fe-ion) irradiation technique is very useful to understand irradiation effects of ferritc alloys with various conditions, such as irradiation temperature, irradiation dose, damage rate, and so on. Applying a nano-indentation technique to evaluate the irradiation hardening of ion-irradiated ferritic alloys, furthermore, the hardness change can be obtained within a surface of only about 1 μm irradiated by the ion.

In the present study, we describe a systematic result concerning the effects of alloying elements on the irradiation hardening of Fe-based binary alloys containing about 1 at.% elements, such as Cr, Cu, Mo, Mn, and Ni, after Fe^{3+} ion irradiation by using a nano-indentation technique.

Fig. 5 shows the dose dependence of the irradiation hardening of the Pure-Fe and Fe-based binary alloys after Fe^{3+} irradiations at 290 °C. While data scattering shown was larger than that of typical micro-hardness tests, difference of the dose dependence between alloys can be clearly seen. Irradiation hardening behavior of Fe-1Cr and Fe-1Mo was similar with that of Pure-Fe showing the lowest irradiation hardening in the alloys investigated at the displacement damage of 1 dpa. Saturation of the irradiation hardening was observed in Fe-1Cu irradiated up to 0.1 dpa. This saturation behavior has been also reported for the iron-copper binary alloys irradiated with electron and neutron, and was explained by formation of Cu-rich precipitates. It is noted that significant irradiation hardening was observed in the Fe-1Mn, Fe-1.5Mn and Fe-1Ni after the irradiation up to 1 dpa as well as the Fe-1Cu irradiated. The irradiation hardening of these alloys increased with displacement damage and was seen to be not saturated at 1 dpa.

Effects of saturation at higher doses were explained by so-called Makin-Minter model. Fig. 5 shows the experimental data and their fitting curves by using $\Delta H = A[1-\exp(-B \times \mathrm{dpa})]^{0.5}$ considering the saturation effects. According to this model and its extended understanding by Yamamoto et al., the saturation can be physically related to the depletion of solutes in the case of a precipitation hardening mechanism, or an excluded zone effect in the case of the accumulation of displacement damage-type defects. The former case is corresponding to typical Cu-rich precipitation hardening observed in the Fe-1Cu. The latter case must be principally related to the irradiation hardening observed in the Pure-Fe. Moreover, the significant irradiation hardening observed in the Fe-1.5Mn, Fe-1Mn, and Fe-1Ni was probably due to the effect of alloying element on the surviving defect and/or defect clusters produced from a displacement cascade. However, the difference of irradiation hardening behavior between the various Fe-based binary alloys could not be explained simply by the size-factor of solutes in bcc Fe matrix on the evolution of dislocation loops and following irradiation hardening because the size factor of Mn (+4.81%) and Ni (+4.65%) in the bcc Fe was similar with Cr (+4.36%). Nagai et al. experimentally revealed formation of vacancy-solute complex in the electron-irradiated Fe-Mn binary alloys and vacancy cluster in the electron-irradiated Fe-Ni. On the other hand, Vincent et al. suggested formation of Fe-Mn mixed dumbbells with a high probability for its formation and its migration from ab initio calculations. Further investigations are necessary to distinguish these complex phenomenons especially concerning state of Mn in bcc Fe matrix under irradiations.

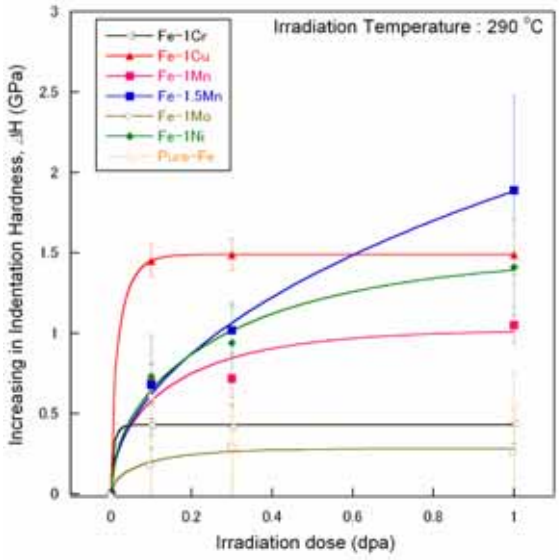


Fig. 5 Fitting curves for the dose-dependence of irradiation hardening of Pure-Fe and Fe-based binary alloys by using $\Delta H = A[1-\exp(-B \times \mathrm{dpa})]^{0.5}$.

4. Theory & modeling of radiation damage processes in fusion materials

SiC/SiC composites are expected as one of blanket structural candidate materials in a nuclear fusion reactor because of good mechanical properties and low activation properties. Point defects and defect clusters such as interstitial loops and voids are produced and accumulated in materials during irradiation. This changes the material microstructure which influences material properties such as mechanical properties, dimensional stabilities and thermal mechanical properties. Irradiation effects on SiC materials have been widely studied by a variety of experiments and numerical analysis. However, defect accumulation process is not well understood, because SiC materials have an ionic nature in addition to a covalent one and therefore point defects and defect clusters can have effective electric charges which will lead to the complicated diffusion and growth behavior of defects. Ryazanov et al. have proposed a model for formation of interstitial loops taking into account charge state of defects in β-SiC under irradiation, where interstitial loops grow and maintain its neutral charge. Interstitial loops treated in the model are relatively large loops recognized as perfect loops. In the present work, the model applied for nucleation and growth of small interstitial loops taking into account defect charge state is developed, where small interstitial loops treated here are small defect clusters recognized as so-called black dots. Such defect clusters in β-SiC are observed during high-energy electron irradiation in high voltage electron microscopy (HVEM).

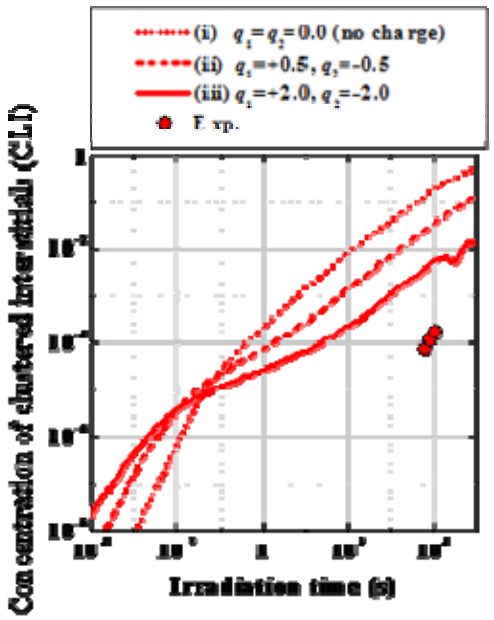


Fig. 6 The time dependence of calculated concentration of clustered interstitials (CLI) under the condition of three different charge states (i) $q_1 = q_2 = 0.0$, (ii) $q_1 = +0.5$, $q_2 = -0.5$ and (iii) $q_1 = +2.0$, $q_2 = -2.0$.

In the present study, the capture efficiency of a mobile defect by a sink is derived, where electric interaction between a sink and mobile defect is considered. And then, numerical analysis of accumulation of small interstitial loops during high-energy electron irradiation is conducted using the kinetic rate theory based model with the derived capture efficiency. Finally, the calculation results were compared with a HVEM experiment.

Consider a charged mobile defect diffusing in the electric field created by a charged spherical sink. When the mobile defect has electric charge in electric field, the defect migrates depending on the gradient of the electric potential in addition to the gradient of the defect concentration. Thus, the diffusion flux of mobile defects can be expressed as

$$\boldsymbol{j} = \frac{1}{\Omega}\left(-D\nabla C - \frac{eqDC}{k_{\mathrm{B}}T}\nabla V\right)$$

where e is the elementary electric charge, q is effective charge of mobile defect, V is electric potential at the position of the defect, k_{B} is the Boltzmann constant and T is the temperature. Interstitial loops treated here are black dots, which are regarded as small spheres. Assuming that the electric potential V is created by a charged spherical sink, then the diffusion flux of the mobile defect by the sink can be described as $\boldsymbol{j}(r, \theta, \varphi)=(J_r(r), 0, 0)$ in polar coordinate, where $j_r(r)$ is the radial component of the diffusion flux at the distance r from the sink. When $V(r)=-eQ/4\pi\varepsilon_0 r$ is applied, the total current of charged mobile defects to the sink is written by

$$J = \frac{1}{\Omega}4\pi R_0 Z\left[DC(\infty) - DC(R_0)\exp\left(\frac{e^2qQ}{4\pi\varepsilon_0 R_0}\frac{1}{k_{\mathrm{B}}T}\right)\right],$$

$$Z = \begin{cases} \dfrac{\dfrac{e^2qQ}{4\pi\varepsilon_0 R_0}\dfrac{1}{k_{\mathrm{B}}T}}{\left[\exp\left(\dfrac{e^2qQ}{4\pi\varepsilon_0 R_0}\dfrac{1}{k_{\mathrm{B}}T}\right)-1\right]} & \ldots \quad (qQ \neq 0) \\ 1 & \ldots \quad (qQ = 0). \end{cases}$$

Fig. 6 represents the time dependence of the concentrations of clustered interstitials (CLI) as a function of cluster charge states, where CLI is defined as the total number density of interstitials in clusters. As shown in the figure, the concentrations of clustered interstitials greatly depend on the charge state and they decrease with increasing the magnitude of charges. Fig. 6 also indicates the result of HVEM experiments. The calculated concentration of clustered interstitials approaches to the experimental data. This may imply that the capture efficiency derived here is important for better description of the experimental observation.

Complex Plasma Systems Research Section

F.Sano, Professor
K.Hanatani, Associate Professor

1. Introduction

The development of magnetic fusion as a commercial reactor of electricity requires the solution to the physics problems such as plasma transport and magneto-hydrodynamics. The goal of the fusion plasma research is the discovery of a magnetic configuration that can efficiently confine high density plasma at high temperature for sufficiently long confinement time to produce net thermonuclear power. This research section seeks to investigate the confinement optimization of high-temperature plasma in the helical-axis heliotron line. For the experimental and theoretical investigation of this theme, the plasma device of Heliotron J has been operated at the Laboratory for Complex Energy Processes since FY2000 to study the magnetic configuration effects of confinement with special regard to the improved bulk plasma confinement regime, H-mode., the bumpiness effects of the high-energy particle confinement, the non-inductive plasma current properties, MHD activities, and particle and heat load handling in the SOL/divertor region. With regard to these experimental studies, the detailed theoretical and computer simulation studies of plasma transport have also been carried out.

2. Effect of toroidal current on rotational transform profile by MHD activity measurement

Non-inductive toroidal currents can modify the MHD equilibrium and stability due to the change in the rotational transform profile. In Heliotron J, the approach to the transition to the improved confinement mode has been investigated [1]. The experiments show that the transition occurs at a certain value of the toroidal current [2]. The modification of the rotational transform by the toroidal current may induce a spontaneous transition. The effect of toroidal current on the rotational transform ($\iota/2\pi$) by measuring MHD activities has been investigated in Heliotron J [3].

Figure 1 shows a discharge with ECH+co-NBI for the $\iota/2\pi$ = 0.48 configuration. Here, "co-" means the toroidal current increases the vacuum rotational transform and "counter-" means the toroidal current decreases the vacuum rotational transform. An ECH power of 208 kW is injected from 165 to 265 ms. An NB port-through power of 573 kW is injected from 175 to 265 ms. The net toroidal current measured by Rogowski coils is increased up to 2.5 kA in the co-direction. The magnitude fluctuation of m/n = 2/1 mode with frequency of 3 kHz is observed from 225 to 245 ms. Here, m and n are the poloidal and toroidal mode numbers, respectively. The fluctuation of soft X-ray (SX) signal also appears simultaneously. Figure 3 shows the relative amplitude of the fluctuations δI_{SX} observed in the SX signal. Peaks are observed at #5 (ρ = 0.90), #15 (ρ = 0.45) and #18 (ρ = 0.80). Here, ρ is the normalized minor radius. The phases of #5 and #18 are almost the same and they are consistent with the m = 2 determined using the magnetic probe arrays. Thus, they correspond to the location of the resonant surface of the m/n =2/1 mode. In the vacuum magnetic surface of the $\iota/2\pi$ = 0.48 configuration, there is no rational surface of the m/n = 2/1, however, the rotational transform is affected by the plasma pressure and co-flowing toroidal current, resulting in crossing of the rational surface of the m/n = 2/1 mode.

The effect of toroidal current on $\iota/2\pi$ was also investigated in an ECH+counter-NBI plasma at the $\iota/2\pi$ = 0.50 configuration. The net toroidal current is increased up to 1 kA in the co-direction. The total toroidal current during counter-NBI injection is smaller than that during co-NBI injection since the NB driven current flows in the counter-direction against the bootstrap current. The m/n = 2/1 mode with frequency of 5 kHz is observed and the fluctuation of SX signal also appears simultaneously. The peak of the relative amplitude is observed at #3 and #19. The phases of #3

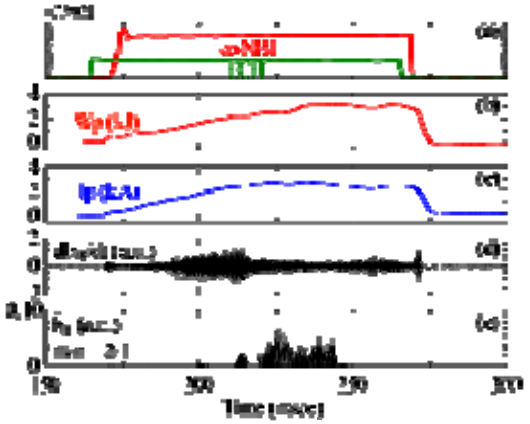


Fig. 1 Time evolution of an ECH + co-NBI plasma. The (a) NBI and ECH, (b) stored energy, (c) plasma current, (d) Mirnov signal, and (e) Magnetic fluctuation of m/n = 2/1 mode.

and #19 are almost the same, which is consistent with the m = 2 identified by magnetic probes. The positions are almost identical to the rational surface of 0.5 in the vacuum configuration. This suggests that the position of the rational surface does not move even though the finite net current exists.

In order to determine the effect of the toroidal current on the equilibrium, fixed boundary VMEC calculations [$4] taking into account the toroidal current were carried out in ECH+co-NBI plasma. The calculation result shows that when a bootstrap current of 1.5 kA and NB driven current of 1 kA flow in co-direction, the rotational transform profile has the rational surface of the m/n = 2/1 mode around ρ = 0.8. The calculated

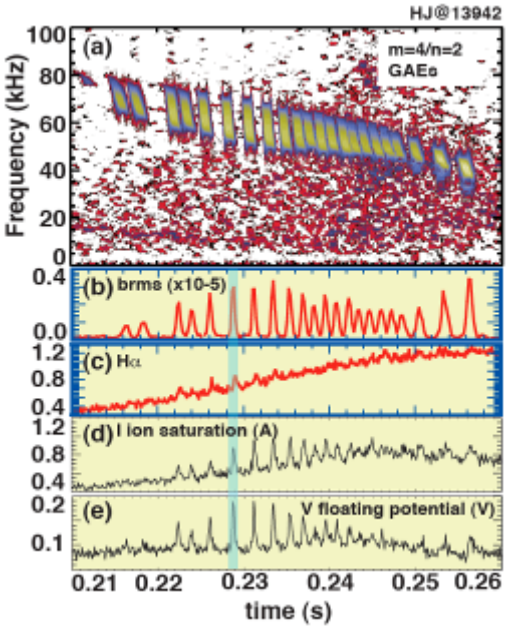


Fig. 2. Time evolution of (a) frequency spectrum and (b) amplitude of observed m=4/n=2 GAEs, (c) H_α/D_α signals, and (d) ion saturation current and floating potential obtained from Langmuir probes.

location of the rational surface of the m/n = 2/1 is consistent with that obtained from the SX signals. The VMEC calculations were also performed in ECH+counter-NBI plasma. In ECH+counter-NBI plasma, there is no significant difference between the rotational transform profile that takes into consideration the toroidal currents and that of the vacuum configuration, as compared with the ECH+co-NBI plasma. These results suggest that the change in the rotational transform profile induced by the toroidal current is small because the counter-flowing NB driven current density decreases the rotational transform. The assumed toroidal current profiles are considered to be valid. However, we will discuss the effect of some current density profiles on $\iota/2\pi$ in the future.

3. Study of energetic ion driven MHD instabilities and their effects in Heliotron J

To clarify the MHD instabilities such as Alfvén eigenmodes (AEs) destabilized by energetic ions is important for the Deuterium-Tritium (D-T) fusion plasmas because the MHD instabilities could lead to the loss of alpha particles from confinement region before their thermalization and the ejected alpha particles might cause significant damage of first wall of a fusion device.

In Heliotron J with low magnetic shear, Global AEs (GAEs), which can exist on just below of upper and above of lower shear Alfvén continuum, are typically observed in the NBI (neutral beam injection) heated plasma. It is important and of interest to investigate the GAEs in the Heliotron J plasmas for an advanced stellarator type fusion reactor with low magnetic shear. We utilize the Alfvénic energetic ions produced by the tangentially co.- and counter-injected hydrogen neutral beams with the energy of 24~27 keV for the destabilization of GAEs.

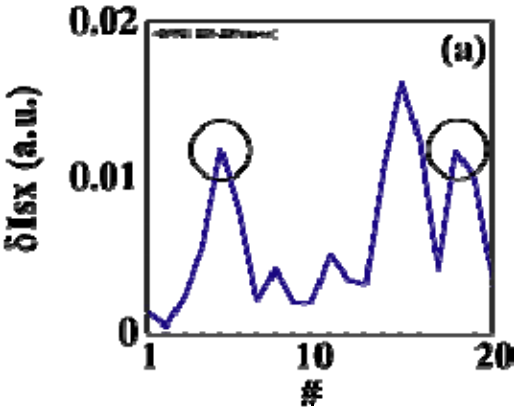


Fig. 3 Radial profile of SX fluctuation amplitude in ECH+co-NBI plasma.

GAEs with m=2/n=1 (m and n: poloidal and toroidal mode number) and m=4/n=2 are typically observed in the NBI heated plasmas of Heliotron J [5]. These modes propagate in the ion diamagnetic drift direction and of which frequency corresponds to that of discrete mode obtained from CAS3D3 [6] analysis. In the plasma with magnetic configuration having the higher bumpy field, bursting GAEs (m = 4/n = 2), of which amplitude is two times larger than that of continuously observed GAEs are often observed, as shown in Fig. 2 (a)~(b). The frequency of bursting GAEs usually chirps down quickly. The time interval between each bursting GAEs increases with the increased amplitude of bursting GAEs. This phenomenon can be explained by predator-prey model between energetic ion driven mode and energetic ions, and might indicate the energetic ion transport induced by the energetic ion driven mode. The some plasma parameters such as H_α/D_α and, ion saturation current and plasma floating potential which are obtained from Langmuir probes located at outside last closed flux surface of plasma, are simultaneously increased with bursting GAEs, as shown in Fig.2 (c)~(e). The increasing of ion saturation current is related to the

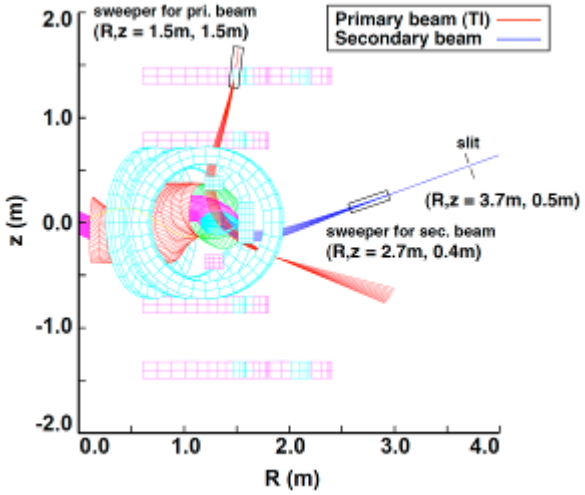


Fig. 2. HIBP system and poloidal cross-section of Heliotron J. Red and blue lines are primary (Tl^{1+}) and secondary (Tl^{1+}) beam. There are two sweepers for the spatial measurement.

increasing of amplitude of GAEs. Although the increase in Langmuir probe signals cannot directly indicate the loss of energetic ion from the confinement region, the simultaneous increasing of the some plasma parameters indirectly indicates the energetic ion loss induced by the busting GAEs. The degradation of stored plasma energy induced by the bursting AEs observed in LHD and W7-AS were not observed in Heliotron J plasmas.

4. Development of HIBP system for Heliotron J

We designed the heavy ion beam probe (HIBP), which can simultaneously measure both the electric field and electron density inside plasma in order to study the neoclassical and anomalous transport. HIBP system consists of the accelerator for primary beam (Maximum voltage for acceleration Vacc=140keV), electrostatic analyzer for detection and measurement of the secondary beam, which has information of plasma such as electric field and electron density, and deflector of primary and secondary beam for the spatial measurement (Fig. 4). The primary beam (Tl^{1+}) is injected from upper port of Heliotron J and secondary (Tl^{2+}) beam ionized at measurement point inside plasma pass through the slit and detect at analyzer where magnetic field, which would effect on beam trajectory is typically less than 50 Gauss. When we control the sweeper voltage, HIBP can measure the plasma parameters from the core toward edge.

5. Development of δf Monte Carlo simulation: Monte-Carlo collision operators and the model of momentum and energy conserving terms.

Neoclassical transport is important to investigate the confinement properties in the nonaxisymmetric systems such as Heliotron J because the radial flux is enhanced due to the particles trapped in helical ripples. Although, the radial flux of particle, momentum, or energy may in general be determined by turbulent transport, the neoclassical transport flux in helical systems determines the radial electrostatic potential with the ambipolar condition; and it also dominate the turbulent flux through the zonal flow generation [8]. And the transport flux tangent to the magnetic surface can be well predicted by the neoclassical theory. The bootstrap current is a key factor to control MHD equilibrium and stability in the helical system. But the analytical theory is not sufficient for calculating the neoclassical transport matrix in the realistic magnetic field, especially in the long mean free path regime. To calculate the neoclassical transport in such a complicated situation, we are developing δf Monte Carlo code. Here, we report its features focusing on the modeling of the collision operator with the good conservation properties.

We briefly describe the formulation of δf scheme [10]. The conventional Monte Carlo simulations [9] have been used to evaluate the radial flux in the nonaxisymmetric system based on the total-f scheme. Many particle orbits are solved by the Langevin-like guiding center equation and the diffusion coefficient is calculated from the mean square displacements. But the parallel fluxes like the bootstrap current cannot be treated accurately by such scheme because of the significant statistical noise. One of that reasons is the profile relaxation effect that is not important for the linear neoclassical theory. On the other hand, in the δf simulation, the distribution function is divided into the known analytical part and the unknown perturbation. This eliminates the profile relaxation effect from the simulation and allows us to reduce the statistical noise by orders of 10^{-2}. The distribution function is written as follows; $f = f_0 + \delta f$, where we choose f_0 as the Maxwellian, $f_0 = n_a \pi^{-3/2} v_{tha}^{-3} \exp(-E/T_a) \exp(e_a\Phi/T_a)$. $v_{tha} = (T_a/2m_a)^{1/2}$ is the thermal velocity of species a, n_a is the density, and T_a is the temperature. The drift kinetic equation that describes the evolution of the perturbed part of the guiding center distribution function is;

$$\frac{\partial \delta f}{\partial t} + v_{\parallel}\mathbf{b}\cdot\nabla\delta f - C^L(\delta f) = -\mathbf{v}_d\cdot\nabla f_0,$$
$$\mathbf{v}_d = \Omega_a^{-1}\mathbf{b}\times(v_{\parallel}^2\mathbf{b}\cdot\nabla\mathbf{b} + v_{\perp}^2\nabla\ln B/2 + e_a\nabla\Phi)$$

Here, $\mathbf{v}_d$ is the guiding center drift velocity and $C^L(\delta f)$ is the linearized Coulomb collision operator, $\Omega = e_aB/m_a$, $v_{\parallel}$ is the parallel velocity, $v_{\perp}$ is the perpendicular velocity, and Φ is the electrostatic potential; f/f_0, $|\mathbf{v}_d|/v_{th}$, and $(e_a\Phi/T_a)$ are considered to order $\varepsilon = \rho_{th}/a$, where ρ_{th} is the gyroradius and a is the plasma minor radius. We solve this equation by the Lagrangian marker with the weight function which varies along the orbits. The evolution of the weight function can be described by

$$\frac{dw}{dt} = (1-w)\mathbf{v}_d\cdot\vec{\kappa} + (\Delta w)_{FP},$$
$$\frac{d}{dt} = \frac{\partial}{\partial t} + v_{\parallel}\mathbf{b}\cdot\nabla - C_{TP},$$

where C_{TP} is the test particle term of the collision operator and $(\Delta w)_{FP}$ is the correction of weight function for satisfying the conservation laws of the operator.

There are several expressions of test particle term, which are used commonly. For example, the pitch angle scattering operator proposed by Boozer and Kuo-Petravic is $\lambda_n = \lambda_o(1-\nu_d\tau) \pm [(1-\lambda_o^2)\nu_d\tau]^{1/2}$. This expression is used for monoenergetic calculation. On the other hand, to consider the thermalization effect due to the like particle collisions, energy scattering terms are needed. In this report, we used the expressions of Lin, Tang and Lee [10];

$$v_{\parallel} = v_{\parallel 0} - \nu_{s\parallel}\Delta t + \sqrt{12}(R_1 - 0.5)\sqrt{\nu_{\parallel}\Delta t},$$

$$v_{\perp}^2 = v_{\perp 0}^2 - \nu_s^{\perp}\Delta t + \sqrt{12}(R_2 - 0.5)\sqrt{\left(\nu_{\perp} - \frac{\nu_{\parallel\perp}^2}{\nu_{\parallel}}\right)\Delta t}$$

$$+\sqrt{12}(R_1 - 0.5)\frac{\nu_{\parallel\perp}}{\nu_{\parallel}}\sqrt{\nu_{\parallel}\Delta t}$$

The notations used above are given in Ref.[10].

Another advantage of the δf scheme is the momentum conservation property can be incorporated with the correction term $(\Delta w)_{FP}$. Several methods are proposed and we tested the following three schemes [10-12];

Dimits & Cohen (DC);

$$\Delta w = -\mathbf{v}\cdot\delta\mathbf{P} - \left(\frac{v^2}{v_{th}^2} - \frac{3}{2}\right)\delta E$$

$$\delta\mathbf{P} = \frac{2}{n v_{th}^2}\sum_i w_i \Delta v_i$$

$$\delta E = \frac{2}{3 n v_{th}^2}\sum_i w_i \Delta v_i^2$$

Lin, Tang & Lee (LTL);

$$\Delta w = -3\sqrt{\frac{\pi}{2}}\phi(x)\left(\frac{v_{th}^2}{v^2}\right)^3 \mathbf{v}\cdot\delta\mathbf{P}$$

$$-3\sqrt{\frac{\pi}{2}}\left(\phi(x) - \frac{d\phi(x)}{dx} - \frac{3}{2}\right)\frac{v_{th}}{v}\delta E$$

Sasinowskii & Boozer (SB);

$$\Delta w = -3\nu\frac{u_{\parallel}}{v}\lambda$$

Because the SB model is used for monoenergetic calculation, this does not contain the energy conserving term for the field particle collisions. The LTL model is the modified version of the DL model and that take into account for the test particle energy dependence of the filed particle contributions. To check the accuracy of the conservation, the shifted Maxwellian is loaded as the initial condition and the marker weights are pushed only by the collision operator. The results can be summarized as follows. When we used no momentum-conserving operator, initially loaded average flow was damped within several collision times. On the other hand, the momentum-conserving operator properly conserve the averaged flow. No clear differences between the LTL and DL, but the SB model showed the low accuracy for the long time calculation. It must be noted that these models treat correctly about the three lowest order moments alone. This artificial correction may distort the distribution function in respect to the higher order moment.

In our code, the individual orbits are calculated in the Boozer coordinates [9]. The well-known problem of the δf neoclassical simulation is the entropy growth of the weight functions [13]. To overcome this difficulty, the appropriate weight averaging schemes or the weight spreading reduction schemes are needed. We are now trying to adopt this method in our δf code.

In summary, we are developing the δf Monte Carlo code for neoclassical transport calculations. In this report we focused on the modeling of the collision operators. Three collision operators were tested to obtain the conservation laws of the Coulomb collision operators. The effects of numerical techniques, such as the weight averaging on the flux surface, should be considered in future work. In applications of the code, the neoclassical transport flux will be calculated for the nonaxisymmetric devices such as Heliotron J.

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Clean Energy Conversion Research Section

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(Kyushu Institute of Technology)

1. Introduction

Photo-conversion efficiencies for dye sensitized solar cells (DSC) have been reported to reach 10-11 % [1] which is almost the same as that of amorphous Si type solar cells. The next target of the efficiency has been set to be 15 %. In order to increase the efficiency, we focused on improving charge carrier collections. Figure 1 shows a working principle of DSCs and parameters we measured for the DSC studies. How to collect charge carriers from the charge separation area is crucial. In this paper, we focus on the fabrication of charge carrier path (electron-paths and ion-paths).

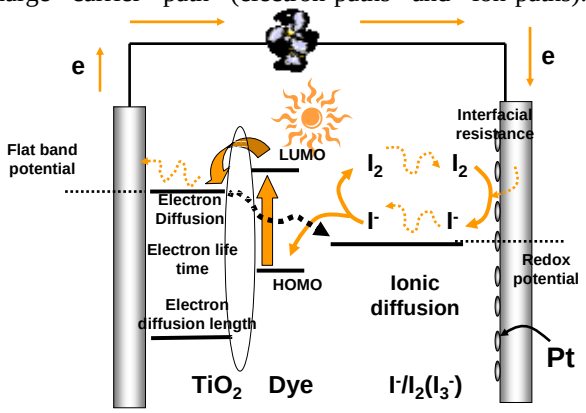

Figure 1. Working principle of DSCs

2. Fabrication of electron-paths[2]

Since electron diffuses by trapping and detrapping electron-traps on TiO_2, the presence of electron traps, especially, surface traps of highly nano-TiO_2 surfaces, disturbs the efficient electron collection. In addition, surface traps become the center for the charge recombination. In order to increase the solar cell efficiency, surface states have to be passivated. It has been reported that organic molecules bearing carboxylic moieties (including Ru dyes) passivated surface traps [3]. In order to passivate highly nano-porous TiO_2, dye-staining of the TiO_2 surface was carried out under pressurized CO_2 conditions instead of the commonly employed dipping conditions, since it has been reported that the diffusion rate of molecules in supercritical CO_2 fluid is 10-7-10-8 m^2/s which is 10 times higher than that in liquid phase.

The black-dye-staining of TiO_2 layer occurred gradually and it took 100 hours for the dye-uptake to level off. On the other hand, the dye uptake on a TiO_2 layer swiftly occurred in the pressurized

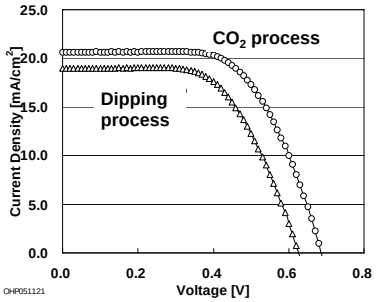

Figure 2. Photovoltaic properties for cells prepared by CO_2 and dipping processes

100mW/cm², AM1.5. LiI:500mM, TBP:580mM, I_2:50mM, MePrImI:600mM in Acetonitrile, BDCO2: 23Mpa. 40 °C, 40 min. BDDIP: 48 h at room temperature. 5 mm x 5 mm.

CO2 atmosphere and completed within 2 hours. Figure 2 shows the photovoltaic properties for the DSC prepared by a dipping process and a CO_2 process. Open circuit voltage (Voc) and short circuit voltage (Jsc) for the cell prepared by the CO_2 process were higher than those prepared by a dipping process. The electron diffusion coefficient in TiO_2 layers for the cell prepared by the CO_2 process was higher and the electron life time was longer. The surface states of TiO_2 layers can be evaluated by measuring thermally stimulated current (TSC) [4] which is associated with trap distributions. After the TiO_2 surface was covered with dye molecules, the TSC for the TiO_2 layer decreased, supporting strongly the explanation that the surface state of the TiO_2 was passivated by the adsorption of the dye molecule. The increase in Voc and Jsc of the cell prepared by the CO_2 process can be explained by the better coverage of the TiO_2 surface with the black dye molecule. Namely, the black dye passivates effectively the surface states of TiO_2 prepared under the pressurized CO_2 condition, which aids to make electron diffusion paths in the TiO_2 layer.

3. Suppression of black dye aggregation in a pressurized CO2 atmosphere

Black dye is commonly adsorbed on TiO_2 layers with co-adsorbents such as deoxycholicacid (DCA) in order to suppress black dye aggregation. It has been reported that dye aggregation decreases photovoltaic performances. When a TiO_2 substrate was dipped in the black dye solution, the absorption peak of stained TiO_2 electrode blue shifted gradually and peaks of 520 -530 nm increased. Considering the fact that the blue shift was not observed in the presence of an aggregation inhibitor (Deoxycholic Acid), it is reasonable to explain

that the shift is associated with the black dye aggregation. Even when the dye was adsorbed on a porous TiO_2 electrode in a pressurized CO_2 condition for two hours, the blue shift was not observed, suggesting that the dye aggregation was also suppressed under the pressurized CO_2 condition. It was found that the reaction rate between black dye molecules and Ti-OHs on TiO_2 layers was much faster in a pressurized CO_2 condition than that in a dipping condition by measuring visible spectra (615 nm) of a stained TiO_2 plate. It has been reported that TiOHs of the TiO_2 surface react with CO_2 molecules under the pressurized CO_2 condition to form Ti-O-CO-O linkages and the linkage disappears again to form Ti-OH and CO_2 at atmospheric pressure. It is reasonable to explain that the reaction between Ti-O-CO-O- and HOCO-dye molecules occurs swiftly than that between Ti-OH and HOCO-dye because of the increased negativity of the Ti atom. In other word, it is reasonable to describe that TiO_2 surface is activated under the pressurized CO_2 condition.

4. TCO-less all-metal electrode type DSC-new cell structure for electron collection [5]

Transparent conductive oxide layer-less DSCs (TCO-less DSC) have been studied [6], because TCO glass is one of the most expensive staffs. We have succeeded in preparing thick Ti layers having highly porous structures fullfilling contradictry properties of higher electron conductivities and better ion-diffusions of redox species throght the Ti electrode. Figure 3 shows a TCO-less DSC structure consisting of all metal electrodes we have reported [5]. The efficiency was 7.43% which was almost the same as that for a DSC consisting of TCO glass.

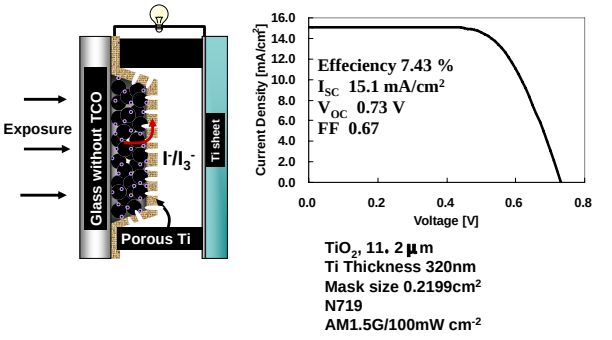

Figure 3. Photovoltaic properties for TCO-less-DSC (all metal electrode)

5. Ion-path in quasi-solid medium [7]

Solidification of electrolytes is one of crucial research items for DSCs. Solidification prevents ionic diffusions and decreases photovoltaic performances for DSCs. The decrease in the photovoltaic performances becomes serious when the content of electro-inactive solidification materials increases and cloth to all solid type DSCs. We solved the problem by fabricating ion-paths in the solidified electrolytes, where, I^-/I_3^- species are concentrated and are carried by Grötthuss mechanism. We used nano-porous Al_2O_3 films as the electrochemically inactive supporter. Figure 4 shows a DSC structure consisting of porous alumina films. In order to concentrate the redox species in the Al_2O_3 film, the surface of these alumina nanopore walls was chemically modified with imidazolium iodide moiety consisting of long alkyl groups (C12 in Figure 4). The long alkyl groups help the alignment of the imidazolium iodide molecules by self-organization mechanisms. The fact that I_2 is concentrated in alumina pore wall was observed by using adsorption experiments using Raman and UV-VIS absorption spectroscopy. Figure 4 shows photovoltaic properties for the quasi-solid DSCs consisting of the surface modified Al_2O_3 films. Jsc increased after the solidification. This is very interesting results because photovoltaic properties usually decrease after quasi-solidification.

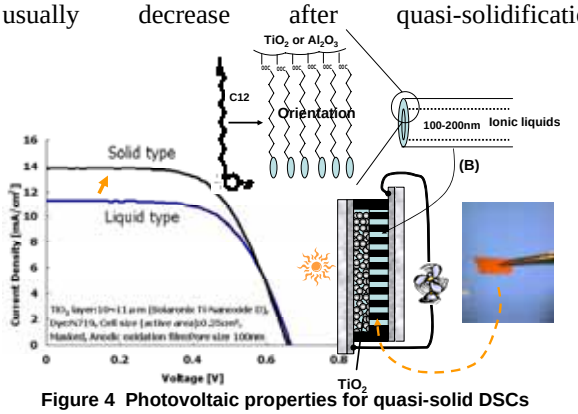

Figure 4 Photovoltaic properties for quasi-solid DSCs consisting of surface modified Al_2O_3 films

6. Conclusion

We discussed the increase in photovoltaic properties from the view point of fabricating charge carrier paths which are the most crucial item for the solar cell performance. Electron paths were effectively fabricated by passivating highly porous TiO_2 surfaces under a pressurized CO_2 condition. Ionic-paths were fabricated on the surface of porous alumina films, surfaces of which were covered with imidazolium iodide moieties consisting of long alkyl groups. This study proved that the fabrication of charge carrier paths is crucial for high efficiency dye sensitized solar cells.

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Advanced Energy Conversion Division

Clean Energy Conversion Research Section

Current status of thermal hydraulics for technological development of water-cooled in-vessel components of fusion reactor

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1. Introduction

In-vessel components such as blanket and divertor in a fusion reactor need to handle high heat and particle loads from plasma to maintain the structural soundness of the reactor. In the International Thermonuclear Test Reactor called ITER, being build by ITER Organization under the framework of collaboration of seven parties including Japan, first wall(FW) of blanket and divertor receive as high heat flux as 0.5 and 20 MW/m^2. In ITER as shown in Fig. 1, most of in-vessel components consist of blanket and divertor is located at bottom part of vacuum vessel. Design parameters of these components are summarized in Tab.1. In ITER, there are two kinds of blankets; one is a shielding blanket of which main function is 14-MeV neutron shielding caused by fusion reaction. Another is Test Blanket Module (TBM) that is a teststand for the breeding blanket being developed for fusion DEMO reactor. This report intends to briefly describe current status and outlook of thermal hydraulics for technological development of water-cooled in-vessel components in JAEA.

2. Test blanket module, TBM[1]

2.1 Outline and technological issues of TBM. TBM aims to demonstrate functions of blankets of fusion reactor in addition to the function of neutron shielding, that is, Tritium breeding used as fusion reactor fuel and power generation. This will be the first integrated test of blanket in fusion environment and each party in the ITER project is developing its original TBM concept. JAEA is pursuing to design and development technology to realize a water-cooled solid breeding TBM as shown in Fig.2. In this concept, a frame made of a Reduced Activation Ferritic Martensitic (RAFM) steels, F82H is housing a multiple layer consisted of T-breeding material, Li2TiO3, and neutron multiplier material, Be. FW has parallel rectangle cooling channels made of F82H to cool a plasma-facing surface uniformly. FW and TBM frame is integrally shaped with a hot-isostatic pressing (HIP) method. The breeding and multiplier materials are contained as small pebble beds in multiple layers separated by membrane panels with cooling tubes. Tritium bred in the pebble beds is released and collected through He purge gas flow. Since the pebble beds are expanded caused by being heated up to more than 500^{o}C due to the nuclear heat, the thermo-mechanical interactions between the pebble beds layer and the frame are necessary to be evaluated. In these analyses, thermo-mechanical properties of the pebble beds of the breeding and multiplier materials such as effective values of thermal conductivity and elastic modulus are indispensable to be modeled as a continuum treatment. In estimating efficiency of Tritium recovery from the pebble beds, modeling of heat and mass transfer phenomena inside the pebble beds are one of critical issues in the blanket system.

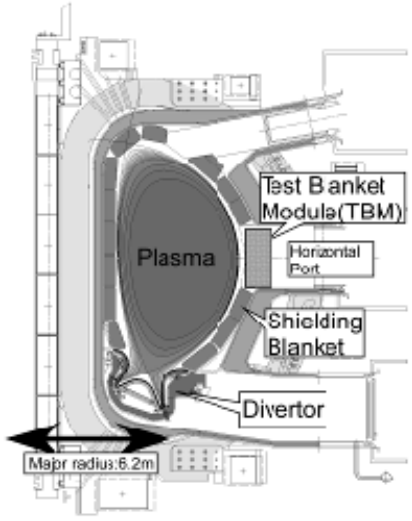

Figure 1Cross-section of ITER

From the view point of thermo-hydraulic system, one of the remarkable features of TBM is to cool the frame and FW by using high pressure and temperature water flowing. In the primary option of JAEA's design, the coolant conditions are the pressure of 15MPa and the inlet temperature of 280^{o}C as almost the same as the PWR conditions. Supercritical pressure water cooling at 25MPa and 360^{o}C are also within the scope of the feasible study for obtaining higher thermal efficiency as an aggressive option of a fusion reactor.

Table 1 Design parameters of in-vessel components

	Shielding blanket	TBM	ITER divertor	DEMO divertor
Surface heat flux	0.5～1.0 MW/m^2	0.5～1.0 MW/m^2	5-20 MW/m^2	10 MW/m^2
Neutron wall load	0.8 MW/m^2	0.8 MW/m^2	MW/m^2	
Cooling water conditions	280-325^{o}C 15MPa	100^{o}C 3MPa	100^{o}C 4MPa	200oC 4MPa
Structural material	SUS316	F82H	Cu-Alloy	F82H

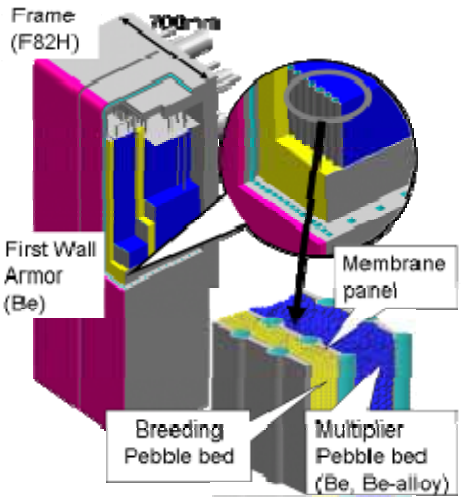

Figure 2 Concept of Solid-breeding water-cooled TBM design

Thermal-hydraulic issues in the water-cooled solid breeding blanket system are related to the follows that are needed to be simultaneously achieved; 1) To control temperatures in the pebble bed of breeding and multiplier materials and in structural material, 2)To obtain required value of tritium breeding ratio and To recover tritium bred in the pebble beds effectively, and 3)To attain the high outlet coolant temperature for high thermal efficiency.

2.2 Thermo-mechanical study on pebble beds[2]. Effective values of thermal conductivity and thermal expansion and stress-strain relationships of pebble bed regarded as a continuum filled in the layer structure inside the TBM are essential. Such data obtained so far is limited without stress loading. In TBM, however, pebble beds receive cyclic transient heat load and are under compressing load condition due to the thermal expansion of pebble itself. Figure 3 shows recent measurements of effective thermal conductivity, λ_{eff}, of the breeding material, Li_2TiO_3 as a function of strain under compressing load. With progress of compressive strain, λ_{eff} increase linearly. At the present stage, using effective thermo-mechanical data of the pebble bed accumulated so far, numerical model is being developed to estimate structural soundness of the TBM frame.

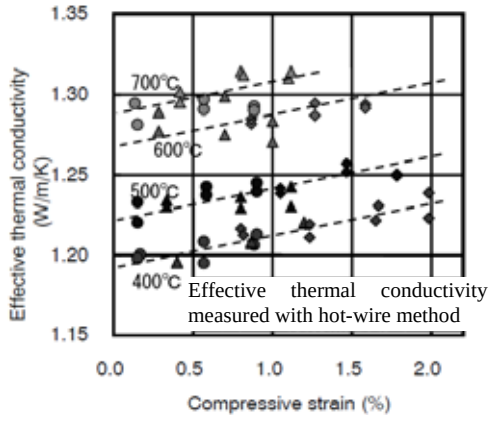

Figure 3 Effective thermal conductivity of a compressed Li2TiO3 pebble bed.

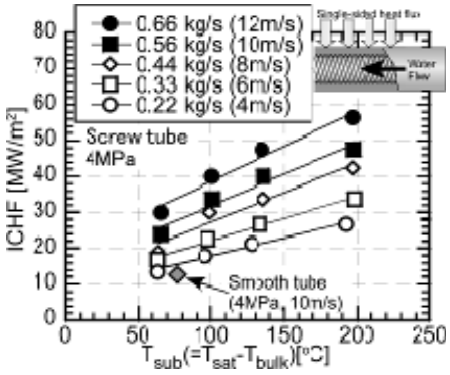

Figure 4 Effect of subcooling on ICHF of Cu screw tube

3. Divertor

3.1 Outline and technological issues of divertor

Divertor has a function to exhaust He-ash and impurity coming from plasma along the magnetic lines and receives the highest heat flux among the in-vessel components. During the transient period in starting plasma, the design value of the heat flux is 20 MW/m^2 for 10 sec in ITER. It is cooled by pressurized water at 4MPa and 100^{o}C flowing inside a swirl tube with the twisted tape inserted. The swirl tube is made of Cu-alloy. The tube transpierces armor tiles made of refractory materials like Tungsten and Carbon-fire composite to protect from the energetic particles.

Although divertor development has no program like the TBM program, JAEA is developing DEMO divertor concept and its technology. In this design, coolant is pressurized water at 4MPa as the same as ITER, but its temperature increases to 200^{o}C to suppress increasing Ductile-Brittle transition temperature of the structural material, F82H, and to improve thermal efficiency. Although the heat flux to the DEMO divertor can be assumed lower value compared with ITER, it needs to handle the steady heat flux at 10MW/m^2. Since the thermal conductivity of F82H is almost 1/10 of that of Cu-alloy, a cooling structure with higher heat removal capability is necessary in the DEMO divertor

3.2 Development of cooling structure for DEMO divertor JAEA has proposed a screw tube instead of the swirl tube. In this tube, cooling wall is threaded like a nut. This triangular fin is expected to improve heat removal caused by continuous mixing and delimitation of water near the wall. Experiments are carried out so far using a screw tube made of Cu at 4MPa and 200^{o}C. Figure 4 shows the Incident Critical Heat Flux (ICHF) of boiling water flowing inside the screw tube as a function of subcooling of water[3]. The ICHF value of the screw tube with M10 thread of 10-mm-pitch is about twice as high as that of a smooth tube. Further experiment will be need using the screw tube made of F82H to confirm its heat removal capability under DEMO-divertor-relevant condition.

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Chemical Reaction Complex Processes Research Section

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T. Sakka, Associate Professor
K. Fukami, Assistant Professor

1. Introduction

The results obtained in Chemical Reaction Complex Processes Research Section during the academic year of 2007 are described in the following sections 2 to 7. In section 2 a new method of time-resolved emission spectroscopy using acousto-optic modulator (AOM) as a switching device is proposed for the measurement of the emission from laser ablation plumes. In section 3 an attempt to obtain Zn/Cu ratio of an electrodeposited Zn-Cu film is made, based on the emission spectroscopy of laser ablation plumes. In Sections 4 and 5 the filling of porous Si with conductive polymer and noble metals are studied. In Section 6 the progress obtained on the subject of photo-assisted control of metal deposition onto Si is described. Finally in Section 7 the preparation of a Si rugate filter starting from a lightly doped Si is discussed.

2. Emission spectroscopy of laser ablation plume with time-gating by acousto-optic modulator

Laser ablation plume spectroscopy (LAPS) is a sort of the measurements generally classified to laser induced breakdown spectroscopy (LIBS) or laser induced plasma spectroscopy (LIPS). LAPS measures atomic emission spectra of the species which are laser-ablated from a solid-sample surface, and hence, can be a convenient method for surface elemental analysis. A notable feature of the method is that it can be applied even to the solid surfaces *in situ* in liquid. To obtain clear spectral lines characteristic to atoms and atomic ions for the purpose of the elemental analysis, it is important to eliminate the initial emission, namely the emission within initial several tens of nanoseconds after the laser irradiation, from the integration of the data collection.

Intensified charge coupled device (ICCD) is a useful detection system, because nanosecond gating is available, which enables to eliminate the initial part of the emission. However, the use of an ICCD system has several disadvantages. The system is large and heavy, which is unfavorable for the development of a portable system, and also the spectral sensitivity curve of the ICCD system is limited by the intensifier response curve, which is inconvenient for simultaneous measurement of many elements. Furthermore, ICCD is usually very expensive.

In the present work we propose a system using acousto-optic modulator (AOM) for the gating of the emission signal, instead of the intensifier attached to a CCD detector. This allows the system to be compact, to cover wide range of the spectrum at once, and to be inexpensive

Experimental setup is shown in Fig. 2-1. A Q-switch pulsed Nd:YAG laser beam was focused onto a target surface by a 60-mm-focal-length lens. The emission from the ablation plume was collected by an 80-mm-focal-length lens. The lens was positioned so that the plume was imaged at 860 mm after the lens. In this configuration the light to be detected converges and diverges again with an angle of ~1.5 degree. This light beam passes through an acousto-optic modulator, the operating frequency of which is 80 MHz. The Bragg angle is 0.33 degree at the wavelength of 515 nm, and hence, the separation angle between the non-diffracted or the zero-order beam and the first-order beam is 0.65 degree at this wavelength. In the experiment the first order diffraction by the Bragg configuration was introduced into an entrance slit of a 25-cm-focal-length polychromator. Although the intensifier is not necessary in this experiment, we used ICCD system as a detector, because we did not have a high-sensitivity CCD system without the intensifier. The gate by the intensifier was set to be open throughout the ablation event in the present experiments, which means that the ICCD was used solely as an alternative to a high-sensitivity CCD detector.

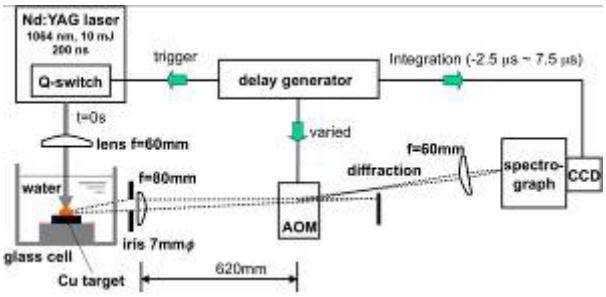

Fig. 2-1. Experimental setup for the use of AOM to gate the detection of emission spectra from the laser ablation plume produced by a solid target in water.

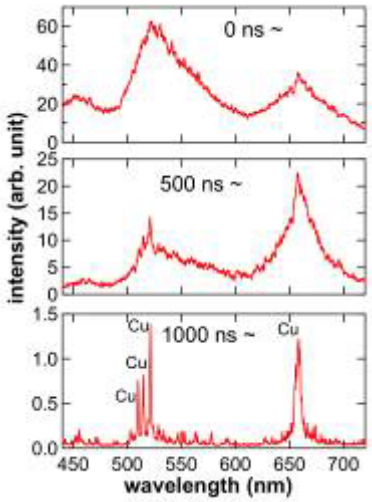

Fig. 2-2. Copper atomic line spectra obtained by the time gate using AOM. The gating time is given in each spectrum.

Fig. 2-2 shows the results of the spectral measurement performed for a Cu plate in water. Gating timing is given in each spectrum. The results show that the elimination of the continuous spectra attributed to the intense emission right after the irradiation is successful by the gating operation based on the AOM switching at 1000 ns. The guideline to obtain clear line spectra is that the emission in initial several hundreds of nanoseconds should be eliminated, although the later part should be efficiently detected. The build-up time of the diffracted beam by AOM is the order of 100 ns for a practical light beam size, although it can be less than 10 ns for very small beam sizes. The present results suggest that this build-up time is fast enough for the application to *in situ* LAPS in water.

3. Analysis of Zn/Cu ratio in an electrodeposited film by *in situ* laser ablation plume spectroscopy

The emission spectroscopy of laser ablation plume has a great potential to be applied to elemental analysis of electrode surfaces in electrochemical processes *in situ* in electrolyte solutions. However, the laser ablation plume in water, or generally in liquids, is confined in a very small volume due to the presence of a liquid, and the density is kept so high that the emission spectral lines suffer from considerable broadenings, as well as an intense continuum. Recently, we have found that the spectral feature is greatly improved by using a long nanosecond pulse for the ablation. Another problem is that pulse-to-pulse intensity fluctuation is too high to be applied to a quantitative analysis. In the present work we examined a data-analysis method taking into account the intensities of all the spectral lines assigned to each element, to minimize the data fluctuation.

The electrodeposited alloy film composed of Zn and Cu formed on a Pt plate was employed as a sample. Aqueous solution of $CuSO_4$ and $ZnSO_4$ was used for electrodeposition. A Cu wire was used as a reference electrode. The deposition was performed under a constant-potential condition. The potential was varied from –1.0 to –1.4 V vs. Cu. The laser irradiation was carried out without removing the solution after the deposition.

The peak-intensity ratio of 481 nm line assigned to Zn atoms and 515 nm line assigned to Cu atoms was obtained and plotted as a function of the deposition potential (Fig. 3-1(a)). The ratio increased with decreasing the deposition potential, as expected from the equilibrium potentials. Error bars represent standard deviations of the data, which result from the pulse-to-pulse fluctuation of the intensities. Another method of the data analysis employed in the present work uses the summation of the intensities of all the peaks assigned to each element, Zn or Cu, for the calculation of Zn/Cu ratio. The results are given in Fig. 3-1(b). The pulse-to-pulse fluctuation as given by an error bar for each point was improved compared with Fig. 3-1(a). Averaging of many lines is equivalent to averaging the population distribution of the relevant excited states, and therefore, the results become less sensitive to the temperature of the measuring region. If the pulse-to-pulse fluctuation is due to the temperature fluctuation, it is reasonable that the averaging of many lines is effective to minimize the fluctuation. We have proposed a simple model, which also supports this discussion.

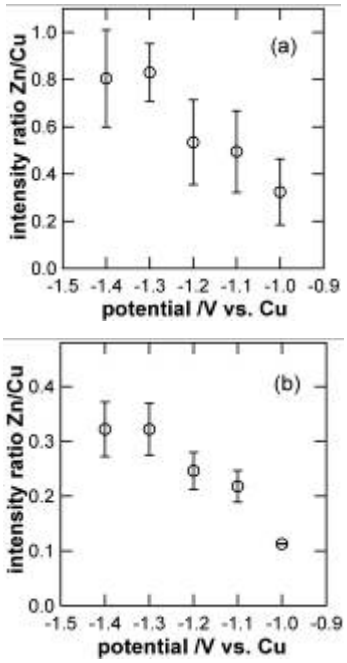

Fig. 3-1 Spectral intensity ratio obtained by (a) two peaks and (b) all the spectral lines in the spectral range of the measurement.

4. Filling of porous silicon with conductive polymer by electropolymerization

The utilization of porous silicon as template is a

promising way to fabricate microstructures. The filling of porous silicon with conductive polymers is interesting from the viewpoint of micro- and nano- actuators, reactors and electric devices. In this study, the filling of porous silicon with polypyrrole under anodic polymerization is investigated.

Porous silicon with macro-, medium-sized and micropores was formed by the anodization of Si(100) in HF solutions. The type and resistivity of the Si for the formation of macro-, medium-sized and mesopores was p-type (10-20 Ωcm), n-type (10-18 mΩcm) and p-type (4.5-6.0 mΩcm), respectively. The obtained pore diameter was 5 μm, 120 nm and 25 nm, respectively. The filling with polypyrrole was carried out at 127 μAcm^{-2}.

Figure 4-1 shows the SEM images of polypyrrole micro- and nanofibers after the removal of porous silicon template. In the case of macropore filling, each fiber has dents on its walls. This means that the electropolymerization in macropores proceeds not as continuous rod but as tubes. On the other hand, in medium-sized and mesopores, the pores are almost completely filled with polypyrrole, leading to the formation of continuous rod.

It is also well known that porous alumina and track-etched polycarbonate are utilized as template for electropolymerization because they are chemically stable during the electropolymerization. Although porous silicon has a disadvantage of surface oxidation in solutions, it has great advantages over the conventional porous templates. The present result indicates the potential of porous silicon as template for micro- and nanofabrication. The wide range of pore diameter can be formed and filled with polypyrrole. The other porous electrode does not achieve this micro- and nano- fabrication. In addition, the plugging at the pore opening by the deposited polymer itself is not observed although metal electrodeposition often shows it (see section 5). In order to understand the inhibition of plugging, the degree of pore filling should be considered. The filling factor is almost 60 % after complete filling in micropores. The rest of volume may trap electrolytic solution and ions. On the other hand, metals are always electrodeposited with 100 % filling. This difference must be the most critical factor to achieve and fail continuous filling from the pore bottom.

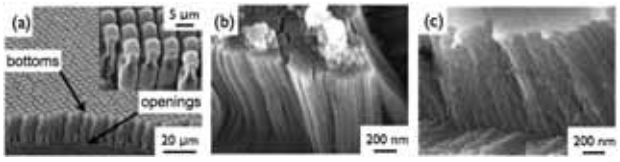

Fig. 4-1 SEM images of PPy nano- and microstructures after removal of PS. Templates used were (a) ordered macro pores, (b) medium-sized pores, and (c) meso pores.

5. Filling of porous silicon with noble metals by electrodeposition

It is of great interest to use well-ordered macroporous silicon as templates for fabricating microstructures of metal. When a silicon template is perfectly dissolved by alkaline etching solution after metal deposition, micro-metal products copying the initial template structure of macroporous Si can be obtained as shown in the section 4.

Electrodeposition of noble metals (Pt, Pd, and Au) in ordered macropores prepared in p-type silicon was investigated. Different behaviors appeared depending on the metals. Pt deposition proceeded continuously from the pore bottoms. Figure 5-1 shows ordered Pt micro-rods obtained in 4-hour electrodeposition followed by removal of the Si template. On the other hand, Pd and Au electrodeposition proceeded on the pore walls, (Figure 5-2). We tried the filling of Pd and Au under different conditions, such as compositions of solution, applied potentials, and pore depth. Though slightly different behaviors appeared depending on the conditions, complete filling of the macroporous Si was not achieved in the Pd and Au systems.

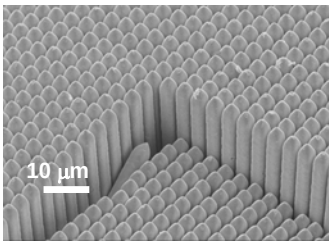

Fig. 5-1 FE-SEM image of ordered Pt micro-rods obtained by electrodeposition of Pt into macroporous Si at -0.3 V for 4 h under dark condition. The Si template was removed by chemical etching in alkaline solution.

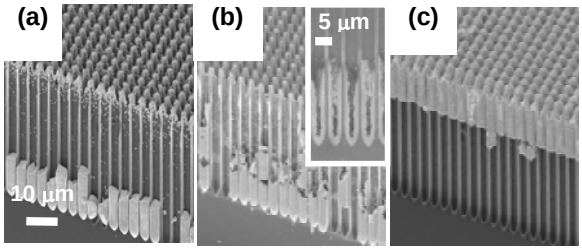

Fig. 5-2 Tilted cross-sectional SEM images of PS electrodeposited for 1 hour: (a) at -0.3 V in 0.1 M H_2PtCl_6 + 0.5 M NaCl, (b) at -0.4 V in 0.1 M $PdCl_2$ + 0.2 M HCl + 0.5 M NaCl, (c) at 0 V in 0.1 M $HAuCl_4$ + 0.5 M NaCl.

Meanwhile the investigation of immersion plating gives an important suggestion. The behaviors of immersion plating are different depending on metals. The electrodeposition mainly proceeded from nuclei formed by immersion plating in Pd and Au systems, while Pt immersion plating rate was much lower than those of Pd and Au. The nucleation of metal, which inevitably occurs before electrodeposition resulting from immersion plating, is an important factor in the noble metal electrodeposition in macroporous Si.

6. Illumination-modulated electrodeposition of less-noble metals

In order to improve electrocatalysis of semiconductor photoelectrode, surface modification like deposition of metal fine particles is required on the surface of semiconductor. An important factor in the improvement is to control the morphology of metal particles on semiconductor such as the size and distribution. Recently, we have found the different behavior of electrodeposition of noble metals such as Pt on p-type Si between in the dark and under illumination. Nucleation is predominant under illumination, while the growth of deposited nuclei is predominant in the dark during the electrodeposition. Using this behavior, we have proposed the novel method for controlling morphology of electrodeposits on p-type Si and named it "Illumination-modulated method". This year, we investigated the influence of illumination on the morphology of the electrodeposition of less-noble metals, such as Ni and Co (Fig. 6-1). In Ni and Co systems, cathodic current increased with the increase in light intensity during electrodeposition on p-type Si, as well as in the Pt and Cu systems. However, the difference of particle density obtained under illumination of different light intensity was quite low compared to the Pt and Cu systems. This fact indicates the difficulty of controlling the morphology by the modulation of illumination during electrodeposition in these less-noble metals systems. We considered the reason of the difficulty and suggested the existence of additional nucleation process, which was not expected by the conventional nucleation model. It is assumed that the attachment of ad-ions or ad-atoms on the Si surface causes the additional nucleation at the initial stage of the electrodeposition.

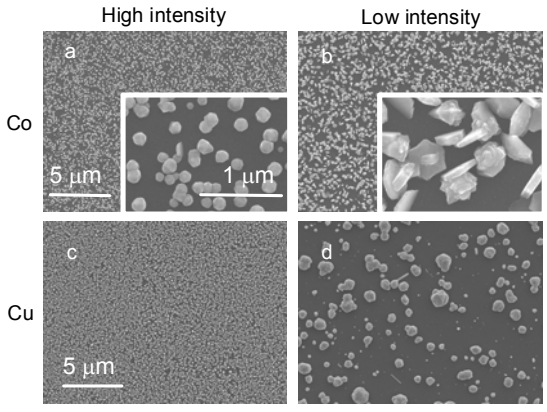

Fig. 6-1 SEM images of Co and Cu deposits on p-type Si under illumination of high and low intensities . Applied potential was −0.6 V and −0.1 V, respectively.

7. Preparation of rugate-type porous Silicon from lightly-doped Silicon

A porous Si multilayer prepared by sinusoidally modulating current density, so-called porous Si rugate filter (PSRF), can be utilized as a chemical vapor sensor. When this structure is exposed to condensable vapor, the capillary condensation takes place and the liquid displaces air in the nanometer-sized pores. This produces an increase in the average refractive index of the matrix, which results in a red shift in the spectrum. PSRF is usually prepared from a heavily doped substrate (p^+), which has a mesoporous structure. However, if a microporous structure with a narrower pore size distribution is used, we can expect a better sensitivity and response than the p^+-PSRF because of the high surface-area and effective capillary condensation ability. The use of a lightly doped substrate (p) produces the microporous structure. We prepared a p-PSRF by adjusting the period of the structure and the upper and lower refractive indices and obtained the expected performance in the sensitivity and response when sensing alcoholic vapor. However, we found some drawbacks in the p-PSRF. One is the mechanical instability due to the high porosity although the stability could be improved by the electrochemical oxidation to some extent. This prevents the layer from growing thick.

Another is a deviation of refractive index from the sinusoidal variation with depth when using a sinusoidal current density waveform. The deviation results from the difference in etching rate during the current density cycling. The p-PSRF required a larger current variation to achieve an appropriate refractive index contrast compared with the p^+-PSRF. This deformed refractive index profile affects the quality of the characteristic resonant peak of the rugate filter. A modification of the current density waveform could allow a true sinusoidal refractive index-depth profile characteristic of the filter exhibiting a low index contrast and a sharp resonant band (Fig. 7-1).

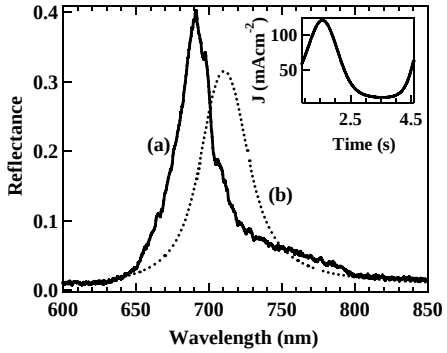

Fig. 7-1 Reflectance spectrum of a 100-cycle PSRF formed by a modified sinusoidal current density shown in the inset (a) and that simulated by assuming a sinusoidal variation of the refractive index with the depth (b).

Molecular Assemblies Design Research Section

S. Yoshikawa, Professor
T. Sagawa, Associate Professor
Y. Suzuki, Assistant Professor
Y. Kobuke, Visiting Professor

1. Introduction

A broad range of organisms have developed complex molecular machinery for the efficient conversion of sunlight to chemical energy over the past 3 billion years, which to the present day has not been matched by any man-made technologies. In this context, in order to build such kinds of efficient system mimicking living system, utilization of self-organized molecular assembly is a quite essential approach. It provides a breakthrough for new energy systems of high performance with minimum energy consumption. This research section focuses on the development of novel one- or two-dimensional nanomaterials of metal oxide and/or conductive polymer, such as nanotubes, nanowires, and/or nanosheets. Furthermore, we are developing highly efficient organic solar cells, such as organic thin-film solar cell and dye-sensitized solar cell (DSC), as well as highly active photocatalyst to realize sustainable energy systems based on next-generation solar technologies.

For the sake of remarkable enhancement of short circuit current J_{sc} and open circuit voltage V_{oc} of organic thin-film solar cell, efficient materials for electron-transporting layer (ETL), light-absorbing layer (LAL), and hole-transporting layer (HTL) were newly developed and novel construction of tandem device inserted the ultra-thin metal or metal oxide layer was performed by our research group. Particularly, supra-hierarchical nano-structured cell through the construction of completely interdigitated "supra-hierarchical nano-structure" is the feasible candidate for the breakthrough of the theoretical limit of bulk heterojunction cells, which is estimated as to less than 9% of photoelectron conversion efficiency.

2. 1D- and 2D-nanostructured conductive materials of organic and/or inorganic hybrid to improve the conversion efficiency in polymer solar cells

Much of the work on improving conjugated polymer–based photovoltaic cells over the next several years will undoubtedly involve an attempt to fabricate nanostructured materials which have the ideal geometry for exciton dissociation from the polymer. While in both components of the bulk heterojunction, carrier transport will be improved simultaneously.

Using the superior nature of TiO_2 for preventing the electron recombination and cell degradation [S.Y., *et. al*., *Appl. Phys. Lett*., **2007**, *90* (16), 163517/1-3], nano-pillar typed metal oxide nanotube array of TiO_2 has been successfully prepared as an ETL as shown in Fig.1. Such new methods of producing electron-transporting materials that are ordered on the nanometer length scale, such as the growth of metal oxide nanowires and the fabrication of well-ordered mesoporous TiO_2 and ZnO films. Particularly, ZnO is one of attractive materials, which easily form nanorod arrays, efficiently to use in hybrid device owing to its high electron transportation. We demonstrated that the charge injection efficiency of hybrid polymer/ZnO solar cell with or without N719 dye. After the modification of the ZnO nanorod arrays with N719, short-circuit current density (J_{sc}) of the hybrid solar cell was obtained as 8.89 mA cm^{-2}, which was 1.5 times higher than the case without the dye. The power conversion efficiency was enhanced from 1.16% to 2.00% by the addition of N719.

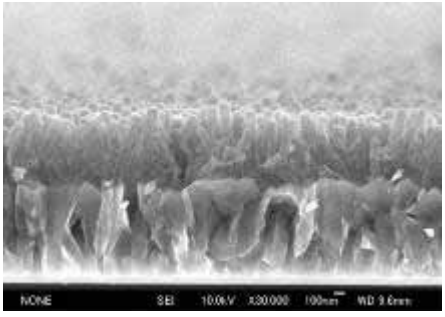

Fig. 1 SEM image of TiO_2 nanotube arrays on FTO preapared from ZnO nanorod array through template liquid phase deposition method (S.Y. *et. al*, *Chem Lett.*, **2007**, *36*, 1508-1509).

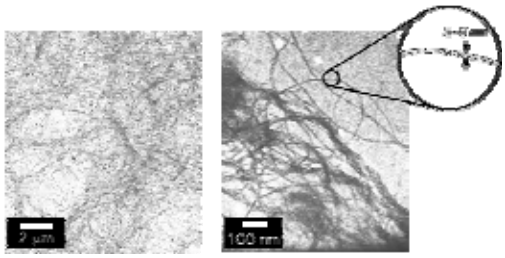

Fig. 2 TEM images of a long chain-alkylated L-glutamide-functionalized tetraphenylporphyrin derivative aggregates in the cast film prepared by 0.2 mM in a cyclohexane-THF (20 : 1) mixture. Stained by 2.0 wt% ammonium molybdate after casting and drying (T.S. *et. al*, *Tetrahedron Lett.*, in press).

Further improvement of highly oriented donor (such as porphyrin as shown in Fig. 2)-acceptor (C_{60} derivative) molecular complex as a light-absorption layer, or various kinds of oligothiophene-C_{60} as alternatives of P3HT-PCBM, and conductive polymer brushes as alternatives of PEDOT:PSS for hole-transport layer were also newly fabricated and characterized their specific photo-conductivity for the sake of remarkable enhancement of J_{sc} and open circuit voltage V_{oc} of organic thin-film solar cell through the construction of completely interdigitated "surpa-hierarchical nano-structure".

Template polymers such as poly(octadecyl acrylate)s (ODA), poly(methyl acrylate)s (MA), and poly(vinylpyridine)s (VPy) were fixed with silane coupling reagent of 3-mercaptopropyltrimethoxysilane onto the surface of quartz glass or silicon substrate through previously reported telomerization method. After the fixation of grafted polymer onto the surface of silicon, the ellipsometric thicknesses of the template layers were 7.37 nm (ODA), 5.51 nm (MA), and 3.50 nm (VPy), respectively. P3HT dissolved in chlorobenzene was spin-coated on the surface of the template-polymer-immobilized glasses and carrier (hole) mobility of thin films were characterized and evaluated through van der Pauw method in order to clarify the effect of the templates. It was found that the hole-mobility of P3HT with VPy and MA increased by 1.5 times higher than P3HT *per se* (*viz*. without the template polymer). On the other hand, remarkable enhancement of the mobility by 1 order of magnitude was observed in the case of ODA.

This research was supported by New Energy and Industrial Technology Development Organization (NEDO) from the Ministry of Economy, Trade and Industry (METI) as R&D for Next Generation PV System: Research and development of high-efficient organic thin-film solar cell with supra-hierarchical nano-structure (FY2006-FY2009) to S.Y., T.S., and Y.K.

3. TiO_2 nanofibers and Ti plates for dye-sensitized solar cells

Dye-sensitized solar cells (DSCs) by using TiO_2 nanoparticles have been studied for over a decade expected as low cost alternatives to conventional Si devices. One limiting factor in the performance of DSCs with nanoparticles is electron transport. Grain boundaries and small cross-sectional area at the interfaces of the nanoparticles lead to the enhancement of the scattering of free electrons, and such scattered electrons reduce the electron mobility and promote the electron recombination. In this context, a specific application of electrospun TiO_2 nanofibers and anodised TiO_2 plate were fabricated as electrodes of DSCs in order to improve the electron transport and enhance the light confinement leading to higher conversion efficiencies.

TiO_2 nanofiber electrodes were electrospun directly onto the fluorine-doped SnO_2 (FTO) substrate from the mixed solutions of titanium butoxide, acetylacetone, and polyvinylpyrrolidone in methanol. Calcination of these composite fibers was carried out at 450˚C. The effects of the calcination temperature on the crystalline phase, morphological appearances, and size of the as-spun TiO_2 fibers were also investigated.

The obtained TiO_2 nanofibers showed 1-D structure with diameters about 100-200 nm, which were very long over several cm. Calcination of the composite fibers at 450˚C for 3 h resulted in predominant content of anatase titania nanofibers. These nanofibers were applied to construct the DSCs with and without the combination of TiO_2 nanoparticles with N719 dye and examined the photovoltaic properties. Fig. 3 shows some examples of photocurrent-voltage characteristics and SEM images of the devices with nanoparticles/nanofibers compared to conventional DSCs with TiO_2 nanoparticles. When the TiO_2 nanofiber-electrode was used, enhanced photovoltaic performance was attained with power conversion efficiency of 8.14% compared with conventional TiO_2 nanoparticles (7.47%). The maximal power conversion efficiency was attained

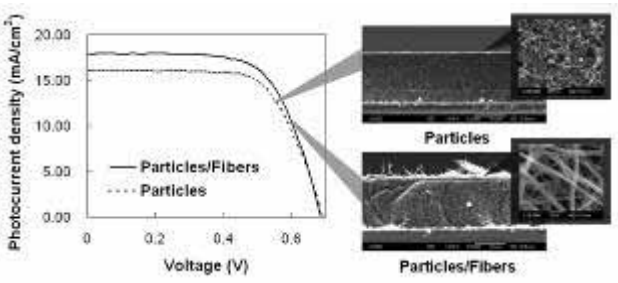

Fig. 3 I-V curves (left) and SEM images (right) of DSCs with or without TiO_2 nanofibers.

to be 10.34% (N719 dye system with 0.0515 cm^2 mask) by some optimizations.

For large-sized DSC based on FTO, metal track is an important component to retain the decrease in cell performance due to the relatively high sheet resistance of FTO. In order to minimize the internal resistance of DSC, the anodised Ti sheet was used as a support of nanocrystalline TiO_2. Although the IPCE of the cell based on Ti was lower than that of conventional FTO based one in the range from 400 to 500 nm, Ti based cell showed higher IPCE in longer wavelength region ascribed to the effective light reflection on Ti substrate. In this sense, Ti plate has superiority for producing the large sized DSC without metal track and reduce the cost of processing (S.Y. *et. al*, *Sol. Energy Mater. Sol. Cell.*, **2007**, *91*, 1176-1181).

4. Photocatalytic evolution of hydrogen by photo/bioreactor with TiO_2 nanoparticles and/or hydrogenases

TiO_2 is an n-type semiconductor and a typical photocatalyst for environmental purification. Recently, we had already reported fabrication of TiO_2/polymer films on a quartz substrate employing cationic poly(diallyldimethylammonium chloride) and anionic potassium poly(vinyl sulfate) through layer-by-layer (LBL) assemblies [T.S., *et. al*., *Chem. Lett*., **2003**, *32*, 962-963]. The transparent ultra thin films showed photocatalytic activity for reduction of methylviologen (MV^{2+}) when irradiated with UV light. This reaction system is applicable to hydrogen evolution in the presence of hydrogenase (H_2ase). As shown in Fig. 4, Sako *et. al*. (Graduate School of Agriculture, Kyoto Univ.) found three hydrogenases derived from three types of bacteria from the deep see nearby Ogasawara Islands (2000), which are extremely stable even at higher temperature over 85˚C.

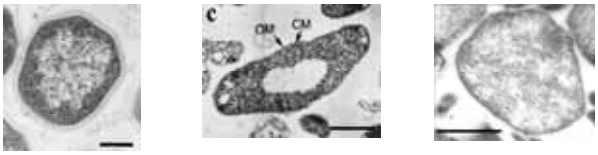

Fig. 4 Optical micrograph images of *Aeropyum camini* (left), *Hydrogenomonas thermphila* (center), and *Aeropyrum perrnix* TB5 (right).

In the present work, H_2ase/polymer thin films are newly presented by LBL method and examined their enzyme activities in joint research with Sako group. It was found that *A. camini* H_2ase-immobilized grassy carbon electrode by LBL method had remarkable durability over 50 h even at 80˚C in air and showed highest specific activity of 9.6 μmol/ml after 120 h for the evolution of hydrogen. The Mechanical Social Systems Foundation to T.S., S.Y. supported this research.

The utilization of flower-like spherical TiO_2 nanosheet as photocatalyst for H_2 evolution was also investigated. Such unique nano-structured TiO_2 was prepared by simple hydrothermal process without any surfactant or template. SEM, XRD, and N_2-sorption analyses of as-synthesized product revealed that the formation of flower-like titanate with diameter of about 250-450 nm and specific surface area of 350.7 m^2 g^{-1}. Upon calcinations at 500˚C, the titanate nanosheets were converted into anatase TiO_2 with moderate deformation of their structures. As-prapared flower-like titanate showed high photocatalytic activity for H_2 evolution from aqueous methanol solution (342 μmol/g). Moreover, the calcined sample at 500˚C exhibited higher activity (H_2 evolved at 588 μmol/g) than that of commercially available TiO_2 anatase powder (92 μmol/g with ST-01).

5. Non-equilibrium nitrogen DC-arc plasma treatment of TiO_2 nanopowder

TiO_2-based nanomaterials have attracted much attention because of their fascinating characteristics relating to renewable energy and environmental applications, such as photocatalytic hydrogen evolution and dye-sensitized solar cells. For both applications, in particular, for photocatalysts, oxygen-vacancy formation and nitrogen doping in TiO_2 have been widely studied to narrow the energy band gap (to be applicable under visible light region) or to prolong the lifetime of excited species.

In collaboration with Ecole des Mines de Paris, non-equilibrium nitrogen DC-arc plasma treatment of a commercial TiO_2 anatase nanopowder was examined to obtain nitrogen-doped TiO_2. By using a non-thermal discharge at low current (150 mA) and high voltage (1200 V) using pure N_2 gas, light yellowish-gray TiO_2 powder was successfully obtained within a short period of 5-10 min. XPS and TEM-EELS studies confirmed the existence of doped nitrogen. Due to the relatively mild conditions (plasma power of 180 W), metastable anatase structure and fine crystallite size of TiO_2 (ca. 10 nm) were maintained after the plasma treatment. The in-flight powder treatment system used in this study is promising for various type of powder treatment. This work was supported by French National Research Agency (ANR), M.I.N.E.S Carnot Institute, Japan Society for the Promotion of Science (FY2006 Scientist Exchanges Program), and MEXT, Japan (Grant-in-Aid for Science Research No. 19685020 For Young Scientist: Category A).

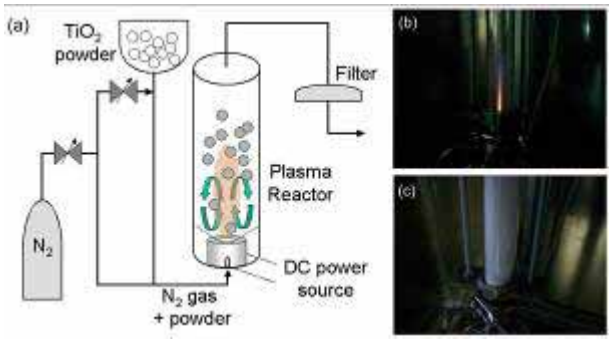

Fig. 5 Non-equilibrium nitrogen DC-arc plasma apparatus with a vertical type reactor: (a) Schematic illustration, (b) typical nitrogen plasma without powder feeding, and (c) nitrogen plasma treatment with powder feeding. (Y. Suzuki et al., *J. Nanosci. Nanotech.*, in press)

6. Synthesis and microstructure of novel TiO_2 aerogel/TiO_2 nanowire composite

Among various TiO_2-based nanomaterials, TiO_2-based aerogels and their derivatives are also expected to be excellent photocatalysts with very high surface area and characteristic mesopores. Meanwhile, TiO_2-derived one-dimensional nano-materials such as TiO_2 (or titanate) nanotubes, nanowires and nanofibers, which are prepared by hydrothermal and post-thermal treatments, have been also extensively studied due to their unique microstructure, rather high specific surface area, good crystallinity, and prospective rapid electron transfer. By combining these two types of nanomaterials, it is possible to prepare a new TiO_2-based nano-composite with homogeneous 3-D mesoporous structure reinforced with 1-D nanodispersoid. In collaboration with Ecole des Mines de Paris, TiO_2 aerogel/10 mol% TiO_2 nanowire composite was prepared by a sol-gel technique with the addition of TiO_2 nanowires into TiO_2 sol, followed by supercritical drying in CO_2. TEM analysis revealed that a unique "nanowire-network" structure was formed within mesoporous aerogel matrix. The aerogel/TiO_2 nanowire composite had relatively large surface area of 427 m^2/g with mesopores of ~16 nm in diameter and pore volume of 1.63 cm^3/g.

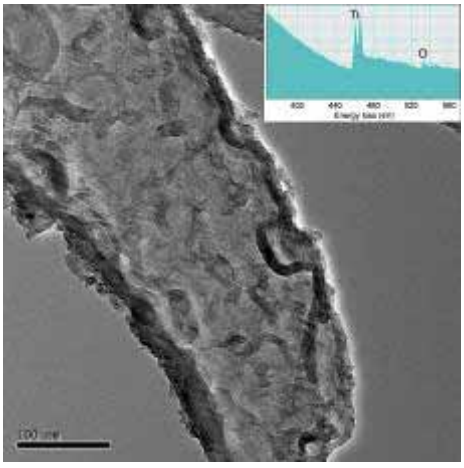

Fig. 6 TEM micrographs of a unique nanowire-network structure existed in TiO_2 aerogel/TiO_2 nanowire composite: (Insert) EELS spectrum of the composite. (Y. Suzuki et al., *Nano*, in press).

Advanced Energy Utilization Division

Biofunctional Science Research Section

T. Morii, Professor
K. Tainaka, Assistant Professor

1. Introduction

The work in our research group takes synthetic, organic chemical, biochemical and biophysical approaches to understand the biological molecular recognition and chemical reactions. Design and application of miniature proteins and functional protein/nucleic acids assemblies are explored to target and to chemically transform biologically important molecules in water, the solvent of life. Followings are main research achievments in the year of 2007.

2. Stepwise functionalization of ribonucleopeptides.

The availability of receptors with appropriate affinity and specificity to the target has been expanded by in vitro selection of RNA or DNA aptamers for targets ranging from small molecules to proteins or even cell membranes. These macromolecular receptors composed of nucleic acids are chemically modified with reporter groups to convert into fluorescent sensors. However, incorporating fluorophore-labeled nucleotides into the aptamers and chemical modifications of the aptamers with a fluorophore with suitable optical characteristics do not always guarantee successful construction of fluorescent biosensors responding at desirable wavelengths.

We have reported a strategy that enables isolation of fluorescent ribonucleopeptide (RNP) sensors with a variety of binding and signal-transducing characteristics, *i.e.*, high signal-to-noise ratios, detection wavelengths and concentration ranges for the ligand detection. The modular structure of RNP is ideal for construction of a fluorescent sensor without introducing a fluorophore-labeled nucleotide to the RNA aptamer (Figure 1).

Adenosine 5'-triphosphate (ATP)-binding RNP receptors were obtained by in vitro selection of a library of stable RNP complexes consist of HIV Rev-peptide and its target RNA sequence RRE. The RNA subunit of RNP was utilized to construct a ligand-binding cavity by in vitro selection. The RNA subunit of the ATP-binding RNP and a Rev peptide modified with a fluorophore group formed a stable fluorescent RNP complex that showed an increase in the

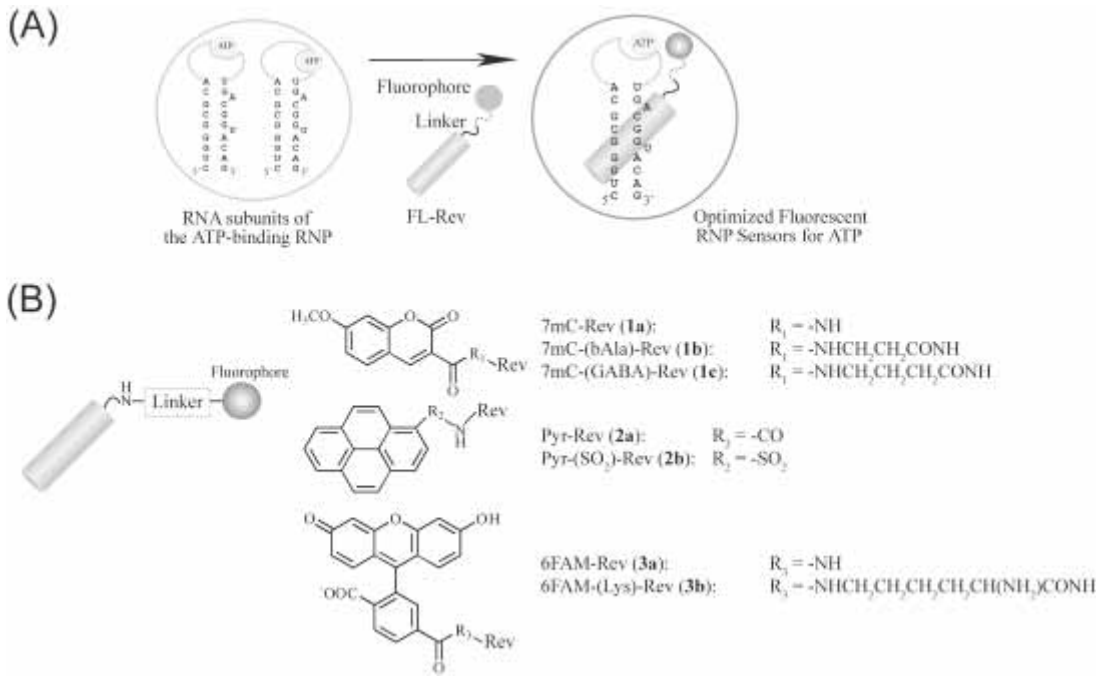

Figure 1. **(A)** A scheme illustrates a strategy to optimize fluorescent RNP sensors. Combination of the RNA subunit of the RNP receptor for ATP and a fluorophore-labeled Rev or a fluorophore- labeled Rev peptide with a linker generates combinatorial fluorescent RNP libraries, from which RNP sensors with desired optical and/or binding properties are screened. **(B)** Structures of fluorescent Rev peptides used in this study.

fluorescence intensity upon binding to ATP. Combination of a pool of RNA subunits obtained from the in vitro selection of ATP-binding RNPs and a fluorophore-modified peptide subunit afforded fluorescent RNP library, from which RNP sensors with expedient optical and binding properties were screened in a convenient manner.

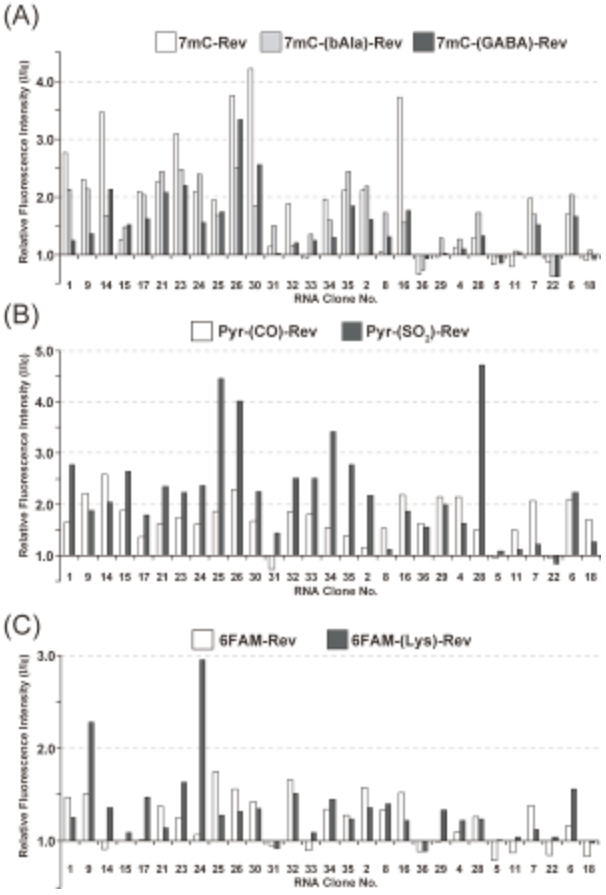

Figure 2. Relative fluorescence intensity changes (I/I_0) of RNPs upon ATP binding are shown in the bar graphs for the fluorescent RNPs derived from **(A)** 7mC-Rev (**1a**), 7mC-(bAla)-Rev (**1b**) and 7mC-(GABA)-Rev (**1c**), **(B)** Pyr-Rev (**2a**) and Pyr-(SO_2)-Rev (**2b**), **(C)** 6FAM-Rev (**3a**) and 6FAM-(Lys)-Rev (**3b**)

Fluorophores are often sensitive to a subtle microenvironmental change. By changing a tethering position of the fluorophore, or by introducing a linker with appropriate length between the amino-terminal of Rev and the fluorophore, fluorescent response of the resulting RNP sensor could be modulated. To test this, Rev derivative peptide with an additional b-alanine linker or g-aminobutyric acid linker at the amino-terminal were modified with 7-methoxycoumarin-3-carboxylic acid to afford 7mC-βAla-Rev (**1b**) and 7mC-GABA-Rev (**1c**). A derivative of Pyr-Rev was synthesized by modifying the Rev peptide with pyrene sulfonyl chloride (Pyr-(SO_2)-Rev, **2b**). A derivative of 6FAM-Rev was synthesized by an addition of a Lys residue at the *N*-terminal of Rev followed by modification with 6-FAM at the e-amino group of the Lys residue (6FAM-(Lys)-Rev, **3b**).

Each fluorophore-labeled Rev (**1a**, **1b**, **1c**, **2a**, **2b**, **3a**, and **3b**) and RNA complex was placed individually on a multi-well plate, and was evaluated by the change of fluorescence intensities in the absence or presence of ATP (0.1 mM) by using a microplate reader. Relative ratios of fluorescence intensity (I/I_0) in the absence (I_0) and the presence (I) of ATP for fluorescent RNPs with (**1a-c**), (**2a-b**), and (**3a-b**) monitored at 390, 390, and 535 nm, respectively, were summarized in Figure 2. Fluorescent RNP libraries derived from the fluorescent Rev peptide of **2b** and **2c** revealed a pattern of I/I_0 ratios different from that of the 7mC-Rev derived RNP library (Figure 2A). Fluorescent RNP of **1b** with A6, A24, A28, and A35 showed improved I/I_0 ratios. The observed I/I_0 ratios of fluorescent RNP of **2b** were much higher than that of the original fluorescent RNP derived from **2a**, as typically shown for the RNP with A02, A25, A26, A28, A34, and A35 (Figure 2B). Comparison of the observed I/I_0 ratios of fluorescent RNP derived from **3a** and **3b** revealed that **3b** afforded fluorescent RNPs with much higher I/I_0 ratios than **3a**, as typically shown for A6, A9, A17, and A24 (Figure 2C).

Even using the same fluorophore, the I/I_0 ratio of fluorescent sensor was affected by a subtle change in the attaching chemistry of the fluorophore on the peptide. Thus derivatizing the fluorophore-attaching position of the Rev peptide also increased a diversity of the fluorescent RNP pool. The degree to which the fluorescence intensity changed upon binding to ATP varied with each fluorescent RNP. These results confirmed the difficulty in predicting the efficiency of optical response of the fluorescence sensor, and demonstrate the advantage of the above strategy to obtain usable fluorescence sensors.

This research was supported by a Grant-in-aid for Scientific Research from Ministry of Education, Science, Sports and Culture, Japan to T.M (17310125, 17026019) to T.M.

3. Sensing multiple ligands at different wavelengths

The adaptability of in vitro selection to a panel of ligands is applicable to the RNP-based selection scheme. RNP receptors for GTP were isolated from the pool of RNP library by the in vitro selection method described for the selection of ATP-binding RNP. Nucleotide sequences of the RNA subunit of GTP-binding RNPs revealed that there are consensus 5'-GCGG-3' and 5'-UGUCUAC-3' sequences (Figure 3).

As with the case for the ATP-binding RNP, combination of the RNA subunit pool of the GTP-binding RNP and several fluorophore-labeled Rev peptide subunits can form combinatorial fluorescent RNP receptor libraries. The fluorescence intensities in the absence and presence of GTP were assessed in the similar manner for fluorescent RNP libraries obtained by combination of the RNA subunits of the ATP-binding RNP and the fluorophore-labeled Rev peptides. A series of fluorescent GTP sensors with I/I_0 ratios of over 2 were obtained from the fluorescent RNP libraries derived from the RNA subunit of GTP-binding RNP and 7mC-Rev, Pyr-Rev or NBD-Rev (Figure 4). In the case of 7mC-Rev derived GTP sensor library, G23/7mC-Rev, G26/7mC-Rev, G05/7mC-Rev, and G14/7mC-Rev had high I/I_0 ratios of

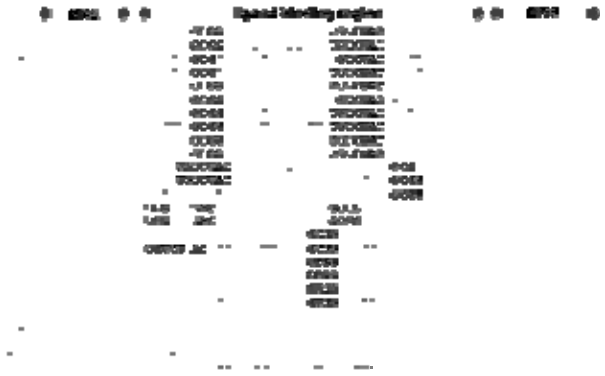

Figure 3. Nucleotide sequences of the ligand-binding region for the RNA subunit of RNP selected against GTP revealed consensus nucleotide sequences (shown in bold). RRE denotes the Rev-binding RNA sequence.

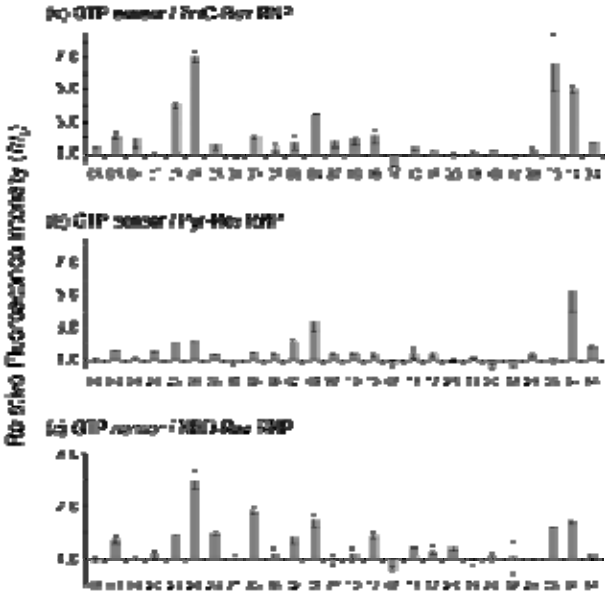

Figure 4. Relative fluorescence intensities (I/I_0) of fluorescent GTP-binding RNPs upon GTP binding are shown in the bar graphs: (a) 7mC-Rev RNP, (b) Pyr-Rev RNP, (c) NBD-Rev RNP.

over 4. As observed for the screening of the ATP sensor libraries, the observed I/I_0 ratios of GTP sensors depended on the nature of fluorophore even using the same RNA subunit. For example, The I/I_0 ratio of G05/7mC-Rev showed almost 6 (Figure 4, panel a), whereas that of G05/Pyr-Rev was measured to be 1 (Figure 4, panel b).

These fluorescent GTP-binding RNPs showed distinct selectivity to GTP over other NTPs. With ATP and GTP sensors emitting at different wavelengths, concentrations of ATP and GTP can be monitored independently in the same solution.

The ATP sensor A32/Cy5-Rev could monitor ATP from a submicromolar to millimolar concentration range at 670 nm. Even when the ATP titration was carried out in the presence of GTP (500 μM), the fluorescence intensity of A32/Cy5-Rev increased with the affinity similar to that in the absence of GTP. Increasing the concentration of ATP did not affect the fluorescence intensity of the GTP sensor G33/NBD-Rev. G33/NBD-Rev showed constant fluorescence intensity in solutions containing 500 μM GTP and increasing concentrations of ATP. The GTP sensor G33/NBD-Rev also responded to GTP from a submicromolar to millimolar concentration range at 535 nm in the absence and presence of ATP (500 μM). The affinity of the GTP sensor G33/NBD-Rev to GTP in the presence of 500 μM ATP was almost equal to that in the absence of ATP.

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4. Fluorescent RNP sensors for biologically important targets.

Protein phosphorylation at the tyrosine residue controls many cellular signaling events. Protein tyrosine kinases, protein tyrosine phosphatases, and their substrates play an important role in regulating processes such as proliferation, differentiation, motility, and immune responses, as well as pathological conditions such as cancer. Methods for the selective recognition and sensing of protein tyrosine phosphorylation are quite important in biochemical, proteomic, and cell biological studies. However, current analytical methods of protein tyrosine phosphorylation generally need multiple laborious steps. Typical methods utilize detection by autoradiography of ^{32}P-radiolabeled proteins separated by SDS-PAGE, and immunoblotting or immunoprecipitation by anti-phosphotyrosine antibody. Antibody detection is an especially sensitive method (fmol level) which is highly suitable for the detection of specific tyrosine phosphorylated residues in individual phosphoproteins. Mass spectral analysis, of peptide fragments obtained by utilizing anti-phosphotyrosine antibody has advantages of being rapid and does not require radiolabelling, which unveiled temporal interaction of many signaling proteins. Although these methods were powerful and exploited our understanding of cellular signaling, it is difficult to obtain quantitative interpretation due to the washing steps involved in the analytical procedures. There still exists a demand for direct detection methods of tyrosine-phosphorylated proteins, such as using artificial biosensors.

RNP-based fluorescent probes would be ideal to detect protein tyrosine phosphorylation directly in the solution. In order to develop a fluorescent RNP sensor for the pTyr residue, RNP receptors has been developed for the pTyr residue.57 RNP receptors for pTyr were isolated from a pool of RNA sequences (430) as described for the ATP-binding RNP receptors. Analysis of the nucleotide sequences of clones revealed distinct consensus sequences. Among the 29 clones, nine revealed the same nucleotide sequence pY03. A 10-nucleotides consensus sequence 5'-UGC---GGUAGAA-3' was thoroughly observed for other clones. Equilibrium dissociation constant for the complex of pY03RNA and the Rev peptide (pY03RNP) to pTyr were determined from the saturation curves to be 376 μM. The RNP complex pY03RNP preferentially bound

pTyr over O-phospho-L-serine (pSer), Gly-Tyr, or Leu-pTyr. The phosphate charge of pTyr contributed to the specific binding complex formation of pY03RNP as the non-phosphorylated dipeptide Gly-Tyr did not bind pY03RNP. Because pSer did not bind pY03RNP, the aromatic ring of pTyr is also a key determinant for the selective binding of pY03RNP. A pTyr-containing dipeptide Gly-pTyr bound pY03RNP with a similar affinity to pTyr. Interestingly, a pTyr-containing dipeptide Leu-pTyr showed almost no affinity to pY03RNP. It is likely that the steric hindrance at the N-terminal portion of pTyr group strongly prohibits the binding of pTyr group to the binding pocket of pY03RNP. It would be possible to isolate RNP receptors that can recognize not only a pTyr residue, but also amino acid residues surrounding the pTyr. By increasing the recognition surface, it would be possible to increase the affinity of RNP to pTyr-containing amino acid sequences. By using the RNP-based modular strategy for the fluorescent ATP sensors, it will be possible to obtain fluorescent pTyr biosensors from the pTyr-binding RNP receptors to detect a pTyr residue within a defined amino acid sequence on the protein surface.

The combinatorial strategy using the modular RNP receptor described here enables efficient tailoring of fluorescent sensors for specific ligands, ATP, GTP, phosphotyrosine, and biogenic amines. The approach consisting of a selection followed by a screening, namely, in vitro selection of RNP receptors and an efficient screening of the combinatorial RNP sensor library, affords fluorescent biosensors with a series of emission wavelengths, high I/I_0 ratios, and/or responding ligand concentration ranges without a chemical modification of the ligand binding RNA region and without detailed knowledge of the three dimensional structure for the RNP receptors. By choosing the fluorescent RNP sensors with appropriate optical characteristics, sensitive detections of multiple ligands without inhibitory effects of intrinsic fluorescence in the samples such as cellular extracts would be possible. Combination of the fluorescent RNP sensors offers a fluorescent sensing system that detects a wide range of ligand concentrations. Fluorescent sensor systems with wide detection concentration ranges were previously achieved by manipulating the ligand-binding affinity of the parent receptor by means of site-directed mutations that needed detailed knowledge of three-dimensional structural information. In contrast, in vitro selection of the RNA-based RNP library provides RNP receptors with various affinities to the target ligands. Such an inherent diversity of the ligand-binding affinity is a definitive advantage for RNP to construct a fluorescent sensor. The non-covalent modular structure of RNP provides a distinct advantage for the screening strategy of fluorescent sensors described here. Once an optimal fluorescent RNP sensor is constructed, the non-covalent complex of RNP sensor could be covalently linked by tethering between the RNA and the fluorophore-labeled peptide subunits. Such a stable RNP sensor can be immobilized by hybridization of the attached RNA sequences to complementary surface-bound DNA probes to organize an RNP sensor microarray that is useful for analyzing biologically active molecules simultaneously.

This research was supported by a Grant-in-aid for Scientific Research from Ministry of Education, Science, Sports and Culture, Japan to T.M (17310125, 19021023) to T.M.

5. Role of tyrosine phosphorylation for the amyloid fibrillization of human tau core peptide.

Phosphorylation/dephosphorylation of the fibrillogenic protein, human tau, is believed to play crucial roles in pathogenesis of the Alzheimer's disease. We have studied the phosphorylation at Tyr310 of tau in the fibril-forming core domain. Previous studies have demonstrated that Tyr310 is essential for the tau fibrillization and a plausible *in vivo* phosphorylation site. For elucidating the events in molecular details, we synthesized a short peptide, $VQIVY_{310}K$, derived from a partial sequence surrounding Tyr310. This peptide forms amyloid-type fibrils similar to those generated by full-length human tau. The Tyr residue in VQIVYK was substituted with phosphorylated tyrosine and a series of non-natural aromatic amino acid residues to elucidate mechanisms by which the Tyr phosphorylation alters the fibril-forming propensity. We found that the phosphorylation strongly stabilized the VQIVYK fibrils and enhanced their secondary lateral segregation. The results have proposed that an alteration in the charged state of peptide by the phosphorylation plays a dominant role in the aggregation enhancement. To confirm whether the electrostatic pairing of charged amino acid residues could enhance the fibril formation of peptides, we performed experiments using a Lys to Glu mutated peptide VQIVYE and its phosphorylated derivative, and found that the phosphorylation of VQIVYE clearly suppressed the fibril formation. Kinetic properties of the peptide aggregations were also studied by a time-resolved method, which found that the phosphorylation and its charge-altering effect substantially affected the fibril formation process. The phosphorylation effects observed for both VQIVYK and VQIVYE were interpreted in a unified view, which concluded that the electrostatic nature of the phosphate group and its interactions with the neighboring charged residue of the peptides strongly alter their amyloid formation properties.

This research was supported by a Grant-in-aid for Scientific Research from Ministry of Education, Science, Sports and Culture, Japan to T.M.(17310125, 19021023) to T.M.

Bioenergy Research Section

K. Makino, Professor
T. Kodaki, Associate Professor

1. Introduction

Our research section seeks to develop environmentally clean and efficient reaction systems by means of chemically or biologically manipulated systems suitable for energy production. For the development of such a process by learning from biological systems, it is essential to understand complex network of biological signal transductions and mechanism of chemical transformations in the system. Following aspects have been investigated to establish the fundamental basis that would emerge a new technology for the energy-efficient utilization of ubiquitous environmental resources.

2. Development of stable DNA microarray platforms suitable for quantitative analysis

DNA microarrays are essential tools for studying gene expression, disease diagnosis and drug discovery, and detection of single nucleotide polymorphisms. In general, DNA microarrays are fabricated by mechanical deposition of the pre-synthesized oligonucleotides on a chemically modified glass slides using automated robotic systems. Glass slides modified with active organic functional groups such as primary amino groups are particularly advantageous for immobilizing probe DNA by several kinds of immobilization chemistries. In most of the cases, amine-modified glass slides are prepared by the self assembly of aminoalkoxysilanes on hydroxylated glass slides. It is, however, reported that because of steric hindrances a significant number of silanol groups are left unreacted during the self assembly process which can induce non-specific interactions with the introduced amino groups on the surface. In addition, these remaining silanol groups may also catalyze the hydrolysis of silylether bonds between the glass slides and the linker and there the capture DNA probe is detached from the array surface. In addition, usage of homo/ heterobifunctional linkers for efficient immobilization of oligonucleotides probes increases the manufacturing steps of DNA microarrays.

In the post genomic era, new approaches to DNA microarray fabrication are very much required to address some of these challenges that are beyond the present day technologies. In this study, we put forward a new strategy of additional surface modification of glass slides and application of functional oligonucleotide probe for fabricating DNA microarrays that could be useful for sensitive and selective detection for conducting quantitative analysis. Microarray substrates were prepared by chemical modification of glass slides with aminopropyltriethoxysilane and butyltrimethoxysilane as described elsewhere. Functional oligonucleotide probes were synthesized by incorporating DMT-dOxo-phosphoramidite to a growing chain of oligonucleotide at 5′-end on DNA synthesizer using phosphoramidite chemistry. These oxanine modified oligonucleotides (Oxa-ODN) were subsequently purified by using reverse phase HPLC.

Then Oxa-ODNs were spotted on the prepared glass slides with an inkjet spotter (Figure 1). At first, fluorescent intensity was normalized on the basis of initial fluorescent intensity measured by hybridizing a part of the samples with Cy3-labelled complementary targets on 0 days. Subsequently, the remaining sets of DNA spotted glass slides exposing to 42°C and 50% humid conditions for 7 days and their respective fluorescent intensity was again measured by hybridizing with Cy3-labelled complimentary targets.

As shown in Figure 2, the stability of the array surface for the immobilized Oxa-DNA probes was assessed by comparing the fluorescent signal intensity of the hybrids formed on the array surface. It was observed that aminopropyltriethoxysilane modified glass slides (APS; Control) without capping treatment and butyltrimethoxysilane capped APS glass slides (APS+BMS) showed decrease in hybridization fluorescent intensity. However, the amount of decrease in hybridization signal intensity from APS+BMS glass slides was comparatively less compared to control. These results suggest that APS+BMS glass slides provide stable surface that can reduce the hydrolysis of silyl ether bonds between the glass slides and silane linkers and thereby increasing the life time of the probes on the array surface. On the other hand, the application of dOxo modified oligonucleotide on the array surface through the functional nucleobase of dOxo eliminated the need for the use of bifunctional linkers making the present system practically convenient for DNA

microarray fabrication.

In this study, amine-modified glass slides additionally modified with butyltrimethoxy silane were prepared and spotted with Oxa-ODN probes. It was observed that APS+BMS glass slides showed improved stability of the silylether bonds. Furthermore, the application of Oxa-ODN probes would be practically convenient for fabricating DNA microarrays. Therefore, we infer that, the present additional surface modification and Oxa-ODN probes could be useful for fabricating reliable and efficient DNA microarray.

3. Chemical natures of 6-formylpterin nucleoside analogs

6-Formylpterin (6FP, Scheme 1) is a pteridine

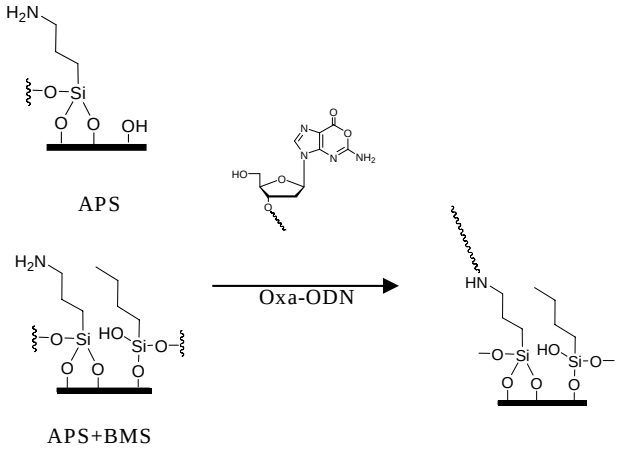


Figure 1. Schematic illustration of Oxa-ODN immobilization on APS and APS+BMS glass slides using inkjet spotter.

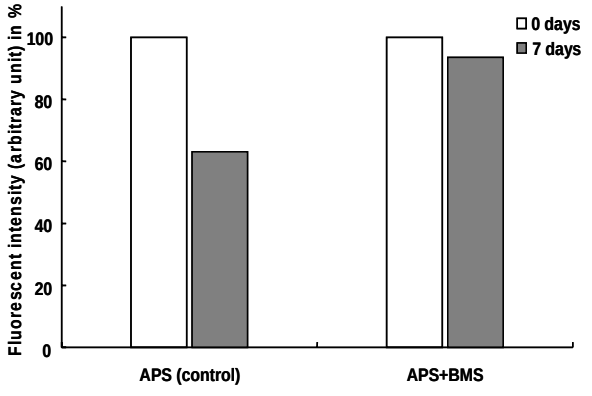


Figure 2. Fluorescence intensity of Cy3-labelled complementary targets hybridized with probe DNA on array surface at 0 days (white bar) and after 7 days exposure to 42°C temperature and 50% humidity (grey bar).

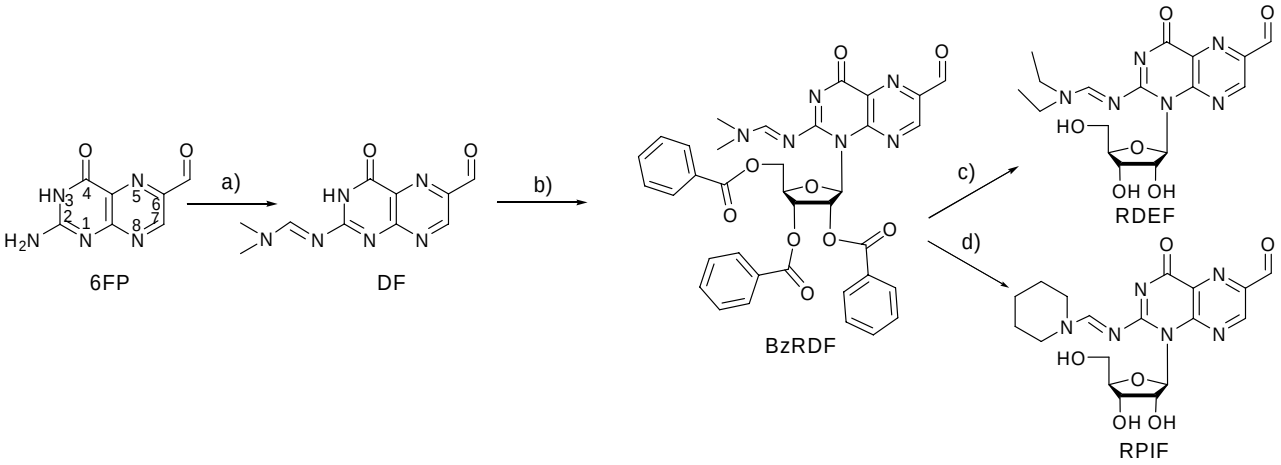
derivative produced from folic acid by photooxidation in the presence of O_2, and *in vivo* from folic acid in some pathological conditions, such as carcinoma. In previous studies, we have shown that reactive oxygen species (ROS) such as H_2O_2 generated by 6FP, induced apoptosis in HL-60 cells, inhibited Fas-mediated apoptosis in Jurkat cells, and suppressed activation of NF-κB, cytokine production, and cell proliferation in PanC-1 and human blood T cells. In addition, 6FP showed neuroprotective effects against transient ischemia-reperfusion injury (IRI) in gerbils and similar effects for retinal IRI in rats; however, the mechanism of such physiological activities has not been elucidated until date. Although 6FP showed notable physiological activities, it has not been used clinically because of its poor watersolubility at neutral pH. In the present study, to overcome this drawback, nucleoside analogs of 6FP, which improved water solubility at neutral pH have been developed and assessed for their ROS generation activities in the presence of NADH in the dark. Since one of the ubiquitously occurring electron sources in living organisms is NADH, we employed it in the present study as an electron source. It is important to explore ROS generation by 6FP derivatives in the absence of light, because most biological events occur in the dark. As reported previously, it is now widely known that ROS is not only involved in cell death but is also involved in the modulation of a variety of cell functions. Therefore, it is important to understand the relationship between ROS activity and biological events. 6FP derivatives, which can generate ROS through NADH oxidation in the dark at neutral pH without any biological systems, can be potent physiological compounds that generate appropriate amounts of ROS in living systems. In addition, it is assumed that by using these compounds the above-mentioned physiological activities of 6FP can be introduced in most biological systems.

When 6FP reacted with *tert*-butoxybis(dimethylamino)methane in *N*,*N*-dimethylformamide (DMF) at 60 ºC for 20 min, 2-(*N*,*N*-dimethylaminomethyleneamino)-6-formylpteridin-4-one (DF) was obtained in a yield of 95%, as shown in Scheme 1. Then the glycosylation reaction of DF was conducted. When DF reacted with 1-*O*-acetyl-2,3,5-tri-*O*-benzoyl-β-D-ribofuranose in the presence of 1,8-diazabicyclo[5,4,0]undec-7-ene and trimethylsilyl trifluoromethanesulfonate in acetonitrile at 60 ºC for 14 h, glycosylation reaction occurred at the 1-position of DF and1-(2,3,5-tri-*O*-benzoyl-β-D-ribofuranosyl)-2-(*N*,*N*-dimethylaminomethyleneamino)-6-formylpteridin-4-one (BzRDF) was obtained in a yield of 68% (Scheme 1).

Next, the deprotection reaction of BzRDF was carried out. When BzRDF was stirred in methanol containing 25% diethylamine solution at room temperature for 24h, 1-(β-D-ribofuranosyl)-2-(*N*,*N*-diethylaminomethylenea mino)-6-formylpteridin-4-one (RDEF) was obtained in a yield of 49% as a nucleoside analog of 6FP derivative (Scheme 1). RDEF shows over 100 mM solubility in PBS at pH 7.4 because of the hydrophilic ribose moiety. In this reaction, the dimethylaminomethyleneamino group on the 2-position was substituted to *N*,*N*-diethylaminomethyleneamino. To determine if this occurred generally, BzRDF was stirred in methanol containing 25% piperidine solution at room temperature for 24 h, and it was found that 1-(β-D-ribofuranosyl)-2-(piperidin-1-ylmethyleneamino)-6-formylpteridin-4-one (RPIF) was produced in a yield of 25% (Scheme 1).

To investigate ROS generation in the reaction of RDEF and NADH, electron paramagnetic resonance (EPR) spectroscopy coupled with the spin trapping technique, using 5-(2,2-dimethyl-1,3-propoxy cyclophosphoryl)-5-methyl-1-pyrroline *N*-oxide (CYPMPO) as a spin trap was employed. PBS solutions (pH 7.4) containing 4 mM RDEF, 4 mM NADH, and 20 mM CYPMPO were incubated at room temperature for 1 h in the dark in an open system and measured using EPR spectroscopy. The results are summarized in Figure 3. The solutions containing either RDEF or NADH and CYPMPO did not show EPR signals. The solutions containing RDEF, NADH, and CYPMPO showed a prominent EPR spectrum pattern of the mixture of $^{\cdot}O_2^-$ and $^{\cdot}OH$ adduct of CYPMPO (Figure 3(a)). These signals were completely quenched by superoxide dismutase (SOD) which converts $^{\cdot}O_2^-$ to H_2O_2 in the presence of a proton source (Figure 3(b)). Figure 3(c) is a computer simulated spectrum for the mixture of $^{\cdot}O_2^-$ and $^{\cdot}OH$ adduct of CYPMPO. Both $^{\cdot}O_2^-$ and $^{\cdot}OH$ adducts are composed of two diastereomers respectively. The computer-simulated spectrum showed good coincidence with the previously reported spectrum of the mixture of $^{\cdot}O_2^-$ and $^{\cdot}OH$ adduct of CYPMPO, and also with the spectrum in Figure 3(a) in this study. These results clearly indicate that O_2 was converted into $^{\cdot}O_2^-$ in the presence of RDEF and NADH.

We synthesized novel nucleoside analogs of 6FP. RDEF, which has a ribose on the 1-position of the pteridin ring, showed $^{\cdot}O_2^-$ generation activity in the presence of NADH in the dark at pH 7.4 without any biological systems.



Scheme 1. Chemical synthesis of RDEF and RPIF: a) Synthesis of DF from 6FP using *tert*-butoxybis(dimethylamino)methane in DMF at 60 ºC for 20 min, 95%; b) Glycosylation of DF using 1,8-diazabicyclo[5,4,0]undec-7-ene, trimethylsilyl trifluoromethanesulfonate, and 1-*O*-acetyl-2,3,5-tri-*O*-benzoyl-β-D-ribofuranose in CH_3CN at 60ºC for 14 h, 68%; c) Deprotection of BzRDF in MeOH containing 25% diethylamine at rt. for 24 h, 49%; d) Deprotection of BzRDF in MeOH containing 25% piperidine at rt. for 24 h, 25%.

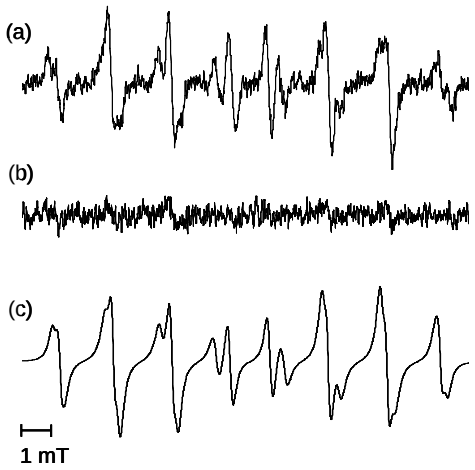


Figure 3. EPR spectra obtained for PBS (pH 7.4) containing (a) 4 mM RDEF, 4 mM NADH and 20 mM CYPMPO, (b) 4 mM RDEF, 4 mM NADH, 20 mM CYPMPO and 2,000 units/ml SOD. The reactions were performed for 1 h at room temperature in the dark in an open system and measured by using EPR spectroscopy. (c) Computer-simulated spectrum for the mixture of $^{\cdot}O_2^-$ and $^{\cdot}OH$ adduct of CYPMPO.

4. Efficient Bioethanol Production from Woody Biomass by Yeast Transformed with Protein Engineered Enzyme

Xylose is one of the major fermentable sugars present in lignocellulosic biomass, the second most abundant carbohydrate polymer in nature. *Saccharomyces cerevisiae* is used widely for industrial ethanol production because of its ability to produce high concentrations of ethanol and high inherent tolerance. Since the native *S. cerevisiae* cannot ferment xylose, molecular engineering of *S. cerevisiae* for xylose utilization has focused on adapting the xylose metabolic pathway from xylose-utilizing yeast such as *Pichia stipitis*. *S. cerevisiae* transformed with the native genes encoding xylose reductase (XR) and xylitol dehydrogenase (XDH) from *P. stipitis* ferments xylose to ethanol, but has not yet been applied to the industrial bio-process due to the unfavorable excretion of xylitol. Intercellular redox imbalance caused by the different coenzyme specificities of XR (using NADPH/NADH with preference for NADPH) and XDH (exclusively using NAD^+) has been considered one of the main factors for xylitol excretion. To address this issue, we first performed site-directed mutagenesis for complete reversal of coenzyme specificity on NAD^+-dependent XDH from *P. stipitis* to generate $NADP^+$-dependent XDH. Asp207, IIe208, Phe209, and Asn211 were selected as amino acid residues to attempt the switch of coenzyme specificity from NAD^+ to $NADP^+$. These mutated XDHs were characterized *in vitro*. The triple mutant (D207A/I208R/F209S), named ARS, and quadruple mutant (D207A/I208R/F209S/N211R), named ARSdR, showed over 4,500-fold higher values of *k*cat/*K*m with $NADP^+$ than the wild-type enzyme, reaching values comparable to *k*cat/*K*m with NAD^+ of the wild-type enzyme. Furthermore, we attempted to improve the thermostability of XDH mutants by introduction of an additional zinc atom. The mutated XDH introduced three cysteine residues, named C4 mutant, on acquiring an additional zinc and had higher thermostability. Then, we constructed recombinant yeasts that were transformed with the genes encoding the wild-type XR and the protein engineered XDH of *P. stipitis*. These recombinant yeasts were characterized based on the enzyme activity and fermentation ability of xylose to ethanol. The recombinant *S. cerevisiae* was shown to effectively express a native XR and a protein engineered $NADP^+$-dependent XDH from *P. stipitis*. In a comparison of fermentation performance of xylose to ethanol with the reference strain, the recombinant yeast gave enhanced ethanol production (Figure 4) and also decreased xylitol excretion. The more improvement of XDH should be attempted for the effective production of ethanol. Introduction of the improved XDH with some other enzyme involved in xylose metabolism to the wild type *S. cerevisiae* is expected for the construction of the effective xylose fermenting yeast.

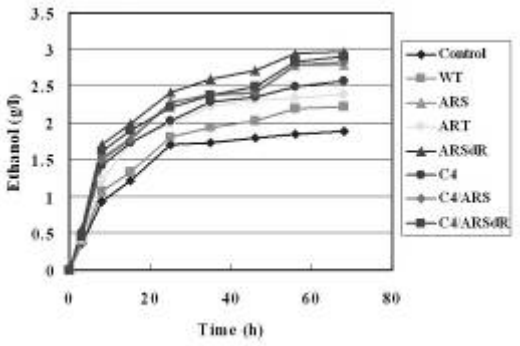


Figure 4. Ethanol fermentation of recombinant *S. cerevisiae* transformed with protein engineered XDH.

These works were supported by a Grant-in-Aid for Scientific Research, and "Kyoto Nanotechnology Cluster Project", a Grants for Regional Science and Technology Promotion from the Ministry of Education, Science, Sports and Culture, Japan. These works were also supported by CREST of the Japan Science and Technology Agency.

Laboratory for Complex Energy Processes Section

M. Kinoshita, Professor
H. Okada, Associate Professor

1. Introduction

A. Theoretical Biophysics

A variety of self-assembling and ordering processes in biological systems, which occur at molecular levels, are sustaining life. Examples of such processes are protein folding leading to a unique tertiary structure, protein traffic, molecular recognition, aggregation of protein molecules forming ordered and often symmetrical quarterly structure, and lipid-membrane formation. Biopolymers, a great diversity of molecular and ionic species, or water is simply *material* when each of them is separately present. However, the complicated correlations among these material constituents can lead to *life*. We are elucidating those correlations, uncovering the mechanism of the biological self-assembly, and clarifying the roles of water by developing special theories based on statistical mechanics and morphometric thermodynamics. The achievements will provide important bases of life science, biotechnology, and nanotechnology. The current subjects are investigations on (A1) the hydrophobic effect, (A2) molecular mechanisms of folding and unfolding (denaturation) of proteins, (A3) molecular recognition, (A4) prediction of the native structure of a protein, and (A5) mechanism of amyloid-fibril formation. Here we briefly describe several developments made in (A1), (A2), and (A4).

B. Plasma Physics

The major subjects concerned with plasma physics in this section are to study fast-ion confinement in plasma confinement devices and to investigate interactions between fast-ions and materials, such as a first wall and a vacuum vessel. The fast-ion confinement is a critical issue for the fusion reactor since the alpha particles produced in the D-T reaction should be utilized to heat plasma efficiently. The interactions between fast-ions and materials cause the impurity problem for the plasma confinement and the damage for the vessel or the first wall materials.

Fast ion velocity distribution is investigated using ICRF minority heating in Heliotron J with special emphasis on the effect of the toroidal ripple of magnetic field strength. Pitch angle distribution of fast ions is observed experimentally and Monte Carlo analysis is performed. Pitch angle dependence of fast ions from the calculation agrees with the experimental results.

(A1) Molecular Origin of the Hydrophobic Effect

The hydrophobic effect plays crucially important roles in a variety of phenomena in aqueous environments such as micelle formation, protein folding and aggregation, lipid membrane formation, and molecular recognition. The elucidation of the molecular origin of the hydrophobic effect is imperative for understanding the mechanism of those phenomena. Even for a small nonpolar solute, however, there are still lots of controversial aspects and many of the issues concerning the hydrophobic effect remain unresolved.

We have investigated the molecular origin of the hydrophobic effect using the angle-dependent integral equation theory combined with the multipolar water model [1]. The thermodynamic quantities of solvation (excess quantities) of a nonpolar solute are decomposed into the translational and orientational contributions. The translational contributions are substantially larger with the result that the temperature dependence of the solute solubility, for example, can well be reproduced by a model simple fluid where the particles interact through strongly attractive potential like water and the particle size is as small as that of water. The thermodynamic quantities of solvation for carbon tetrachloride, whose molecular size is ~1.9 times larger than that of water, are roughly an order of magnitude smaller than those for water and extremely insensitive to the strength of solvent-solvent attractive interaction and the temperature. The orientational contributions to the solvation energy and entropy are further decomposed into the solute-water pair correlation terms and the solute-water-water triplet and higher-order correlation terms. It is argued that the formation of highly ordered structure arising from the enhanced hydrogen bonding does not occur in the vicinity of the solute. Our proposition is that the hydrophobic effect is ascribed to the interplay of the exceptionally small molecular size and the strongly attractive interaction of water, and not necessarily to its hydrogen-bonding properties.

(A2-1) Thermodynamics of Apoplastocyanin Folding: Comparison between Experimental and Theoretical Results

It has recently been shown experimentally that the folding of apoplastocyanin (apoPC) illustrated in Fig. 1 accompanies a very large enthalpic loss. This implies that an even larger entropic gain occurs in stabilizing the folded structure to overcome the enthalpic loss. We have calculated the water-entropy gain upon the folding of apoPC using the angle-dependent integral equation theory combined with the multipolar water model and the morphometric approach [2,3]. It is demonstrated that the calculated value is in quantitatively good accord with the value estimated from the experimental data by accounting for the conformational-entropy loss.

According to a prevailing view, the water adjacent to a hydrophobic group is unstable especially in terms of the rotational entropy and the folding is driven primarily by the release of such unfavorable water to the bulk through the burial of nonpolar side chains. We show, however, that the resultant entropic gain is too small to elucidate the experimental result. The great entropic gain observed is ascribed to the reduction of the restriction for the translational motion of water molecules in the whole system.

Fig 1. Folding of apoplastocyanin.

(A2-2) Molecular Mechanism of Pressure Denaturation of Proteins

The native structure of a protein is unfolded by various perturbations such as the addition of chemical substances, the change in the temperature or pH, and the application of a high pressure. Investigating these processes will provide physical insights into the folding/unfolding transition of proteins. Above all, the pressure-induced unfolding is one of the most challenging subjects.

We have investigated the molecular mechanism of pressure denaturation of proteins using the angle-dependent integral equation theory combined with the multipole water model and the morphometric approach. We argue that the hydration entropy of a protein is the key quantity. In accordance with the experimental observations, it is verified that at an elevated pressure a swelling structure, which has only moderately less compact than the native structure but has a much larger water-accessible surface area, turns more stable than the native structure in terms of the water entropy [4]. The swelling structure is characterized by the penetration of water into the interior. In order to elucidate the molecular mechanism of the denaturation, we decompose the hydration entropy into contributions from the translational and rotational restrictions for the molecular motions of water. Each contribution is further decomposed into the water-protein pair correlation component and the water-water-protein triplet and higher-order correlation component. The pair correlation component in the translational contribution is divided into two terms arising from the excluded volume and the water structure near the protein, respectively. It is found that pressure denaturation accompanies a loss of the translational and rotational entropies at the pair correlation level but a much larger gain of the translational entropy at the triplet and higher-order correlation levels. This implies that the water penetration leads to a great reduction of the translational restriction for the water molecules which are sufficiently far from the protein.

(A4) Novel Scoring Function for Discriminating Native Fold from Misfolded Decoys

The prediction of the native structure of a protein from its amino-acid sequence is an extremely difficult problem which has remained unresolved for these fifty years. It appears that a breakthrough is not likely to be obtained unless a unique concept, which is totally different from the conventional approaches, is employed.

As the first step toward developing a reliable method for predicting the protein native structure, we have developed a novel scoring (free-energy) function which enables us to discriminate the native fold from a number of misfolded decoys [5]. The function consists of the hydration entropy calculated under the isochoric condition and the total dehydration penalty. Compared to the fully extended structure which possesses the maximum number of hydrogen bonds with water molecules and no intramolecular hydrogen bonds, in a more compact structure some donors and acceptors (e.g., N and O, respectively) are buried in the interior after the break of hydrogen bonds with water molecules (CO···W, NH···W, etc.). There is no problem if the intramolecular hydrogen bonds (CO···HN, etc.) are formed. However, it is not always formed, leading to the dehydration penalty.

In the protein research community, “decoy” sets are prepared for a number of proteins so that people can check the power of their scoring function. Each decoy set comprises the data of nearly one thousand *misfolded* structures. Most of them were generated using bioinformatics and related techniques. Structures which are similar to the native fold are included in the data set. The power of the scoring function can be evaluated by checking if the native fold gives the lowest value of the scoring function and it can thus be selected from among the decoy and native folds. We have tested our scoring function for the decoy sets, “Four-State Reduced (see Fig. 2)”, “Fisa” (except for one unreasonable set), and “Baker CASP3”, and succeeded in identifying the native fold as the structure with the lowest scoring function for all the sets (15 proteins). Up to now no other group has made the same success.

Our method thus developed is best suited to selecting the most stable structure from among the candidate structures. The number of the candidate structures is allowed to be huge, because in our method the scoring function is calculated with minor computational effort. In the next stage, we intend to develop a practical tool for predicting the native structure of a protein from its amino-acid sequence, by combining our method with the bioinformatics techniques which can generate a variety of candidate structures. Our method is capable of handling much larger proteins than those considered

above and can also be extended to analyses of protein-protein interaction and protein aggregation. The application to the drug design is also possible.

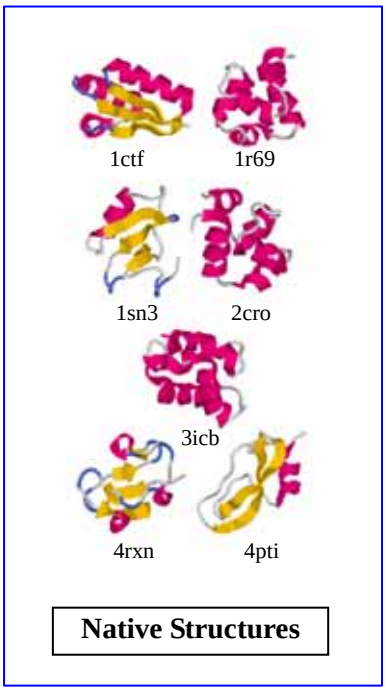


Fig. 2. Seven proteins in the "Four-State Reduced" decoy sets.

(B-1). Dependence of the Bulk-Ion Temperature on the Bumpy Field Component

The effect of the magnetic configuration on the fast ion confinement is one of the most important issues in helical devices. In this paper, fast ion confinement is studied using ion cyclotron range of frequency (ICRF) heating in the minority heating scheme in Heliotron J [6,7,8], a low-shear helical-axis heliotron (R_0 = 1.2 m, a = 0.1-0.2 m, $B_0 \leq$ 1.5 T). The effect of the bumpiness,

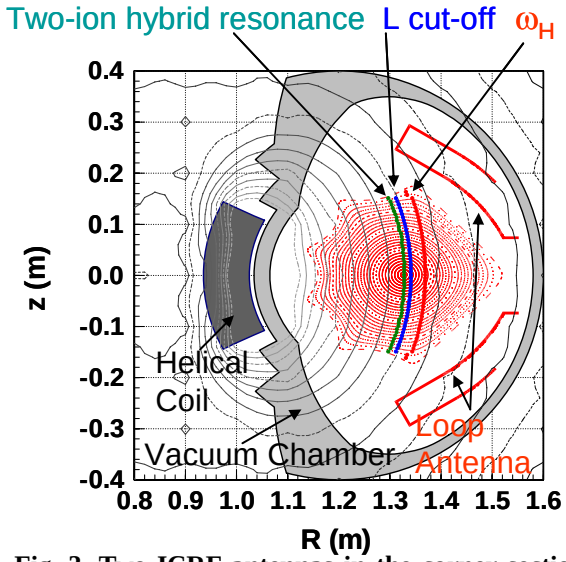
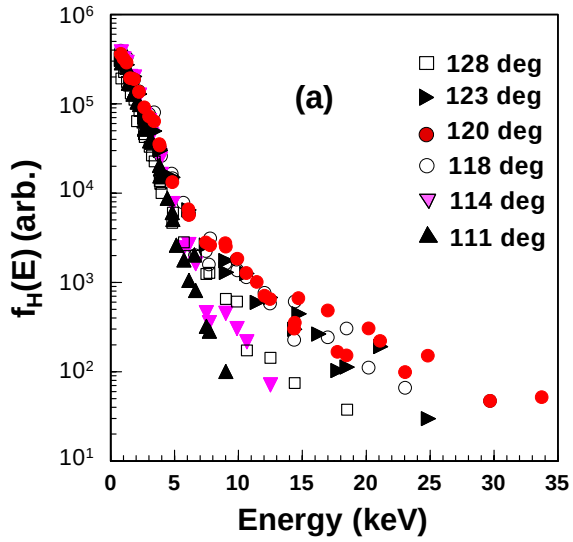


Fig. 3. Two ICRF antennas in the corner section of Heliotron J.

which is the toroidal field ripple, on fast ion confinement and heating efficiency are discussed in the previous papers [9,10,11]. The good confinement of fast ions and the high efficiency of ICRF heating in the high bumpy case are reported. Here, the pitch angle dependence of energy spectra for high bumpy case is measured for the first time and compared with the medium bumpy case, then, the fast ions up to 34 keV are observed during ICRF heating in Heliotron J. The configurations used in this study are as follows; the bumpiness (B_{04}/B_{00}, where B_{04} is the bumpy component and B_{00} is the averaged magnetic field strength) are 0.15



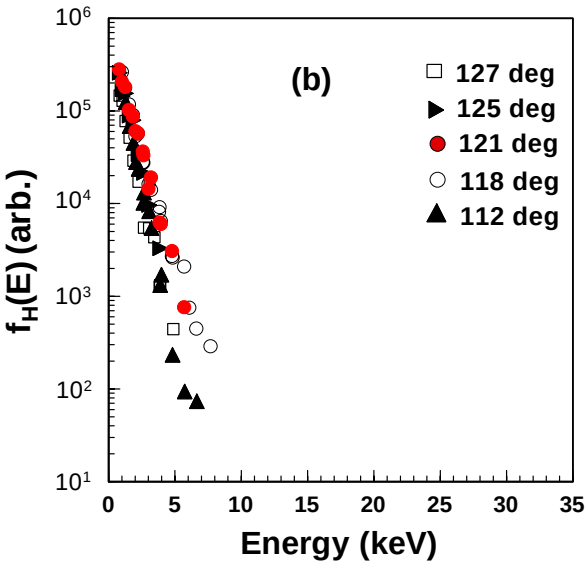


Fig. 4. Minority hydrogen energy spectra for various pitch angles in the high bumpy configuration (a) and in the medium bumpy configuration (b).

(high) and 0.06 (medium) at ρ = 0.67. Herem r is the normalized minor radius. The configuration of B_{04}/B_{00} = 0.06 corresponds to the standard configuration in Heliotron J.

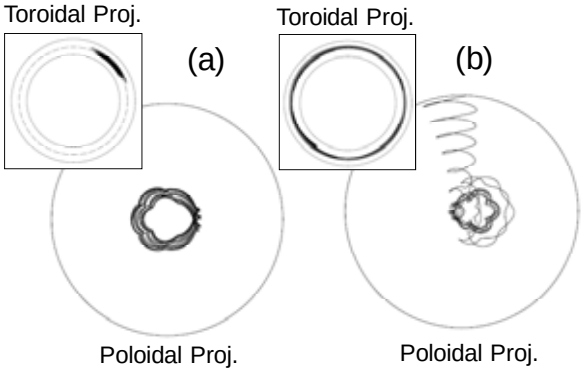


Fig. 5. Trapped ion orbits starting in the straight section for the high bumpy case (a) and the medium bumpy case (b).

The ICRF loop antennas are installed on the low-field side of the corner section of Helitoron J as shown in Fig. 3. An ICRF pulse of 23.2 MHz is injected into an ECH target plasma where $T_i(0)$ = 0.2 keV, $T_e(0)$ = 0.8 keV and $\bar{n}_e = 0.4 \times 10^{19}$ m^{-3}. ICRF injection power is about 270 kW. The majority species is deuterium and the minority is hydrogen. Figure 4 shows measured minority hydrogen energy spectra for various pitch angles by changing toroidal angle of a charge exchange neutral energy analyzer (CX-NPA) for two bumpy cases. In high bumpy case, the ion flux is measured up to 34 keV at the pitch angle of 120 deg. Such high energy particles cannot be observed in the medium bumpy configuration. In both cases, the higher energy flux is measured near 120 deg in pitch angle although the ions are considered to be accelerated by ICRF heating in the perpendicular direction.

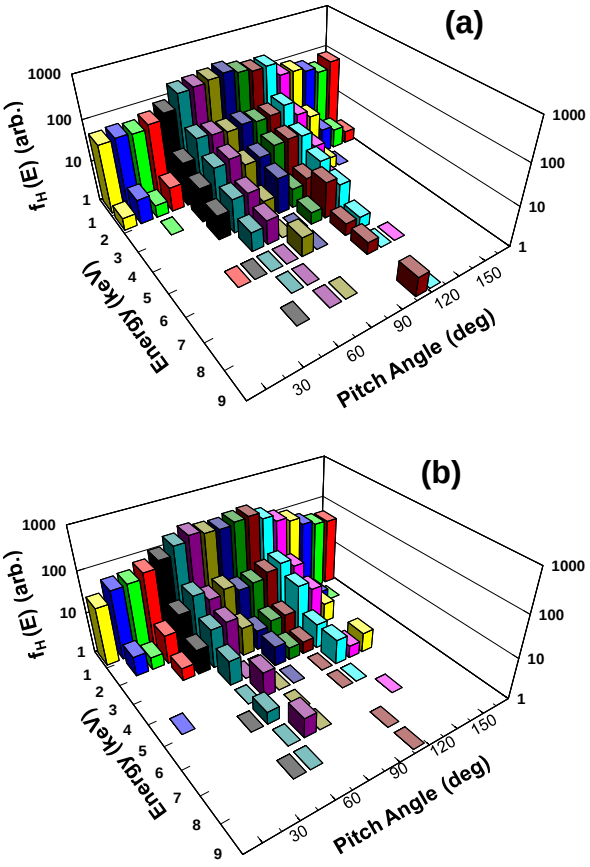


Fig. 6. Calculated pitch angle dependence of energy spectra in the high bumpy case (a) and the medium bumpy case (b).

To understand experimental results, Monte Carlo calculation is performed. The numerical model consists of orbit tracing, Coulomb collisions and acceleration by the ICRF heating. Minority protons are regarded as test particles and the heating is simulated by the velocity kick in the perpendicular direction in velocity space when ions cross the cyclotron layer. The initial distribution of test ions is uniform in the toroidal and poloidal directions, and parabolic in the radial direction. The energy is chosen randomly due to the Maxwell distribution of bulk ions. Examples of the orbit of trapped ion with energy of 1 keV in Boozer coordinates are shown in Fig. 5 for two configurations under the same initial conditions. The ion in Fig. 5(a) is trapped in the toroidal direction due to high bumpiness and confined in the plasma. The ion in Fig. 5(b) is barely trapped, then, lost finally. Figure 6(a) shows the relation of calculated energy spectra to the pitch angle in the high bumpy case. The high energy ions are generated near 60 deg and 120 deg in pitch angle. There are high energy ions at 90 deg; however they are not so large. The property is almost same in the medium bumpy case shown in Fig. 6(b). The calculation results using Monte Carlo Method represents that the accelerated ion distribution has its peak in the range of 20 deg to 30 deg from the perpendicular direction. This result is considered to be caused mainly due to the existence of the loss region around the perpendicular direction [11].

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COLLABORATION WORKS IN THE LABORATORY FOR COMPLEX ENERGY PROCESSES

1. Introduction

The laboratory was established in 1996 simultaneously with the Institute as an attached facility for research on advanced processes of energy production, conversion and application. In order to perform the research objectives of the Institute of Advanced Energy, it is essentially necessary to organize the cooperative research program with much close connection between related research fields in the Institute. The laboratory takes charge of organizing and promoting the cooperative research project as a center of research activity in the Institute. The research staffs in the Institute participate in specific projects to carry out their subjects. The scientists of other faculties in Kyoto University can also participate the cooperative project to enhance the progress of research and educational activities. The laboratory also manages various functions such as symposium and seminar for related topics on energy field.

The cooperative research activities will be published in a publication edited in the laboratory at the end of the year.

Research activities have been directed mainly toward the following cooperative projects as the principle research subject of the Institute.

A1 Interdisciplinary Field of Plasma Energy

The scientific and technological researches on advanced plasma energy system aim at the development of a frontier field "complex plasma energy systems research" based on plasma, hydrogen and material sciences. This field includes the basic research on advanced plasma energy related to the plasma confinement improvement, the effective transport of heat and particle fluxes, the system construction for hydrogen fuel cycle, the basic study on POP of advanced divertor, the control of plasma surface interaction and the development of materials under extremely severe environment, application of plasma energy based on plasma basic research, and advanced fission energy research.

A2 Interdisciplinary Field of Bioenergy

Researches in this field include development of highly efficient material/energy-transformation systems on the basis of bio-nano-technologies and biomimetic approaches and elucidation, improvement, and utilization of the biological processes. The environmental aspects associated with the energy utilization are also studied in this field.

A3 Interdisciplinary Field of Photon and Quantum Energy

This specially-promoted field includes researches for extending advanced functions peculiar to photon, quantum and related materials, studies of fundamentals and /or technology for generating new functions of the energy, and interdisciplinary studies using the energy functions aiming at the creation of a new field of science and technology.

B. Cooperative use of facilities and equipments

Facilities and equipments of the laboratory are provided to cooperated researches for the scientists in the university.

2. The cooperative research project consists of (a) a specific program for "Promotion of a priority project" and (b) a standard program.

(a) A specific program was not planned in this year.

(b) Summary of the standard cooperative research subjects carried out in the year of 2007.

A public collection of cooperative research application was carried out, in this year, for a program which consits of 3 group of "Kiban" , "Syorei" and "Kikaku-chosa" cooperative research. The "Kiban " cooperative research means a program to promote leading research themes of the Institute projects. The "Syorei" cooperative research means a program to promote general research themes with respect to the Institute projects. The "Kikaku-chosa" cooperative research means a program to promote the cooperative research through a seminar or symposium.

As a result, the research themes of 51 were applied and applications of 50 were accepted after the approval by a steering committee of the laboratory. The number of research subjects are listed in Table 1 according to the project categories.

Table 1 Number of the accepted research subjects according to the standard project theme
The whole sum 50

		category A			B	total
		A1	A2	A3		
Kibann *1	inside	1	2	1	0	4
	outside	0	0	0	0	0
Syorei *2	inside	11	5	5	0	21
	outside	15	4	3	2	24
Kikaku -chosa *3	inside	1	0	0	0	1
	outside	0	0	0	0	0

"inside" or "outside" : Numbers applied by the inside or outside of the Institute
The individual research subjects are as follows, *1, *2 and *3 mean the "Kiban", "Syorei" and "Kikaku-chosa' cooperative research theme, respectively,

The individual Research subjects are as follows.

A1

"Controllability of Spontaneous Transition in NBI Plasmas of HeliotronJ"

(1) S. Kobayashi, T. Mizuuchi, K. Nagsaki, F. Sano, H. Okada, K. Hanatani
(2) K. Kondo, Y. Nakamura, H. Arimoto, F. Hamagami
(3) S. Yamamoto
(4) T. Minami, S. Okamura, S. Nishimura, K. Nagaoka, Y. Suzuki
(5) S. Kitajima
(6) Y. Nakashima
(7) S. Murakami
(1) Institute of Advanced Energy, Kyoto University
(2) Graduate School of Energy Science, Kyoto University
(3) Kyoto University Pioneering Research Unit for Next Generation
(4) National Institute for Fusion Science
(5) School of Engineering, Tohoku University
(6) Plasma Research Center, University of Tsukuba
(7) Graduate School of Engineering, Kyoto University

"Optimization of Helical System Concept"

(1) F. Sano, T. Mizuuchi, K. Nagasaki, K. Hanatani, H. Okada, S. Kobayashi
(2) K. Kondo, Y. Kishimoto, Y. Nakamura, G. Motojima, H. Arimoto
(3) T. Mutoh, S. Okamura, K. Ida, K. Toi, Y. Suzuki, H. Iguchi, A. Fujisawa, M. Yokoyama, T. Minami, S. Nishimura, Y. Yoshimura, M. Isobe, C. Suzuki, K. Nagaoka, Y. Takeiri, O. Yamagishi, T. Akiyama
(4) S. Yamamoto
(5) A. Isayama
(6) S. Murakami
(7) T. Fukuda
(1) Institute of Advanced Energy, Kyoto University
(2) Graduate School of Energy Science, Kyoto University
(3) National Institute for Fusion Science
(4) Kyoto University Pioneering Research Unit for Next Generation
(5) Japan Atomic Energy Agency
(6) Graduate School of Engineering, Kyoto University
(7) Graduate School of Engineering, Osaka University

"Study of Cluster Injection Technique for Density Control"

(1) T. Mizuuchi, H. Okada, S. Kobayashi
(2) J. Miyazawa
(3) L. Yao
(4) S. Watanabe
(1) Institute of Advanced Energy, Kyoto University
(2) National Institute for Fusion Science
(3) South Western Institute of Physics
(4) Graduate School of Energy Science, Kyoto University

"Confinement Improvement in Helical Systems"

(1) T. Mizuuchi
(2) S. Kitajima
(3) A. Fujisawa, M. Yokoyama
(4) Y. Kishimoto
(1) *Institute of Advanced Energy, Kyoto University*
(2) *School of Engineering, Tohoku University*
(3) *National Institute for Fusion Science*
(4) *Graduate School of Energy Science, Kyoto University*

"Control of Non-inductive Current Drive in Toroidal Plasmas"

(1) K. Nagasaki, T. Mizuuchi, S. Kobayashi, K. Sakamoto, F. Sano, K. Hanatani, H. Okada
(2) K. Kondo, Y. Nakamura
(3) Y. Yoshimura
(4) A. Fernandez, A. Cappa
(1) Institute of Advanced Energy, Kyoto University
(2) Graduate School of Energy Science, Kyoto University
(3) National Institute for Fusion Science
(4) CIEMAT, Spain

“Study of ICRF Heating Efficiency for Helical-Axis Heliotron Plasmas”

(1) H. Okada, S. Kobayashi, T. Mizuuchi, K. Nagasaki, F. Sano, K. Hanatani
(2) K. Kondo, Y. Nakamura
(3) T. Mutoh
(4) Y. Nakashima
(5) S. Yamamoto
(6) N. Nishino
(1) Institute of Advanced Energy, Kyoto University
(2) Graduate School of Energy Science, Kyoto University
(3) National Institute for Fusion Science
(4) Plasma Research Center, University of Tsukuba
(5) Kyoto University Pioneering Research Unit for Next Generation
(6) Graduate School of Engineering, Hiroshima University

“Studies of the Cylindrical Discharge Type Fusion Neutron Source -Study of Long Line Source-”

(1) Y. Yamamoto, Y. Takeuchi, S. Konishi, K. Noborio
(2) A. Ishidou
(1) Institute of Advanced Energy, Kyoto University
(2) Graduate School of Energy Science, Kyoto University

“Effect of Catalyst on Hydrogen Production Rate in Hydrogen Production Process with Biomass”

(1) Y. Takeuchi, Y. Yamamoto, S. Konishi
(2) T. Hasegawa
(3) T. Kamei
(1) Institute of Advanced Energy, Kyoto University
(2) Graduate School of Energy Science, Kyoto University
(3) Institute of Sustainability Science, Kyoto University

“Strength and Microstructure Interaction of Ceramic Matrix Composites for Advanced Nuclear Fusion Blanket System : (1)Strength at the Fiber/Matrix Interface”

(1) T. Hinoki, A. Kohyama
(2) T. Nozawa
(1) Institute of Advanced Energy, Kyoto University
(2) Japan Atomic Energy Agency

“Research of Investigation Technique for Thermal Conductivity Modification under Irradiation Environments Using Ion-Irradiation Methods”

(1) H. Kishimoto, A. Kohyama, T. Hinoki
(2) K. Shimoda, K. Ozawa
(1) Institute of Advanced Energy, Kyoto University
(2) Graduate School of Energy Science, Kyoto University

“Turbulent Heat Transfer for Heating of Water in a Short Vertical Tube”

(1) K. Hata
(2) K. Fukuda
(3) N. Noda
(1) Institute of Advanced Energy, Kyoto University
(2) Graduate School of Maritime Sciences, Kobe University
(3) National Institute for Fusion Science

“A Modeling Study of Fusion Plasma-Material Interactions”

(1) K. Morishita, A. Kohyama, K. Hanatani
(2) Y. Watanabe
(3) H .Iwakiri
(4) D. Kato
(5) Y. Kaneta
(1) Institute of Advanced Energy, Kyoto University
(2) Graduate School of Energy Science, Kyoto University
(3) Research Institute for Applied Mechanics, Kyushu University
(4) National Institute for Fusion Science
(5) Graduate School of Engineering, The University of Tokyo

“Study on Mechano-Chemical Effects Emerged During Fabrication Processing of Oxide Dispersion Strengthened Steel”

(1) R. Kasada, A. Kimura
(2) J.H. Lee
(3) Q. Xu, T. Yoshiie
(1) Institute of Advanced Energy, Kyoto University
(2) Graduate School of Energy Science, Kyoto University
(3) Kyoto University Research Reactor Institute

“Evaluation of Diffusion Coefficient of Impurity in HeliotronJ ECH Plasmas”

(1) K. Kondo, H. Arimoto, H. Matsuoka, G. Motojima, S. Watanabe, D. Katayama, H. Takahashi, A. Nakajima, F. Hamagami, K. Mukai, K. Murai, H. Yasuda
(2) F. Sano, T. Mizuuchi, H. Okada, K. Nagasaki, S. Kobayashi
(3) N. Nishino
(4) S. Yamamoto
(1) Graduate School of Energy Science, Kyoto University
(2) Institute of Advanced Energy, Kyoto University
(3) Graduate School of Engineering, Hiroshima University
(4) Kyoto University Pioneering Research Unit for Next Generation

“Measurement of HeliotronJ Peripheral Plasma Using Fast Cameras”

(1) N. Nishino, Y. Fukuyama, K. Kaga, S. Sakurada, S. Honda, T. Izuka
(2) K. Kondo
(3) F. Sano, T. Mizuuchi, H. Okada, K. Nagasaki, S. Kobayashi
(1) Graduate School of Engineering , Hiroshima

University
(2) Graduate School of Energy Science, Kyoto University
(3) Institute of Advanced Energy, Kyoto University

"Wide-Band Simultaneous ECE Measurement in Heliotron -J"
(1) Y. Yoshimura, S. Okamura, S. Kubo, T. Shimozuma, H. Igami
(2) K. Nagasaki, F. Sano, T. Mizuuchi, H. Okada, S. Kobayashi
(3) H. Yasuda, K. Mukai
(1) National Institute for Fusion Science
(2) Institute of Advanced Energy, Kyoto University
(3) Graduate School of Energy Science, Kyoto University

"Spectroscopic Measurement and Neutral Transport Analysis in Non-Axisymmetric System Using Monte-Carlo Simulation"
(1) Y. Nakashima, M. Ichimura
(2) S. Kobayashi, T. Mizuuchi, F. Sano, K. Nagasaki, H. Okada
(3) K. Kondo, Y. Nakamura, H. Arimoto, F. Hamagami
(4) S. Yamamoto
(1) Plasma Research Center, University of Tsukuba
(2) Institute of Advanced Energy, Kyoto University
(3) Graduate School of Energy Science, Kyoto University
(4) Kyoto University Pioneering Research Unit for Next Generation

"Studies of Identification of MHD Equilibrium of HeliotronJ Plasmas"
(1) S. Sakakibara, Y. Suzuki
(2) S. Yamamoto
(3) H. Okada, F. Sano, T. Mizuuchi, K. Nagasaki, S. Kobayashi
(4) K. Kondo, Y. Nakamura, G. Motojima
(1) National Institute for Fusion Science
(2) Kyoto University Pioneering Research Unit for Next Generation
(3) Institute of Advanced Energy, Kyoto University
(4) Graduate School of Energy Science, Kyoto University

"Microstructural Evolution of Oxide Dispersion Strengthened Ferritic Steel under Stress Loading and Sodium Exposure at Elevated Temperatures"
(1) M. Inoue, S. Kim, S. Ohtsuka, T. Kaito
(2) A. Kohyama, T. Hinoki
(1) Japan Atomic Energy Agency
(2) Institute of Advanced Energy, Kyoto University

"High-Fluence Irradiation Behavior of Reduced Activation Fusion Reactor Materials"
「Evaluation of Dual Beam Irradiation Behavior in Reduced Activation ODS Steels at High Temperature」
(1) H. Tanigawa, H. Ogiwara
(2) A. Kohyama, T. Hinoki, H. Kishimoto
(1) Japan Atomic Energy Agency
(2) Institute of Advanced Energy, Kyoto University

"Evaluation of SiC and W Coated SiC as Plasma Facing Materials"
(1) Y. Ueda
(2) T. Hinoki
(1) Graduate School of Engineering, Osaka University
(2) Institute of Advanced Energy, Kyoto University

"Ion Irradiation Effects on Mechanical Properties of SiC and W Joining Interface"
(1) T. Shibayama, S. Watanabe
(2) A. Kohyama, T. Hinoki, H. Kishimoto
(1) Center for Advanced Research of Energy Conversion Materials, Hokkaido University
(2) Institute of Advanced Energy, Kyoto University

"Oxidation Effect on Stress Relaxation Behavior of SiC-Based Materials"
(1) S. Nogami, A. Hasegawa
(2) T. Hinoki, A. Kohyama
(1) School of Engineering, Tohoku Univerrsity
(2) Institute of Advanced Energy, Kyoto University

"Studies on Thermal and Electrical Conductivities of SiC/SiC Composites"
(1) T. Tanaka, T. Muroga
(2) A. Kohyama, T. Hinoki
(1) National Institute for Fusion Science
(2) Institute of Advanced Energy, Kyoto University

"Evaluation of Thermal and Physical Properties for NITE-SiC/SiC Composite under Severe Service Environment"
(1) G. Sasaki, Y. Choi
(2) T. Hinoki, J.S. Park
(1) Graduate School of Engineering, Hiroshima University
(2) Institute of Advanced Energy, Kyoto University

"Dynamical Behavior and Segregation of Helium in Austenitic and Ferritic Steels"
(1) M. Miyamaoto, K. Ono
(2) K. Moroshita
(1) Interdisciplinary Faculty of Science and Engineering, Shimane University
(2) Institute of Advanced Energy, Kyoto University

"Theoretical Simulations for Irradiated Materials Based on First Principles Method"
(1) Y. Kaneta, Y. Chen
(2) A. Kohyama, K. Morishita
(3) Y. Watanabe
(1) Graduate School of Engineering, The University of Tokyo

(2) Institute of Advanced Energy, Kyoto University
(3) Graduate School of Energy Science, Kyoto University

"Irradiation Effects on Solid Materials with Complicated Crystal Structure"
(1) H. Iwakiri, N. Yoshida
(2) K. Morishita
(1) Research Institute for Applied Mechanics, Kyushu University
(2) Institute of Advanced Energy, Kyoto University

A2

"Highly Efficient Bioethanol Production from Woody Biomass"
(1) T. Kodaki, K. Makino, S. Watanabe
(1) Institute of Advanced Energy, Kyoto University

"Fabrication of Materials for Photon-Electron Transfer by Using Functional Peptide"
(1) T. Sagawa, S. Yoshikawa, Y. Kobuke
(2) H. Ihara, H. Jinntoku
(3) O. Yoshikawa
(1) Institute of Advanced Energy, Kyoto University
(2) Faculty of Engineering, Kumamoto University
(3) Graduate School of Energy Science, Kyoto University

"Light Scattering Spectrum of Monolayers of Molecules and Particles at Liquid Interfaces"
(1) T. Sakka, K. Fukami, Y.H. Ogata
(2) L.E. Noreng, G. Oye
(1) Institute of Advanced Energy, Kyoto University
(2) Department of Chemical Engineering, Norwegian University of Science and Technology

"Morphological Control of Conducting Polymer on Porous Silicon Template"
(1) K. Fukami, Y.H. Ogata, T. Sakka
(2) T. Yamauchi
(3) Y. Suzuki
(1) Institute of Advanced Energy, Kyoto University
(2) Graduate School of Science and Technology, Niigata University
(3) Uyemura &Co.,Ltd.

"Design of Mobil RNA-Protein Catalytic Complexes"
(1) T. Morii, M. Fukuda
(1) Institute of Advanced Energy, Kyoto University

"Development of Energy-and Material-Recycling Systems by Use of Supercritical Fluid and Biological Functions"
(1) K. Makino, S.P. Pack, T. Kodaki, S. Watanabe
(2) K. Tajima
(1) Institute of Advanced Energy, Kyoto University
(2) Kyoto Institute of Technology

"Identification of Novel Metabolic Pathway of Sugar for Development of Highly Efficient Bioethanol Production"
(1) T. Kodaki, K. Makino, S. Watanabe
(1) Institute of Advanced Energy, Kyoto University

"Energies of ∑ 3 Coincidence Small Angle Boundaries in Si"
(1) K. N.Ishihara
(2) A. Ohtsuki
(1) Graduate School of Energy Science, Kyoto University
(2) Institute of Advanced Energy, Kyoto University

"Structure-Based Design of Miniature Methane Monooxygenase Hydroxylase"
(1) N. Fujieda
(2) T. Morii
(3) M. Inoue, Y. Tatsuyama
(1) Kyoto University Pioneering Research Unit for Next Generation
(2) Institute of Advanced Energy, Kyoto University
(3) Graduate School of Energy Science, Kyoto University

"Photocurrent Generation Devices Using Nucleic Acid Derivatives"
(1) K. Yamana
(2) T. Morii
(1) Graduate School of Engineering, University of Hyogo
(2) Institute of Advanced Energy, Kyoto University

"Formation of Nano-Scale Structures and Molecular Recognition by Synthetic Peptide"
(1) S. Konno
(2) T. Morii
(1) Faculty of Medical Sciences, University of Fukui
(2) Institute of Advanced Energy, Kyoto University

A3

"Nano-Scale Processing with a Near Field Generated in Femtosecond Laser Irradiation"
(1) G. Miyaji, K. Miyazaki
(2) N. Yasumaru
(3) M. Obara
(4) A.E. Kaplan
(1) Institute of Advanced Energy, Kyoto University
(2) Fukui National College of Technology
(3) Faculty of Science and Technology, Keio University
(4) Johns Hopkins University

"Study on the Electron Beam Stabilization in KU-FEL"
(1) H. Ohgaki, K. Masuda, T. Kii
(2) H. Zen, S. Sasaki, T. Shiiyama
(1) Institute of Advanced Energy, Kyoto University
(2) Graduate School of Energy Science, Kyoto

University

“Performance Improvement of KU-FEL RF Gun by Use of RF Triode Structure (Ⅱ)”
(1) K. Masuda, H. Ohgaki, T. Kii
(2) T. Shiiyama, H. Zen
Institute of Advanced Energy, Kyoto University
Graduate School of Energy Science, Kyoto University

“Nonlinear Laser Spectroscopy with Femtosecond-Laser-Induced Molecular Alignment”
(1) K. Miyazaki, G. Miyaji
(2) F.H.M. Faisal
(3) K. Yoshii
(1) Institute of Advanced Energy, Kyoto University
(2) Bielefeld University
(3) Graduate School of Energy Science, Kyoto University

“Ultrafast Phenomena Induced by Intense Far-Infrared Free-Electron Lasers”
(1) T. Nakajima, H. Ohgaki, T. Sakka, C. Liu, H. Tang
(1) Institute of Advanced Energy, Kyoto University

“Pulse Duration and Wavelength Effects upon the Surface Elemental Analysis Based on Liquid-Phase Laser Ablation Plume Spectroscopy”
(1) T. Sakka, T. Nakajima, K. Fukami, Y.H. Ogata
(2) T. Sasaki
(3) Y. Suzuki
(1) Institute of Advanced Energy, Kyoto University
(2) National Institute of Advanced Industrial Science and Technology
(3) Uyemura &Co.,Ltd.

“On the Frontier Study of Atomic Interaction Processes Involving Relativistic Electrons”
(1) A. Itoh, H. Tsuchida, M. Imai
(2) H. Ohgaki, T. Kii, K. Masuda
(1) Graduate School of Engineering, Kyoto University
(2) Institute of Advanced Energy, Kyoto University

“**Preparation of Colloidal Solutions of High-Purity Monodispersed Nobel Metal Nanoparticle Using Laser Ablation in Liquid Media”**
(1) T. Sasaki
(2) T. Sakka
(1) National Institute of Advanced Industrial Science and Technology
(2) Institute of Advanced Energy, Kyoto University

“Promotion of Bond-Forming Reaction by Means of Infrared Pulse Laser Irradiation”
(1) K. Fugami
(2) T. Sakka
(1) Graduate School of Engineering, Gunma University
(2) Institute of Advanced Energy, Kyoto University

B

“Study of Scintillation Efficiency Universal Curves for Crystals”
(1) Y. Uozumi, G. Wakabayashi, M. Imamura, Y. Koba, Y. Fukui
(2) H. Ohgaki, T. Kii
(1) Graduate School of Engineering, Kyushu University
(2) Institute of Advanced Energy, Kyoto University

“Interaction of Helium and Defects in Iron”
(1) Q. Xu, K. Sato, T. Yoshiie
(2) K. Morishita, H. Kishimoto, T. Hinoki, A. Kohyama
(1) Kyoto University Research Reactor Institute
(2) Institute of Advanced Energy, Kyoto University

SYMPOSIUM IN THE LABORATORY

Symposium
The Symposium has been arranged in order to introduce the research activities in sections and to enhance the mutual cooperation among different fields. In 2007 Three regular meetings and the annual meeting for the cooperative research results were held with following theme.

1. The regular meeting

The First Meeting, April 26, 2007
Karl Verfondern「ヨーロッパにおける水素エネルギーシステムの安全に対する取り組み」
Karl Verfondern, "Research Efforts in Europe on the Safety of Hydrogen ", *Institute for Safety Research and Reactor Technology (IEF-6), Research Center Juelich*

The 2nd. Meeting, June 27, 2007
Y.Kotake「新しいスピントラップ剤 CYPMPO の開発」
Y.Kotake, "Development of CYPMPO, a novel spin-trapping agent", *Institute of Advanced Energy, Kyoto University*

The 3nd. Meeting, February 27, 2008
Patrik Schmuki「自己組織化形成する酸化チタンナノチューブ層：形成、特性とその応用」
Patrik Schmuki, "Self-Organized Titanium Oxide Nanotube-Layers : Formation, Properties and Applications", *Department of Material Science, University of Erlangen-Nuremberg*

2. The Annual Meeting for the Cooperative Research Results, April 4, 2008

小林進二「ヘリオトロン J の NBI プラズマにおける自発的遷移現象の制御性」
S.Kobayashi, "Controllability of Spontaneous Transition in NBI Plasmas of HeliotronJ", *Institute of Advanced Energy, Kyoto University*

長崎百伸「トーラスプラズマにおける非誘導電流駆動の制御」
K.Nagasaki, "Control of Non-inductive Current Drive in Toroidal Plasmas", *Institute of Advanced Energy, Kyoto University*

竹内右人「高温核熱利用によるバオイマスからの水素製造反応器の概念設計」
Y.Takeuchi, "Conceptual Design of Hydrogen Production Reactor Using High Temperature Nuclear Heat With Biomass Resource", *Institute of Advanced Energy, Kyoto University*

小瀧　努「木質バイオマスからの高効率バイオエタノール生産」
T.Kodaki, "Highly Efficient Bioethanol Production from Woody Biomass", *Institute of Advanced Energy, Kyoto University*

佐川　尚「有機薄膜太陽電池用光捕集アンテナ色素集合体の開発」
T.Sagawa, "Fabrication of Materials for Photon-Electron Transfer by Using Functional Peptide", *Institute of Advanced Energy, Kyoto University*

深見一弘「多孔質シリコンを鋳型に用いた電解重合における導電性高分子の形態制御」
K.Fukami, "Morphological Control of Conducting Polymer on Porous Silicon Template", *Institute of Advanced Energy, Kyoto University*

藤枝伸宇「単量体ミニチュアメタン水酸化酵素の創製」
N.Fujieda, "Structure-Based Design of Miniature Methane Monooxygenase Hydroxylase", *Kyoto University Pioneering Research Unit for Next Generation*

宮地悟代「フェムト秒レーザー照射により発生する近接場を用いたナノ物質加工」
G.Miyaji, "Nano-Scale Processing with a Near Field Generated in Femtosecond Laser Irradiation", *Institute of Advanced Energy, Kyoto University*

佐々木毅「液相レーザーアブレーションによるナノ粒子分散コロイド溶液の調製」
T.Sasaki, "Preparation of Colloidal Solutions of Nanoparticle Using Laser Ablation in Liquid Media", *National Institute of Advanced Industrial Science and Technology*

中嶋　隆「高強度遠赤外自由電子レーザーによって誘起される超高速現象」
T.Nakajima, "Ultrafast Phenomena Induced by Intense Far-Infrared Free-Electron Lasers", *Institute of Advanced Energy, Kyoto University*

COLLABORATION WORKS WITH OTHER UNIVERSITIES AND ORGANIZATIONS

1. Core University Program between Seoul National University and Kyoto University on Energy Science and Engineering from 1998 to 2007

This Core University Program (hereinafter referred as "CUP"), started in 1998 between the Kyoto University and the Seoul National University has been actively implemented and completed the 10 year collaboration in 2008 March successfully. During this decade, situation in energy research has been drastically changed in both countries, and the role of the collaborative program, that always responded the requests from the both country well, has made a significant difference from the beginning of the program 10 years ago.

Energy policies in both countries now emphasize global environment problem more seriously, and the study of nuclear energy technology is now given more efforts. With the progress of the activities, exchanges have been expanded to involve active research groups in each fields, and the member universities and institutes increased as many as some 70. Particularly in Korea side, nuclear technology and fusion technology are mainly pursued in research institutes. Because this program intends not only to assist collaborative exchanges, the more important objectives of it was to establish a "core" of the personnel and information exchange between both countries. This program achieved very high completion ration of the planned exchanges in each year, particularly from Japan, well over 90%. But it is more important that as the result of this entire program, we can conclude that both Kyoto university and Seoul National University were established as the "Hub" of the exchanges, work as cores in each countries to coordinate over 70 universities and institutes at the both sides, and organize appropriate groups for the desired technical issues.

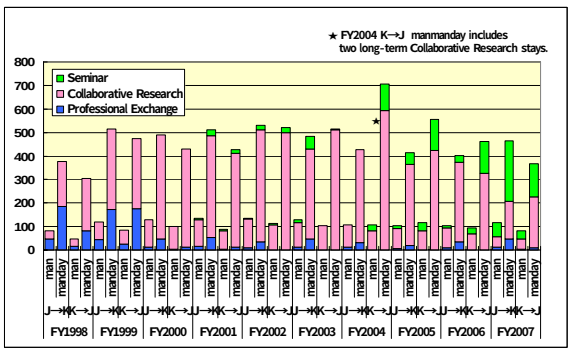

Table 1 personnel exchanges under the Japan-Korea Core University Program.

Table 1 summarizes the numbers of the exchanges and its history in 10 years. As seen in the figure, personnel exchanges took place in the early stage, and as approaching to the completion of the program, most of the activities became collaboration, and seminars are held at the end to conclude each task areas. Approximately 110-130 exchanges were implemented in Accomplishments of the collaboration have been published in the form of joint papers or conference proceedings of exceeding 1200 to date.

Based on our accomplishments, we plan to expand our collaboration to include China for tri-lateral program on the study of advanced energy field, that is the most important technical challenge in all three countries,

We have thus planned a new application, and was selected for the JSPS (Japan Society for the Promotion of Science) Asian CORE (Center Of Research and Education) Program for the "Advanced Energy Science" between Japan, Korea and China. In this program, Japan and core institutes in Asian nations will establish the network of research and education by the extensive collaboration of mutually equal contribution, and eventually provide leading young researchers in the advanced and important field of science. The Institute of Advanced Energy is assigned as a hub institute in Japan to represent universities and research institutes, and collaborate with Seoul National University in Korea and Tsinghua University in China as the counterparts. The 5 year program will start in 2008, and collaborative research, meeting and personnel exchanges will be implemented in the advanced energy science such as

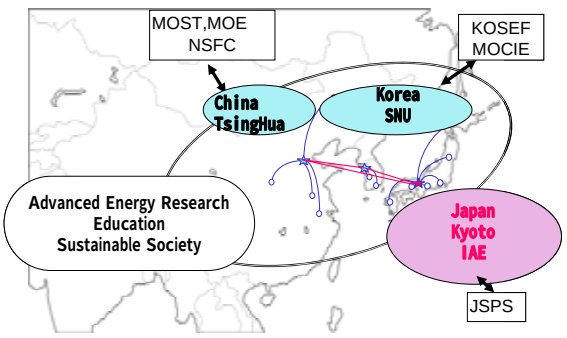

Fig.1 Concept of the newly approved Asian CORE Program.

advanced nuclear energy, fusion, and bio-energy. Figure 1 shows the concept of this program. Coordinators are, Kyoto University: Satoshi Konishi, Seoul National University : Joo HanGyu, and Tsinghua University : Wang Kan. Figure 2 illustrates the structure of the entire program of the Asian CORE, that is operated by JSPS under the MEXT.

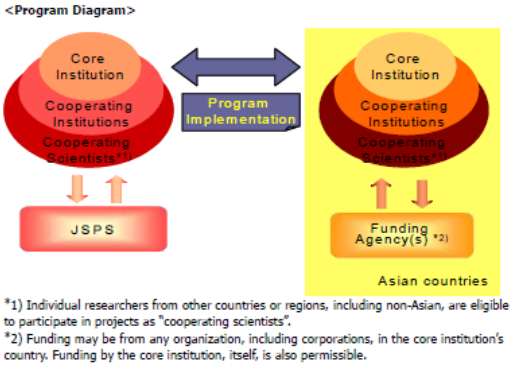

Fig.2 The structure of the Asian CORE Programs under the JSPS.

2. Other Collaboration Works

Advanced Energy Generation Division

Quantum Radiation Energy Research Section

産業技術総合研究所、「新しい量子放射エネルギー発生に関する要素技術研究」、大垣英明、紀井俊輝、増田開、全炳俊、椎山拓己、佐々木怜

Advanced Particle Beam Energy Research Section

CIEMAT（スペイン）、「国際先進ヘリカルにおける改善閉じ込めの研究」、長崎百伸、A.Cappa, A.Fernadez, V.Tribaldos

Australian National University（オーストラリア）、「国際先進ヘリカルにおける改善閉じ込めの研究」、長崎百伸、B.Blackwell

核融合科学研究所、「密度揺動構造とサイクロトロン高調波電子バーンシュタイン波による電流生成の相関について」、長崎百伸

核融合科学研究所、「ヘリオトロン J におけるプラズマ閉じ込め性能の閉じ込め磁場配位からの理論考察」、長崎百伸

核融合科学研究所、「トロイダル電流が MHD 平衡に与える影響の理論的考察とその実験的検証」、長崎百伸

核融合科学研究所、「ヘリオトロン配位における MHD 平衡・安定性に関する研究」、長崎百伸

核融合科学研究所、「磁気計測による磁気島検出器の開発」、長崎百伸

核融合科学研究所、「Heliotron J 装置における電極バイアスによる径方向電場制御」、長崎百伸

核融合科学研究所、「方向性プローブによるプラズマ流計測と揺動の相関」、長崎百伸

核融合科学研究所、「先進ヘリカルによるプラズマ輸送・安定性改善の双方向型共同研究」、長崎百伸

核融合科学研究所、「乱流揺動の構造解析を目的とした長距離揺動相関計測装置の開発と HJ-LHD 比較研究応用」、長崎百伸

核融合科学研究所、「トリムコイルを用いた Heliotron J プラズマの高エネルギー粒子閉じ込め改善の検討」、長崎百伸

核融合科学研究所、「大電力定常ミリ波伝送システムの真空化」、長崎百伸

核融合科学研究所、「高次高調波 ECH によるプラズマ生成の物理」、長崎百伸

日本原子力研究開発機構、「高ベータプラズマの安定維持のための能動的制御と MHD モード解析」、長崎百伸

Advanced Plasma Energy Research Section

Oak Ridge National Laboratory（米国）、Australian National University（オーストラリア）、CIEMAT（スペイン）、Max-Planck Institute（ドイツ）、University of Wisconsin（米国）、Kharkov Institute（ウクライナ）、「先進ヘリカルにおける改善閉じ込めの研究」、佐野史道、水内亨、長崎百伸、岡田浩之、小林進二、J. Hariss (Oak Ridge National Laboratory), B. Blackwell, D. Pretty, H. Punzmann (Australian National University), E. Ascasibar, Á. Cappa, A. Fernández (CIEMAT), N. Marushchenko (Max-Planck Institute), F.S. Anderson (University of Wisconsin), V. Chechikin (Kharkov Institute)

核融合科学研究所、「磁場分布制御技術を用いたプラズマ流制御による輸送・安定性改善研究」、佐野史道、水内亨、花谷清、長崎百伸、岡田浩之、小林進二

核融合科学研究所、「ヘリカル系における核融合プラズマ閉じ込め改善」、水内亨

核融合科学研究所、「ヘリオトロン型プラズマ実験装置開発に関する歴史的資料収集・整理」、水内亨

核融合科学研究所、「LHD における ECH/ECCD プラズマでのイオンテールの発生に関する実験研究」、小林進二

核融合科学研究所、「磁気計測による磁気島検出器の開発」、岡田浩之、水内亨、長崎百伸、小林進二

Advanced Energy Conversion Division

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1. Grant-in-Aid for Scientific Research
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（その他の補助金）

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