RESEARCH ACTIVITIES AT JAERI ON CORE MATERIAL BEHAVIOUR UNDER SEVERE ACCIDENT CONDITIONS

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Abstract

At the Japan Atomic Energy Research Institute(JAERI), experimental studies on physical phenomena under the condition of a severe accident have been conducted. This paper presents the progress of the experimental studies on fuel and core materials behavior such as the thermal shock fracture of fuel cladding due to quenching, the chemical interaction of core materials at high temperatures and the examination of TMI-2 debris.

The mechanical behavior of fuel rod with heavily embrittled cladding tube due to the thermal shock during delayed reflooding have been investigated at the Nuclear Safety Research Reactor(NSRR) of JAERI. A test fuel rod was heated in steam atmosphere by both electric and nuclear heating using the NSRR, then the rod was quenched by reflooding at the test section.

Melting of core component materials having relatively low melting points and their eutectic reaction with other materials significantly influence on the degradation and melt down of fuel bundles during severe accidents. Therefore basic information on the reaction of core materials is necessary to understand and analyze the progress of core melting and relocation. Chemical interactions have been widely investigated at high temperatures for various binary systems of core component materials including absorber materials such as Zircaloy/Inconel, Zircaloy/stainless steel, Zircaloy/(Ag-In-Cd), stainless steel/B_4C and Zircaloy/B_4C. It was found that the reaction generally obeyed a parabolic rate law and the reaction rate was determined for each reaction system.

Many debris samples obtained from the degraded core of TMI-2 were transported to JAERI for numerous examinations and analyses. The microstructural examination revealed that the most part of debris was ceramic and it was not homogeneous in a microscopic sense. The thermal diffusivity data was also obtained for the temperature range up to about 1800K.

The data from the large scale integral experiments were also obtained through the international cooperation such as Phebus-SFD, Phebus-FP and CORA programs to supplement the research at JAERI.

1. Severe Accident Research at JAERI

The wide range of research activities on the severe accident of light water reactors(LWRs) is conducted analytically and experimentally at Japan Atomic Energy Research Institute(JAERI). Outline of present research program at JAERI is shown in Fig.1. The program consists of four research fields, core melt progression, containment integrity, source term and piping integrity.

Research Program Related to Severe Accident

Core Melt Progression
- Evaluation and Analysis of Large Scale Core Melt Progression Tests (Phebus-SFD, CORA)
- Fuel Quench Test at NSRR
- Core Materials Interaction Test
- TMI-2 Debris Examination
- Integrity Evaluation of Reactor Pressure Vessel Lower Head

Containment Integrity
- ALPHA Program
- Analytical Activities on Containment Loads
  - Direct Containment Heating
  - Molten Core/Concrete Interaction
  - BWR Mark-I Containment Liner Melt-through
  - Hydrogen Behavior in Containment
- Containment Structural Analysis

Source Term
- FP Release Tests with Irradiated Fuel
- Experimental Research on FP Iodine Behavior in Containment
- EPSI
- Analytical Activities
  - Integral Analysis
  - Aerosol Behavior in Containment

Piping Integrity
- Aerosol Behavior in Piping
- Structural Behavior of Piping
- Analysis

Fig. 1 Outline of present severe accident research at JAERI

The research on the behavior of LWR core materials under severe accident conditions is included in the first research item of core melt progression which is concerned with physical phenomena in the pressure vessel, such as melting behavior of fuel rod, thermal shock fracture of fuel rod by reflooding, core material interaction at high temperature, etc. In this field of research activities, fuel melt tests[1] and debris coolability tests at the Nuclear Safety Research Reactor(NSRR) are already finished and other experimental programs are still progressing. The examination of TMI-2 debris which provides valuable data for severe accident research is also conducted as well as laboratory scale experiments on core materials interaction. These researches at JAERI consist of relatively small scale experiments and analytical work. To supplement the research at JAERI, information from the large scale integral experiments have also been obtained through international cooperation such as Phebus-SFD and Phebus-FP programs in France and CORA program in Germany.

This paper describes the current status and main results of the experimental studies on fuel and core materials behavior, i.e., fuel rod fracturing by thermal shock, chemical interaction of core materials at high temperature and TMI-2 debris examination.

2. Fuel quench test at the NSRR

In a loss of coolant accident(LOCA) of a LWR, fuel rods will be heated by both decay heat and highly exothermic reaction between steam and Zry, if the emergency core cooling system(ECCS) does not work well by some accidental reason. In such case, fuel rod temperature

Table 1 Test conditions of fuel quench test at NSRR

Test No.	952	954
Test Fuel Rod		
Cladding Material	Zry-4	←
Cladding Outer Diameter(mm)	10.72	←
Cladding Tube Thickness(mm)	0.62	←
Pellet Material	UO_2	←
Pellet Diameter(mm)	9.29	←
Pellet Length(mm)	10.0	←
Enrichment(%)	10	20
Internal Pressure(MPa)	0.5	0.4
Pellet Stack Length(mm)	320	310
Linear Heat Rate of Test Fuel(W/cm)	18	23
Atmosphere	He, Steam	←
Wall Temperature of Test Section(K)	1,133	1,183
Operation Time of NSRR(min)	8	14

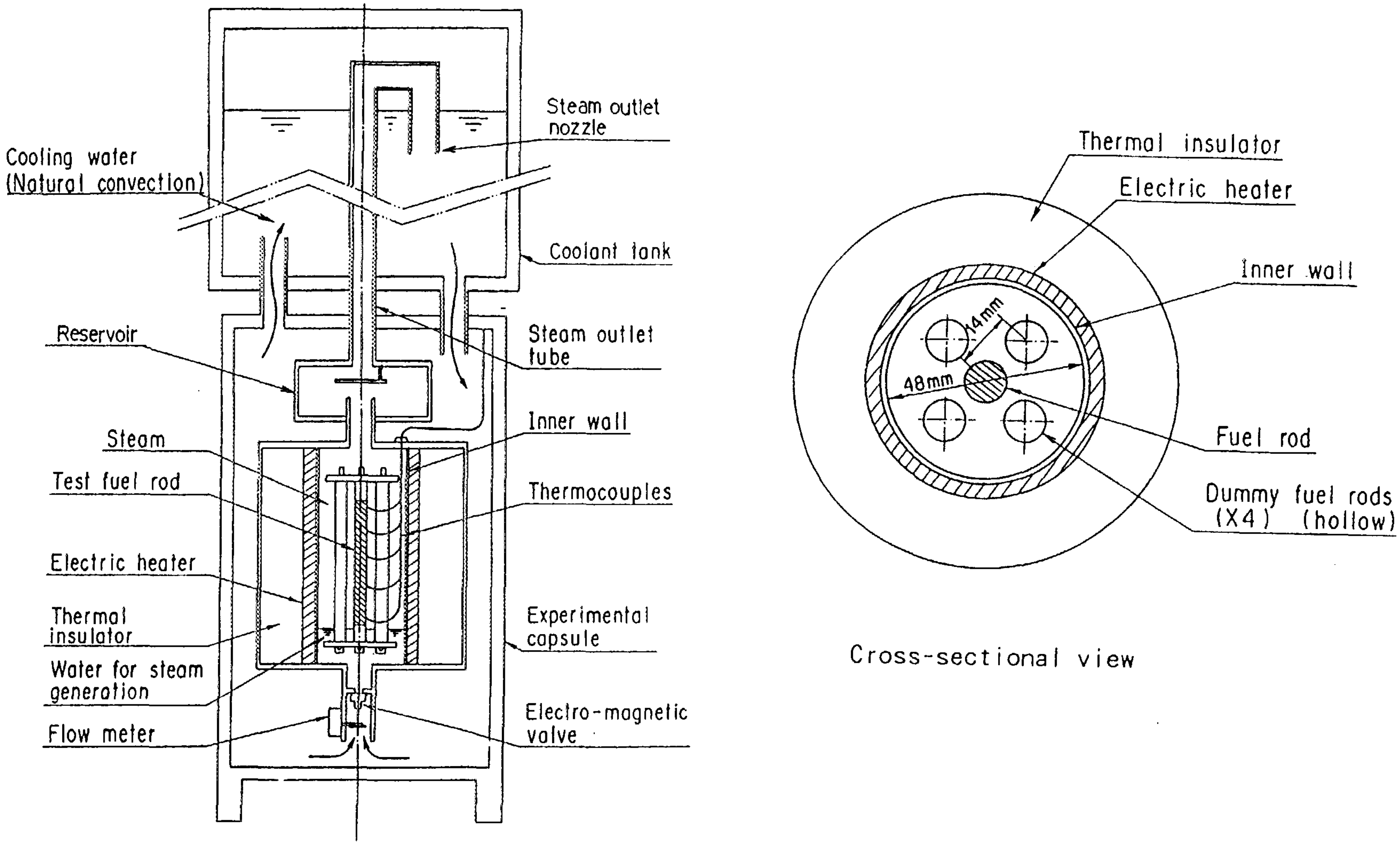


Fig. 2 Schematic of the test capsule for NSRR quench test

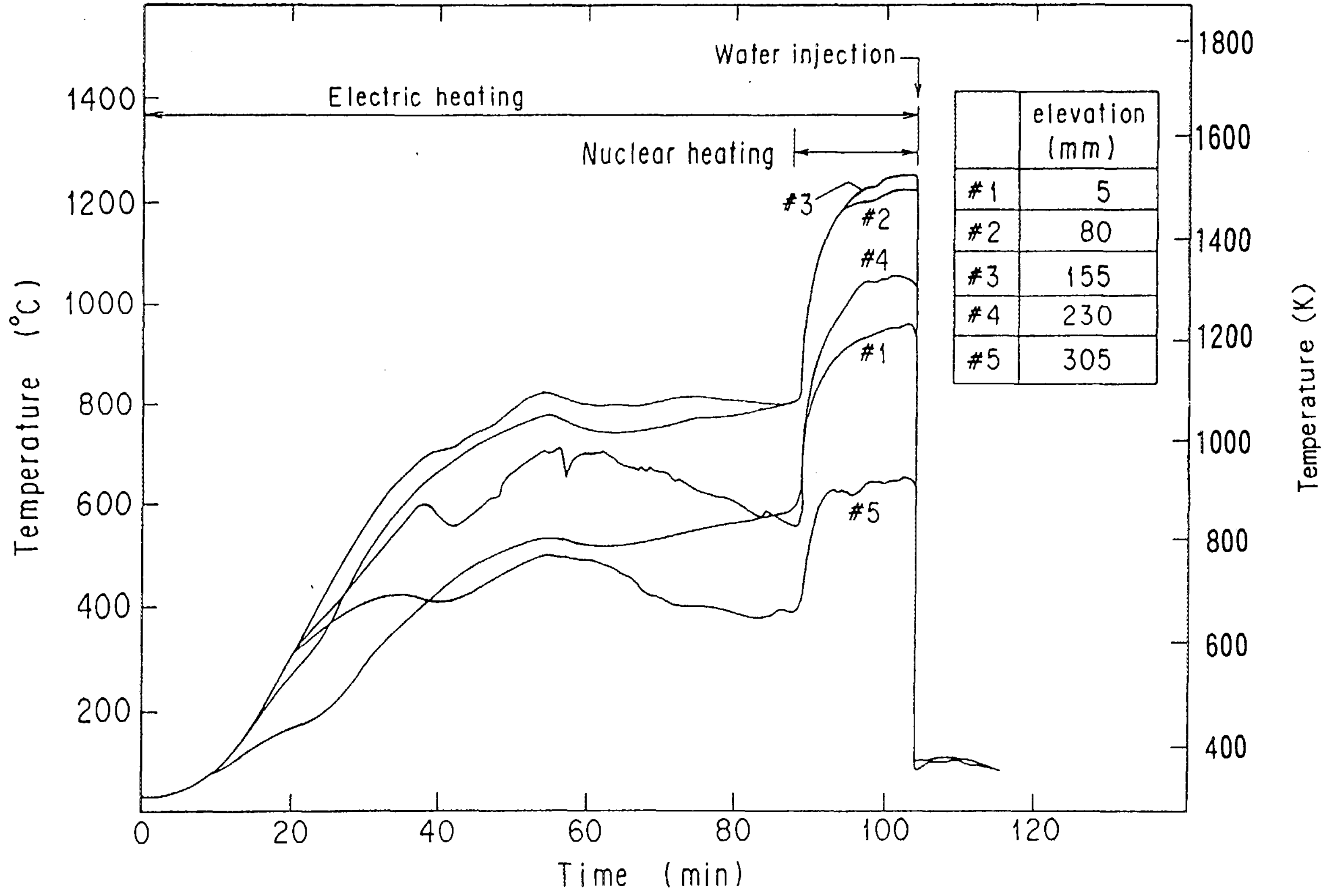


Fig. 3 Temperature histories of cladding outer surface at each elevation in test no. 954
(Maximum cladding temperature : 1533K)

would be raised to very high level beyond the temperature of the design basis event(DBE) criteria, and fuel rods might be damaged severely. Under such severe accident conditions, the cladding tube of fuel rod will rupture and be oxidized on its inner and outer surfaces by high temperature steam. Consequently, severe oxidation of the cladding tube may result in fuel degradation due to thermal shock if delayed reflooding occurs.

The in-pile tests on the fuel behavior during delayed reflooding were performed at the NSRR[2]. The objectives of these tests were to clarify the fuel behavior and degradation conditions during delayed reflooding after the cladding tube was heavily oxidized.

Conditions of the fuel quench tests at the NSRR are summarized in Table 1. A test fuel rod was a shortened PWR type fuel rod containing fresh UO_2 pellets. Initial internal pressure of the test fuel rod was 0.5 or 0.4MPa at the room temperature. With this pressure, the cladding tube is estimated to rupture when the cladding temperature reaches at the level of about 1300K.

Tests were conducted by using a high-temperature flooding capsule. Figure 2 shows a schematic drawing of the capsule. The test fuel rod was placed in the center of the test section. The test fuel rod and four dummy fuel rods were surrounded by an electric heater and a thermal insulator. This capsule with the test fuel rod was fixed at the experimental cavity of the driver core of the NSRR[3] and was irradiated. The environment of the fuel rod during heating was steam which was generated by evaporation of water at the bottom of the test section. An electro-magnetic valve was installed at the bottom of the test section in order to inject water into the test section to simulate reflooding.

Figure 3 shows temperature histories of cladding outer surface measured with thermocouples at each elevation in Test No.954 which exhibited the highest cladding temperature in the test series. The elevations, #1 to #5 denotes the axial position of fuel rod from the bottom of fuel stack. The fuel rod was heated initially only by the electric heater to the temperature beyond 1000K. Then, nuclear heating was started at the linear heat rate of 23W/cm, and the cladding surface temperature reached 1533K. The test was terminated by the water injection into the test section, and the fuel rod was quenched. Another test(Test No.952) was conducted under the same condition except for the linear heat rate.

In Test No.952 which exhibited lower maximum cladding temperature of 1300K, the cladding tube ballooned and ruptured at the middle elevation where the temperature was the highest as shown in Fig.4. Though whole surface of the cladding was oxidized, fragmentation of the cladding tube did not occur in this test. Sum of the oxide layer thickness on the outer and inner surfaces corresponded to about 10% of the cladding thickness which became thinner than the initial value due to ballooning. In contrast to this, the cladding tube ballooned and fractured into several pieces in Test No.954 as shown in Fig.5. These fragments of the cladding tube were 1 to 2cm long. Fragmentation occurred between the elevation of 32mm and 160mm where the temperature was the highest. Inner surface of cladding tube was heavily oxidized in the region above an elevation of 70mm, and its metallic luster was lost there as shown in the magnified view in Fig.5. The sum of the oxide layer thickness of both surfaces reached 35% of the cladding thickness.

In the NSRR quench tests, cladding tube was broken into several fragments during quenching when the cladding temperature exceeded 1500K and the cladding was oxidized to the extent equivalent to 35% oxidation of cladding thickness after occurrence of ballooning. On the other hand, it did not fail in case of the temperature of around 1300K and oxide layer thickness of 10%. These results are in agreement with that of out-of-reactor tests such as Chung's study[4] which was obtained under isothermal oxidation conditions with no constraint force. After fragmentation of cladding tube, fuel pellets were exposed to the environment at the broken part of the cladding tube. This suggests a possibility of the enhancement of fuel degradation in the case of irradiated reactor core with cracked fuel pellets.

In this test series, some other tests have already been conducted and detail information will be obtained after post test examinations.

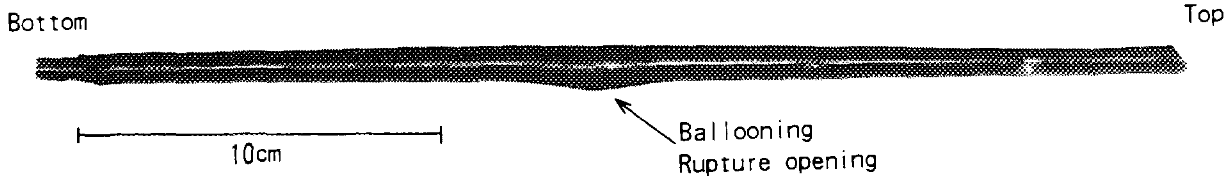
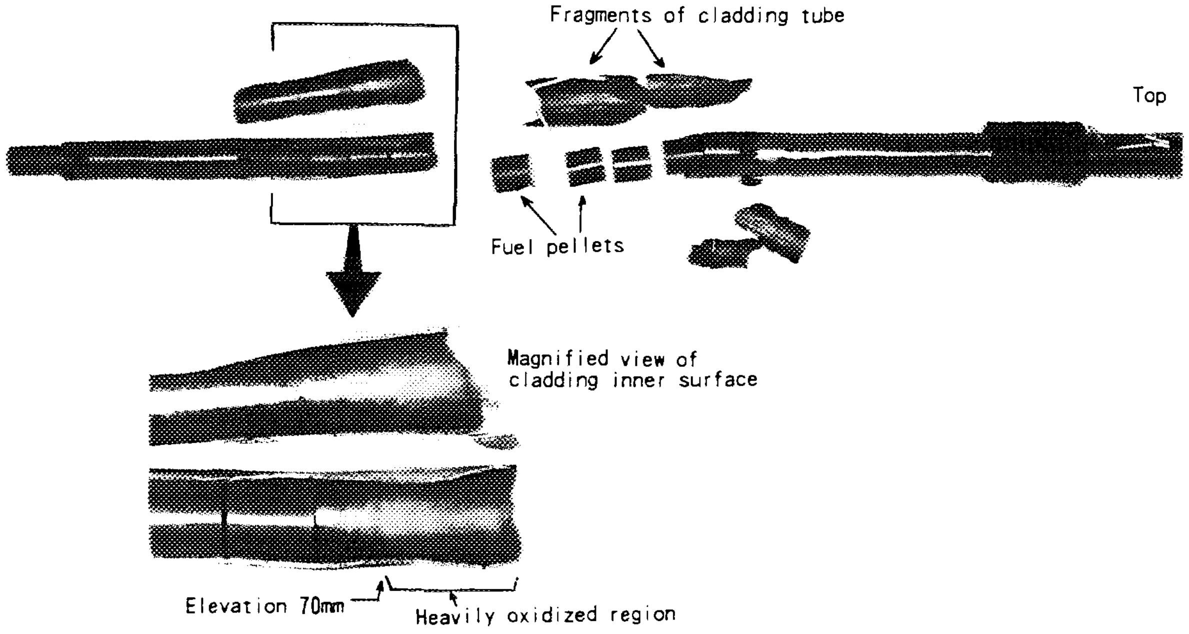


Fig. 4 Post test appearance of the fuel rod after test no. 952
(Max. cladding temperature:1300K, Oxide layer thickness: ~40μm)



'Fig. 5 Post test appearance of the fuel rod after test no. 954
(Max. cladding temperature:1533K, Oxide layer thickness:~200μm)

3. High Temperature Behavior of Fuel and Core Materials

The core of an LWR consists of many materials including UO_2 fuel, Zircaloy cladding, stainless steel, Inconel, and various control rod materials. The high temperature expected in a severe accident would cause fuel failures and core component material interactions. Therefore, extensive information concerning the high temperature behavior and interaction of core materials is required to evaluate and analyze the progression and extent of core damage.

The large scale bundle tests such as PBF-SFD, LOFT/LP-FP-2, PHEBUS-SFD, ACRR-DF, CORA etc. have been performed in many countries. In these experiments, the fuel bundles composed of UO_2 fuel, Zircaloy cladding, spacer grids and control rods were heated up to very high temperatures above 2100K. The experiments have produced significant information that is essential to understand the phenomena that occur in fuel assemblies under a severe accident condition.

In parallel to the large scale bundle tests, out-of-pile separate effect tests focused on the individual interaction between core materials have been performed at Forschungszentrum Karlsruhe(FZK) and JAERI. These tests have aimed to obtain more quantitative information for

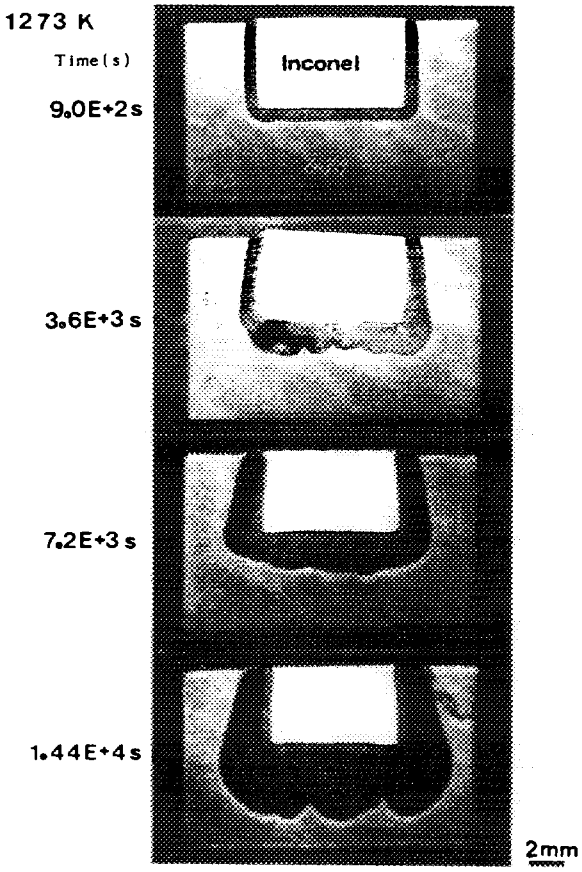


Fig. 6 Macrophotographs of Zry/Inconel diffusion couples isothermally heated at 1273K

Table 2 Kinetics for materials interaction measured at JAERI

Reaction System	Temperature Range (K)	Kinetics $K(m^2/s)=K_0 exp(-Q/RT)$	Applicable Temperature Range (K)
Metal / Metal			
Zry-4/Inconel-718	1248 - 1523	$K=2.69\times10$ exp(-259/RT)	1248 - 1423
		$K=5.33\times10^{-1}$ exp(-192/RT)	1473 - 1523
Zry-4 /SS-304	1273 - 1573	$K=3.11\times10^{9}$ exp(-496/RT)	1273 - 1573
Zry-4/Ag-In-Cd	1273 - 1473	$K=6.30\times10^{3}$ exp(-331/RT)	1273 - 1473
Metal / Ceramics			
SS-304/B_4C	1073 - 1623	$K=1.49\times10^{-2}$ exp(-250/RT)	1073 - 1473
		$K=1.40\times10^{10}$ exp(-549/RT)	1498 - 1623
Zry-4/B_4C	1173 - 1953	$K=2.42\times10^{-8}$ exp(-173/RT)	1173 - 1773
		$K=8.79\times10^{43}$ exp(-1965/RT)	1498 - 1623
Zry-4/(20wt%-B_4C+ 80wt%-SS-304)	1473 - 1923	$K=7.04\times10^{-9}$ exp(-41.3/RT)	1473 - 1673
		$K=7.01\times10^{10}$ exp(-666/RT)	1773 - 1923
Fuel / Metal			
UO_2/(Zry-4+silver)	1473 - 1703	$K=5.98\times10^{-12}$ exp(-34.9/RT)	1473 - 1573
		$K=2.10\times10^{-10}$ exp(-675/RT)	1618 - 1703

Kinetics were measured for decrease in Zry thickness (ex.SS-304 in SS-304/B_4C, UO_2 radius in UO_2/(Zry+silver) reaction), R=8.314 J/mol/K, Q:activation energy in kJ/mol

evaluating the phenomena observed in the large scale bundle tests and TMI-2 accident. The experiments have been conducted under well-defined conditions and reaction rates have been evaluated based on the measurement data. Many reaction systems, Zircaloy/Inconel, Zircaloy/SS, Zircaloy/Ag-In-Cd, SS/B_4C etc. were investigated in the temperature range from 1073 to 1953K at JAERI. Figure 6 shows an example of Zircaloy/Inconel interaction at 1273K. To estimate the reaction rate, the extent of reaction was measured as the thickness of reaction layer formed at the interface of reaction couple and/or the decrease in the material thickness by the reaction. The reaction approximately obeyed the parabolic rate law in every case of reaction system in the examined temperature range, so that the parabolic rate law constant for each system was determined and the Arrhenius equation was obtained. The kinetics data obtained at JAERI and the applicable temperature range is summarized in Table 2. The reaction rates for the various reaction systems are compared with each other in the Arrhenius type plotting in Fig.7. The reaction between materials used in the core of PWR, Zircaloy/Inconel and Zircaloy/Ag-In-Cd, showed the highest rates. The drastic change in the reaction rate at certain temperature level seen for some reaction systems indicates the change of reaction mechanism and this could be related to the eutectic formation occurs at this temperature. For the some reaction systems, the effect of oxidizing atmosphere on the reaction rate was partly investigated and the qualitative information was obtained, which indicated the increase in the initiation temperature of eutectic formation.

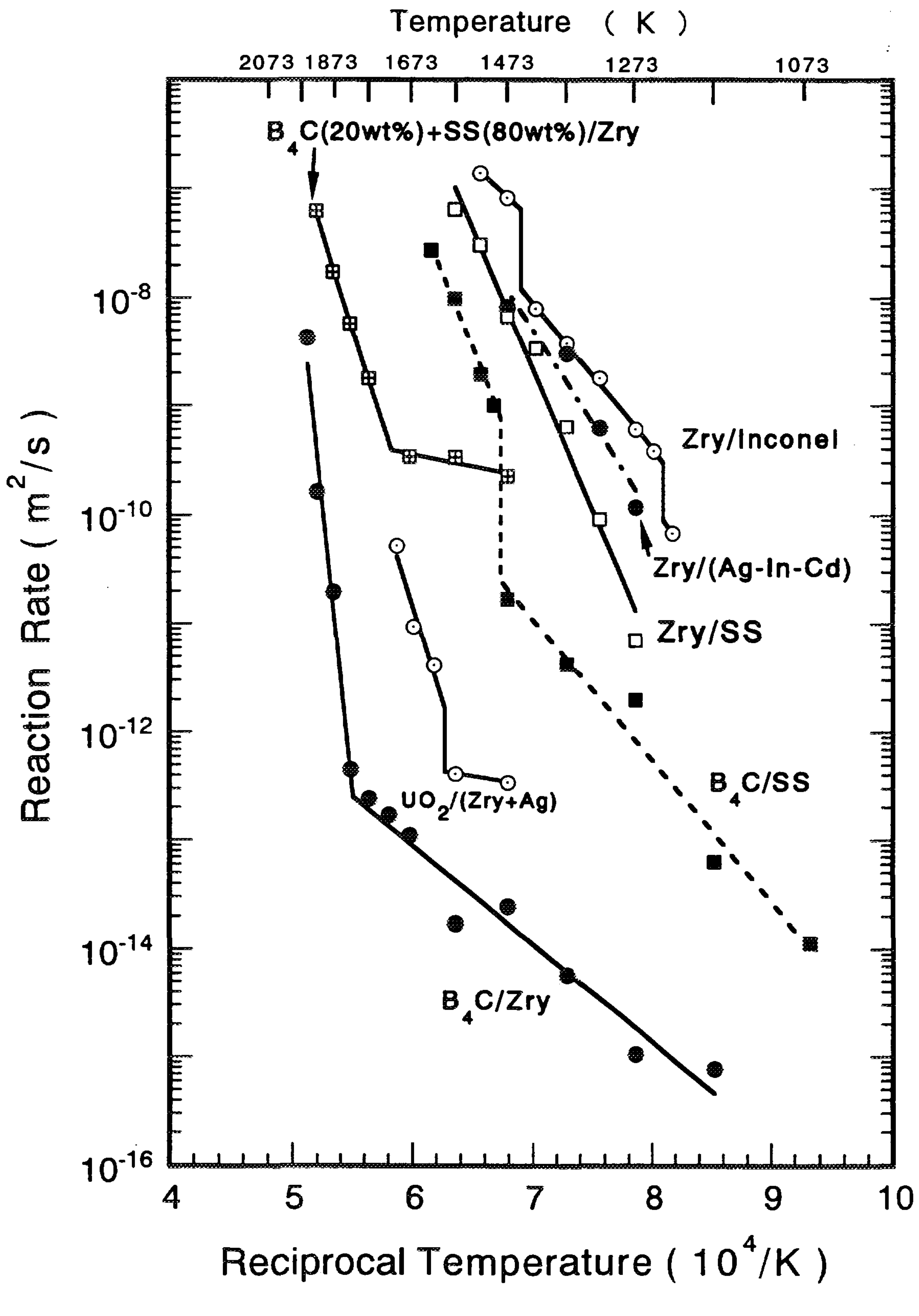


Fig. 7 Comparison of reaction rates between core materials measured at JAERI

The obtained kinetics has been incorporated into computer codes such as ICARE2 which predict the fuel degradation such as melt formation and progression in the bundle under a severe accident condition.

4. TMI-2 debris examination

The TMI-2 reactor accident in March 1979 resulted in severe fuel damage, including melting, interaction and relocation of reactor core materials. About 19 tons of molten core material was assessed to have relocated onto the lower head of the reactor vessel during the accident. JAERI has participated in the TMI-2 Vessel Investigation Project conducted by the OECD-NEA and has obtained sixty samples including ten companion samples from locations adjacent to the lower head. Many examinations and analyses such as ceramography, SEM/EPMA analysis, γ-ray spectrometry and thermal property measurement are being performed at the Reactor Fuel Testing Facility(RFTF) of JAERI. These examinations are being performed to obtain behavioral data for fuel and core materials under LWR severe accident conditions.

Visual examinations indicated that the samples were composed primarily of once molten ceramics. They were generally dull grey in appearance. The histogram of debris density for the 30 measurements data is shown in Fig.8. The density ranged from 6.3 to 9.3g/cm^{3}and the average

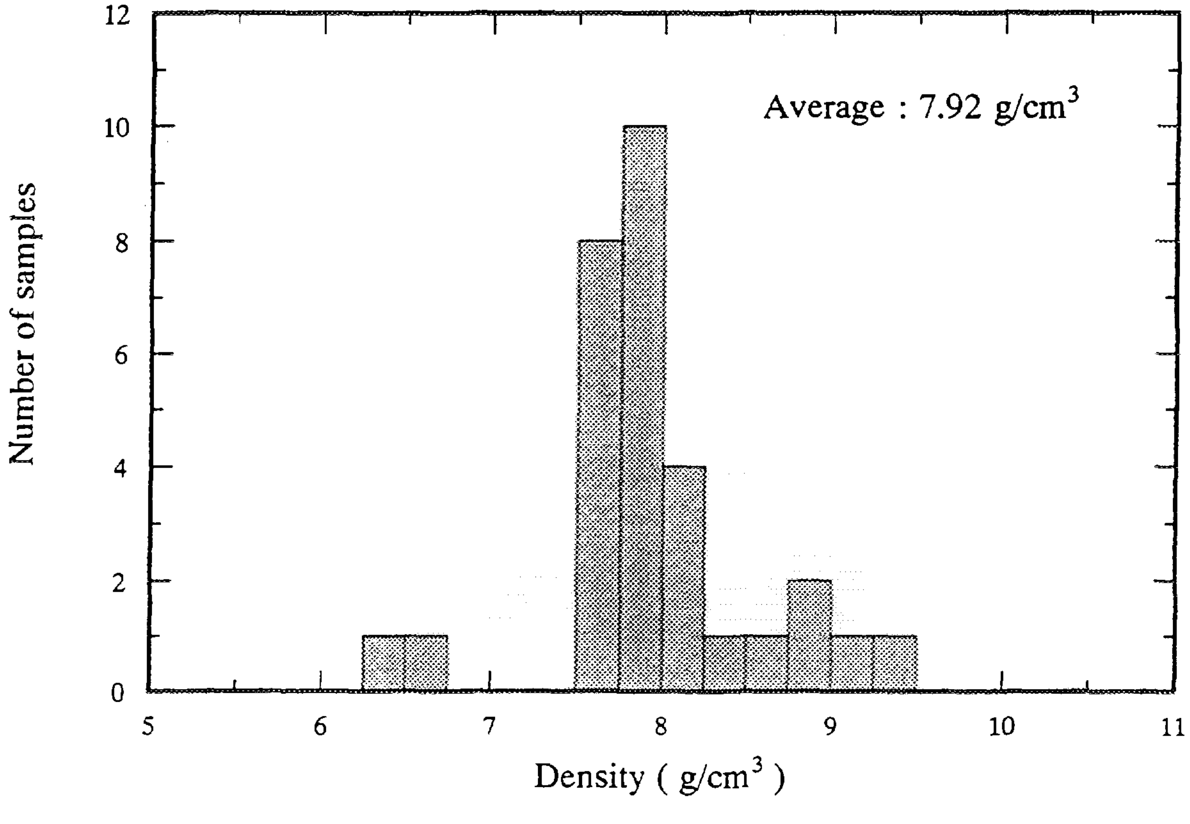


Fig. 8 Histogram of debris density

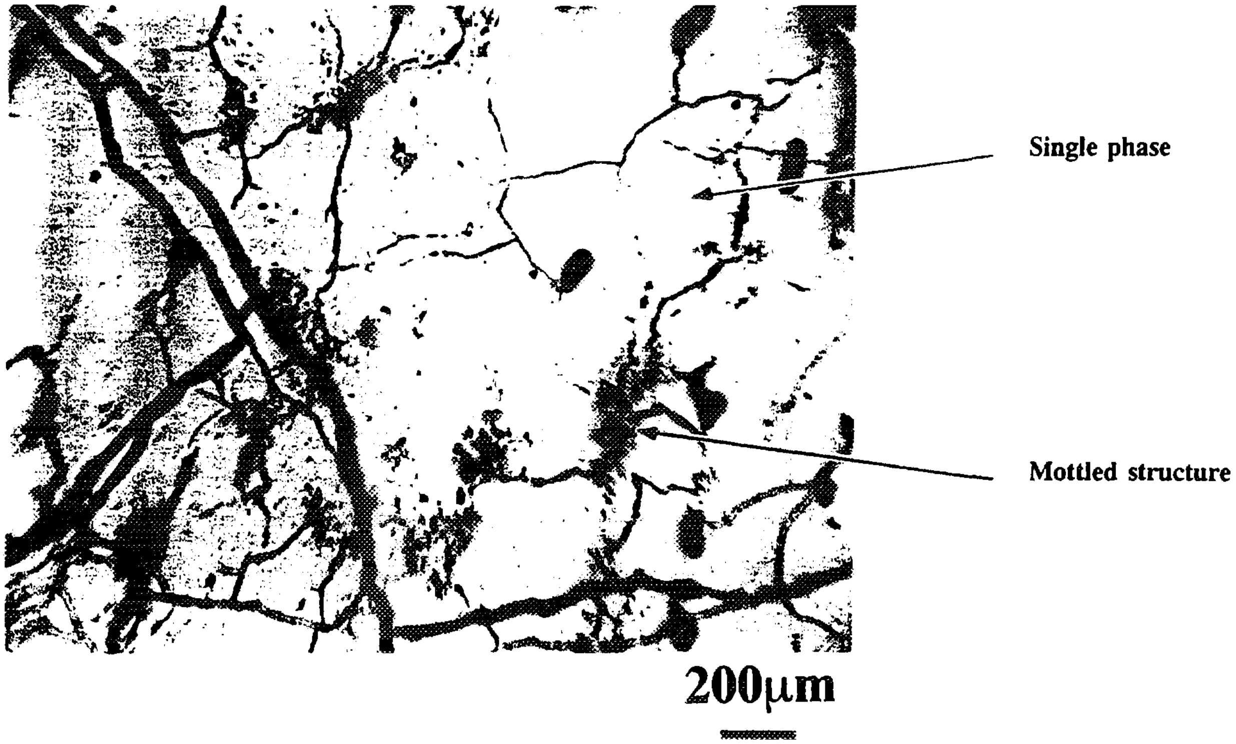


Fig. 9(A) Microstructure of ceramic debris(VIP-12A)

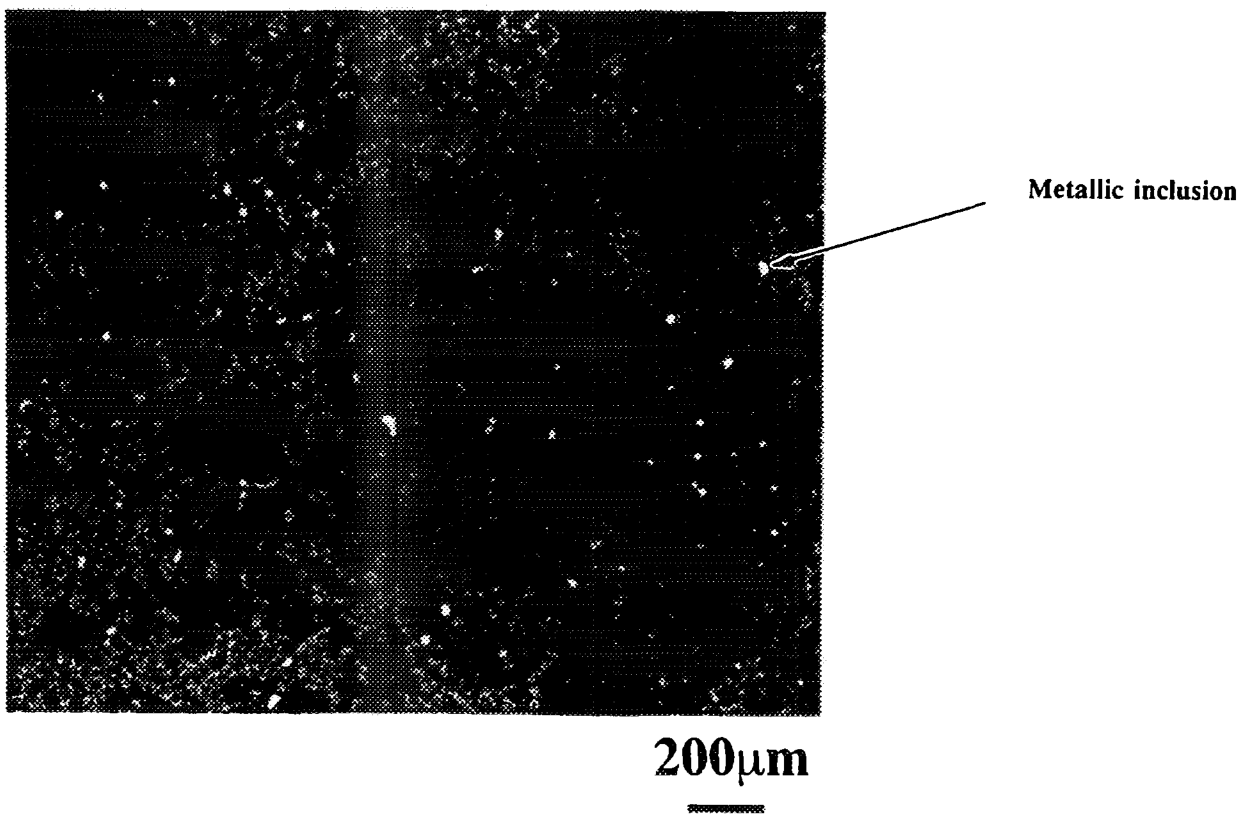


Fig. 9(B) Metallic inclusions dispersed in ceramic debris(VIP-12A)

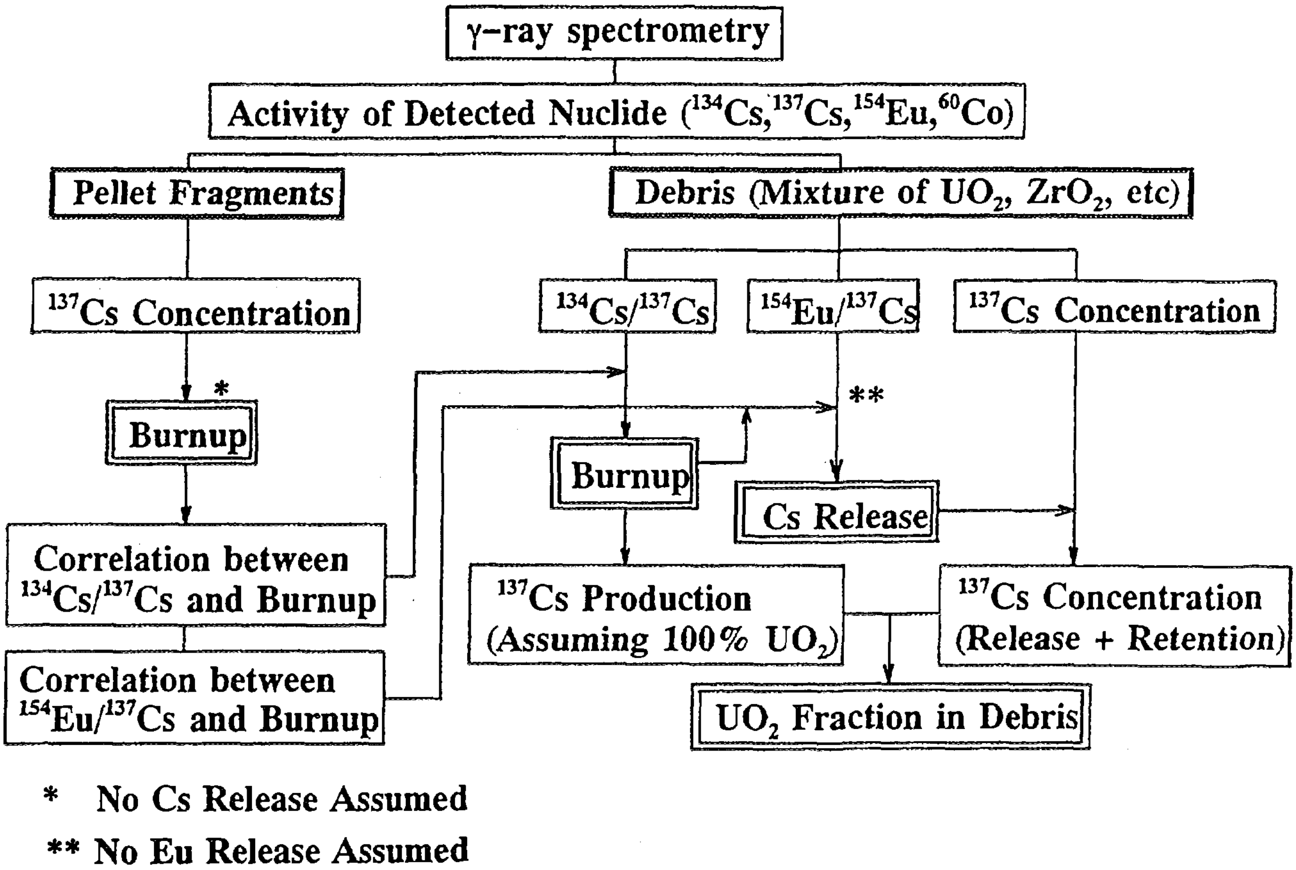


Fig. 10 Flow diagram of estimation for Burnup and Cs retention of TMI-2 debris

Table 3 Burnup, Cs retention and UO_2 fraction in debris

Sample	*Burnup [MWd/t]*	*Cs Retention [%]*	*UO_2 Fraction [%]*
E9-4	*3300*	*0.4*	*64.5*
H8-1	*3700*	*4.2*	*72.5*
VIP-9H-a	*3500*	*5.3*	*79.4*
VIP-9H-b	*3500*	*3.3*	*83.3*
VIP-10C-a	*3600*	*3*	*76.7*
VIP-10C-b	*3600*	*5.9*	*73.9*

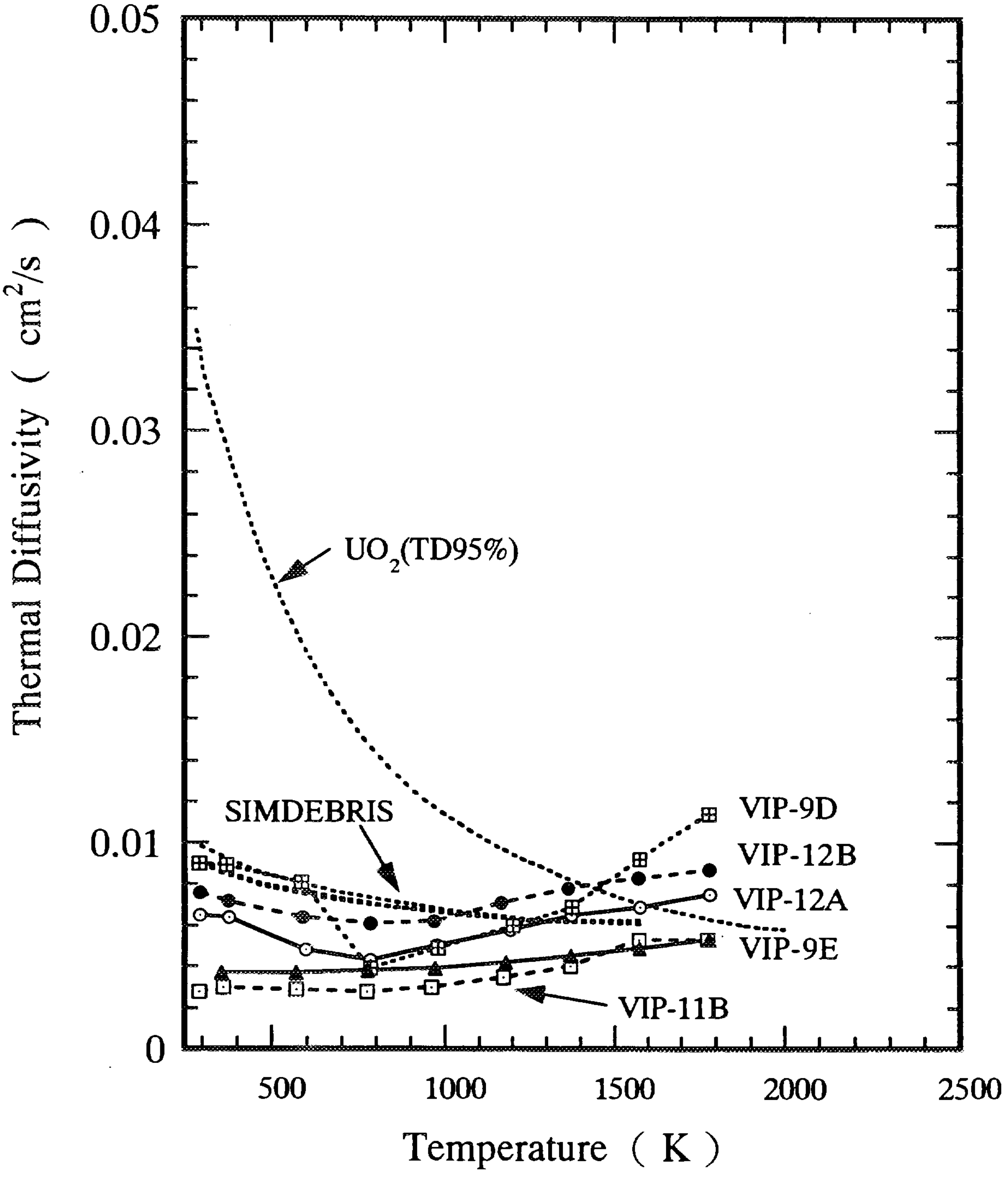


Fig. 11 Thermal diffusivity of TMI-2 debris measured with the laser flash method

density was about 7.9g/cm^3. Figure 9(A) is a high magnification photograph that shows the typical microstructure of ceramic debris. The structure is composed of both the clear single-phase region and the mottled region. The SEM/EPMA analysis revealed that the single-phase was uranium-rich $(U,Zr)O_2$ phase and the secondary phase consisted of microporosity and ceramic two-phase structure, uranium-rich $(U,Zr)O_2$ phase and zirconium-rich $(Zr,U)O_2$ phase. The metallic inclusions were sometimes found in the ceramic debris as shown in Fig.9(B). These melts are probably silver-rich metallic particles.

In the γ-spectrometry, four nuclides of Cs-134, Cs-137, Eu-154 and Co-60 were detected for each sample. The retention of Cs, burnup of fuel and fuel fraction in debris were estimated based on the measurement data and by the method shown in Fig.10. The results of present estimate are summarized in Table 3. The burnup of fuel contained in debris was estimated to be in the range of 3,300 - 3,700MWD/t. The retention of Cs in debris was quite low, it ranged from 0.4 to 5.9%. This indicates debris experienced very high temperature during the accident. In the experiments at ORNL, 37% Cs retention was measured after the heating at 2300K for 60min in steam. The UO_2 fraction in debris was estimated in the range between 64.5% and 83.3%.

Thermal diffusivity of debris was measured with a LASER flash equipment in the temperature range from room temperature to 1773K. Fig.11 summarizes the measurement data showing the correlation between the thermal diffusivity and the temperature. The data of 95%-TD UO_2 and simulated fuel debris(SIMDEBRIS) having similar chemical composition to TMI-2 debris is also plotted in the figure for comparison. The thermal diffusivity of debris is much lower than that of UO_2 at relatively low temperature range, while the data becomes comparable each other in the highest range of examined temperature. There was large difference in diffusivity among debris samples. It was confirmed that the difference in thermal diffusivity between samples was caused by the difference in chemical composition and microstructure.

5. Summary and future plan

To investigate the core materials behavior under severe accident conditions, fuel quench tests at the NSRR, core materials interaction tests at high temperature and TMI-2 debris examination were conducted at JAERI. Experimental data to determine the boundary condition of the fuel fracturing upon quenching have been obtained through fuel quench tests. Chemical interactions have been widely investigated at high temperatures for various binary systems of core component materials. Also, valuable information on the TMI-2 debris was obtained through debris examination. The NSRR quench test program will be finished after information on the additional oxidation by steam generation during reflooding is obtained through post test examination. The experiments on the materials interaction and TMI-2 debris examination are planned to be finished in this year, 1995.

As a next phase of the experimental study on material behavior at JAERI, Verification Experiment of Gas/Aerosol Release(VEGA program) is planned. Objective of this program is to investigate the fission product(FP) release behavior and materials interactions by heating tests of burnup fuels. JAERI's participation in the international collaboration of Phebus-FP program will be continued in order to obtain the information from large scale experiments on FP behavior under severe accident conditions.

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