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HTGR FUEL BEHAVIOR AT VERY HIGH TEMPERATURE

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(Received February 5, 1986)

Fuel behavior at very high temperature simulating abnormal transient of the reactor operation and accidents have been investigated on TRISO coating LEU oxide particle fuels at JAERI. The test simulating the abnormal transient was carried out by irradiation of loose coated particles above 1600 °C. The irradiation test indicated that particle failure was principally caused by kernel migration. For simulation of the core heat-up accident, two experiments of out-of-pile heating were made. Survival temperature limits were measured and fuel performance at very high temperature were investigated by the heatings. Study on the fuel behavior under reactivity initiated accident was made by NSRR(Nuclear Safety Research Reactor) pulse irradiation, where maximum temperature was higher than 2800 °C. It was found in the pulse irradiation experiments that the coated particles incorporated in the compacts did not so severely fail unlike the loose coated particles at ultra high temperature above 2800 °C. In the former particles UO_2 material at the center of the kernel vaporized, leaving a spherical void.

Keywords: Coated Particle Fuels, Very High Temperature, JMTR, Out-of-pile Heating, NSRR, Core Heat-up, Abnormal Transient, Reactivity Initiated Accident.

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超高温下における高温ガス炉燃料挙動

日本原子力研究所東海研究所燃料工学部
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（１９８６年２月５日受理）

高温ガス実験炉の運転及び事故時の異常な過渡変化を模擬した超高温下の燃料挙動を，原研が開発しているＴＲＩＳＯ被覆，低濃縮酸化物粒子燃料について調べた。異常な過渡変化を模擬した試験は，ルーズな被覆粒子を１６００℃以上で照射することにより行った。照射試験の結果，粒子破損は大部分が燃料核移動によるものであった。炉心昇温事故を模擬した試験としては，二種の炉外加熱試験を行った。加熱により耐熱限界温度の測定と超高温下での燃料挙動を調べた。反応度事故時の燃料挙動の研究は，ＮＳＲＲ（Nuclear Safety Research Reactor）によるパルス照射により行い，この時の最高温度は２８００℃以上であった。パルス照射試験では，コンパクトに成形した被覆粒子は２８００℃以上の超高温でも，ルーズな被覆粒子にみられた非常にはげしい破損はみられなかった。コンパクトに成形した粒子では燃料核の中心でUO_2が蒸発し，球状ボイドを呈していた。

東海研究所：茨城県那珂郡東海村白方字白根２の４

INTRODUCTION

Under abnormal occurrences of HTGR operation, coated particle fuels are exposed to very high temperature circumstance. Such events include an abnormal transient of the operation, core heat-up accidents caused by e. g. a loss of forced coolant, and a reactivity initiated accident by ejection of control rods. Although current HTGR designs introduce the passive cooling concept for the accident where the maximum core temperature by gamma-ray heating is about 1600°C, knowledge on fuel behavior at very high temperature is still significant for safety analysis in HTGR design.

Concerning performance of TRISO coated particle fuels at very high temperature, a core heat-up simulation test (CHST) had been carried out at GAT(1,2). High temperature fuel behavior was studied on BISO and TRISO coated particles at 2300°-2500°C at KFA(3). At JAERI, TRISO coated LEU oxide particles have been tested under abnormal conditions covering the three occurrences mentioned above, by capsule irradiation, pulse reactor irradiation and out-of-pile heatings for unirradiated fuels.

EXPERIMENTS

The present study is summarized in Table 1, where abnormal occurrences are classified in three categories. The abonormal transient of the operation was simulated by irradiation in JMTR(Japan Material Testing Reactor). The coated particles were loosely loaded in the capsules and irradiated in the temperature range between 1000°C and 1600°C. Temperature during irradiation was increased up to 1600°-1800°C and kept there for a short time. In the PIE, the coated particles were examined by visual inspection,

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The fuel behaviors at very high temperatures simulating the core heat-up accident and the reactivity initiated accident were investigated by out-of-pile heating and pulse irradiation in NSRR, respectively.

Two experiments for out-of-pile heating were carried out in the temperature range between 1800°C and 2600°C for three forms of unirradiated fuels; loose coated particles, fuel compacts which incorporated the coated particles with the graphite matrix, and minuature fuel rods where a fuel compact was contained in a graphite sleeve.

Coating failure during heating in Experiment II(Table 1) was monitored by variation of carbon mono-oxide (CO) concentration released from failed particles into the sweep gas through the furnace. Post-heating measurements on coating failure, on the other hand, were carried out by visual inspection, ceramography, X-ray microradiography and EPMA(Electron Probe Micro Analysis).

The NSRR pulse irradiation experiments were made by total of six capsules as comprized in Table 1, which loaded either the loose coated particles or the compacts. Due to pulse irradiation, duration at very high temperature lasted for only 10 msec or less, but maximum temperature exceeded 2800°C, a melting point of UO_2. For the examination of the fuels irradiated in NSRR, visual inspection, ceramography, acid leaching and EPMA were carried out.

RESULTS and DISCUSSION

Irradiation experiments simulating abnormal transient

Fig. 1 shows the coated particle performance during irradiation simulating the abnormal transient of the reactor operation. In this figure, 95% confidence limits of failure fractions measured by X-ray radiography and visual inspection, and the averaged failure fraction measured by acid-leaching are illstrated together with temperature variation of each coated particle sample. Although duration at peak temperature lasted for only several hours, the coated particles failed with high fraction ranging from $10^{-3}$to 10^{-1}. For instance, the coated particles irradiated at peak temperature of 1810°C remained in order of 10^{-2}. Particle C prepared in a different method from Particles A and B indicated relatively high failure fraction.

Surface appearance and ceramographs of the coated particles are presented in Fig. 2. It was found by ceramography that most of the coated particles were failed by kernel migration(amoeba effect). It is seen in the surface appearance that outer PyC layer was suffered by the kernel migration. Besides such failure manner, mechanically failed particles having coating cracks were found. This is discussed later. The SiC layer in the coated particles did not show significant deterioration.

Out-of-pile heating simulating core heat-up

Fig. 3 shows variations of failure fraction of three forms of the unirradiated fuels during isothermal heating at about 2400°C. Seen in this figure is that the loose coated particles are the most undurable among the fuels, whereas the fuel rod keeps integrity for the longest time. This fuel behavior would be explaied as follows; due to decomposition of UO_2, resulting in burst release of CO gas at very high temperature(3), some of the coated particles balloon as shown in Fig. 4. This causes swelling of the compacts, thus affecting stress onto the graphite sleeve. In other words, the coated particles are protected from ballooning or burst by mechanical interaction with the graphite matrix and the sleeve, resulting in increase of durability of the coated particles in the compact and all the more in the fuel rod. When the coated particles which were loaded loosely in a long holes of the graphite holder in the capsule, were irradiated at very high temperature, ballooning might cause the mechanical interaction between the coated particles, thus leading to failure of the coatings. Cracks shown in Fig. 2 is probably the typical mechanical interaction caused by ballooning.

Isothermal heatings for both unirradiated loose coated particles and compacted ones were conducted at various temperatures ranging from 1800°C to 2600°C. The results are given in Fig. 5. Survival temperatures of the loose coated particles and compacted ones were about 2400°C and 2500°C on a conservative estimation, respectively, where the compacted particles indicated better durability than the loose particles. These temperatures were not so greatly different from that measured in US, 2660°C(2).

Ceramography and EPMA on the coated particles after heating are given in Fig. 6 as typical examples. The coated particles heated at 2400°C(Fig. 6(A)) exhibited significant deterioration of SiC layer as seen in the ceramographs, but Si image of EPMA did not indicate great dispersion of Si atom from SiC layer to the other layers. By heating at 2600°C(Fig. 6(B)) the coated particles had failed before SiC layer began decomposition. EPMA showed that Si atom did not disperse from SiC layer, although Si atom migrate to some extent into the kernel material from a spot where SiC layer and the kernel material interacted. By this experiment it was found that particle failure was characterized by thermal decomposition of SiC layer at temperature less than 2600°C as reported by GAT and KFA(3,4), followed by ultimate failure of the particles, while the coated particles failed by burst at 2600°C before SiC thermal decomposition. In this case the inner CO pressure taking part in the burst was supposedly about 10-20 MPa in the coated particles under equilibrium(5).

Pulse irradiation in NSRR

Appearance of the loose coated particles after pulse irradiation in NSRR are shown in Fig. 7. It is clearly seen that the particle failure depends on heat generation in the coated particles. The particles remained intact at the heat generation less than 900 J/gUO_2, cracks appeared in some of the coated particles at 1700 J/gUO_2, failure was seen in most of the coated particles at 2500 J/gUO_2, and all of the coated particles failed at 4000 J/gUO_2. Appearance of the fuel compacts after irradiation, on the other hand, is somewhat different as seen in Fig. 8. The fuel compacts irradiated at the heat generations of 2800 and 3400 J/gUO_2 cracked, while the compacts irradiated at less than 2000 J/gUO_2 remained intact. Ceramography of the coated particles in the compacts given in Fig. 9 indicates that the coated particle irradiated at the heat generation less than 2800 J/gUO_2 does not exhibit significat change, while at the heat generation higher than 2800 J/gUO_2 the kernel of the coated particles vaporize at the center region, leaving a spherical void. Therefore, it is certain that temperature at the center exceeded fully 2800°C, a melting point of UO_2. Comparison of the fuel behavior of the coated particles in the compacts with the loose coated particles, indicates that coating damage in the compacted particles is less severe by protection by the graphite matrix.

For the coated particles in the compacts irradiated at the heat generation of 3400 J/gUO_2, ceramography by polarized light, OPTAF(Optical Anisotropy Factor) measurement and EPMA were made. The typical results are shown in Fig. 10. Polarized light observation did not reveal any significant changes in the coatings. Uranium image of EPMA showed that small amount of uranium was detected in a coating crack and the boundary between the particle and the graphite matrix. However, most of uranium equivalent to the center void volume of UO_2 kernel was not seen in the graphite matrix by this observation. It is supposed that most of UO_2 material effuse as gas into the matrix through coating cracks locating in different direction to this sectioned surface. SiC layer did not decompose as seen in Si image which implys that temperature of SiC layer was not so high as that in the out-of-pile heatings.

OPTAF measurements on inner and outer PyC layers of the coated particles yielded OAF values as 1.02 and 1.01, respectively,while pre-irradiation OAF value for both PyC layers was 1.01, thus showing no significant change in preferred orientation of crystals in the PyC layer. It means that the PyC layers were not exposed also to very high temperature , because PyC would become anisotropic under stress field originated in inner gas pressure at very high temperature.

SUMMARY

From the experiments simulating HTGR abnormal conditions, the fuel behavior at very high temperature was summarized in the following way. The particle failure originated predominantly from decomposition of SiC layer in the temperature range from 1600°C to 2500°C. However, when the fuel was irradiated under temperature gradient above 1600°C, the kernel migration would play an important role for the failure. Above 2600°C, the failure was attributed to burst of the particles, where SiC layer did not decompose severely. UO_2 material at the center of the kernel vaporized at ultra high temperature above 2800°C, any significant changes in the coatings. Uranium image of EPMA showed that small amount of uranium was detected in a coating crack and the boundary between the particle and the graphite matrix. However, most of uranium equivalent to the center void volume of UO_2 kernel was not seen in the graphite matrix by this observation. It is supposed that most of UO_2 material effuse as gas into the matrix through coating cracks locating in different direction to this sectioned surface. SiC layer did not decompose as seen in Si image which implys that temperature of SiC layer was not so high as that in the out-of-pile heatings.

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It was indicated in both the out-of-pile heatings and NSRR irradiation that the coated particles incorporated in the compacts were more durable than the loose coated particles because of protection by the graphite matrix.

ACKNOWLEDGEMENT

The authors are indebted to the Department of JMTR Project and Department of Research Reactor Operation for irradiation and post-irradiation examination, and also to Department of Fuel Safety Research for NSRR irradiation.

REFERENCES

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[2] B. I. Shamasunder, O. M. Stansfield and D. D. Jensen, "HTGR Fuel Performance Basis", GA-A 16767(1982).

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Table 1 Experiments for studying the fuel behavior at very high temperature.

<table>
<tr><th>Objective occurrence</th><th>Experimental method</th><th colspan="7">Description of experiment</th></tr>
<tr><td rowspan="6">Abnormal transient of reactor operation</td><td rowspan="6">Irradiation in JMTR</td><td>No.</td><td colspan="2">73F-12A</td><td colspan="2">73F-13A</td><td colspan="2">74F-9J</td></tr>
<tr><td>Max. temperature(°C)</td><td colspan="2">1810</td><td colspan="2">1730</td><td colspan="2">1600</td></tr>
<tr><td>Max. burnup(%FIMA)</td><td colspan="2">3.2</td><td colspan="2">4.4</td><td colspan="2">2.0</td></tr>
<tr><td>Fast neutron fluence (n/cm^2, E 29 fJ) $x10^{21}$</td><td colspan="2">1.5</td><td colspan="2">3.1</td><td colspan="2">0.7</td></tr>
<tr><td>Irradiation time (EFPD)</td><td colspan="2">63</td><td colspan="2">111</td><td colspan="2">51</td></tr>
<tr><td>Sample</td><td colspan="6">Loose coated particles</td></tr>
<tr><td rowspan="4">Core heat-up accident</td><td rowspan="4">Out-of-pile heating for unirradiated fuel</td><td>No.</td><td colspan="3">I</td><td colspan="3">II</td></tr>
<tr><td>Sample</td><td colspan="3">Loose particles, Compacts</td><td colspan="3">Loose particles Compacts, Minuature fuel rods,</td></tr>
<tr><td>Heating mode</td><td colspan="3">Isothermal</td><td colspan="3">Isothermal, Temperature ramp,</td></tr>
<tr><td>Temperature range(°C)</td><td colspan="3">1800 - 2600</td><td colspan="3">1900 - 2500</td></tr>
<tr><td rowspan="3">Reactivity initiated accident</td><td rowspan="3">Pulse irradiation in NSRR</td><td>No.</td><td colspan="3">I</td><td colspan="3">II</td></tr>
<tr><td>Sample</td><td colspan="3">Loose particles</td><td colspan="3">Compacts</td></tr>
<tr><td>Heat generation (J/gUO_2)</td><td colspan="3">900 - 4000</td><td colspan="3">1500 - 3400</td></tr>
</table>

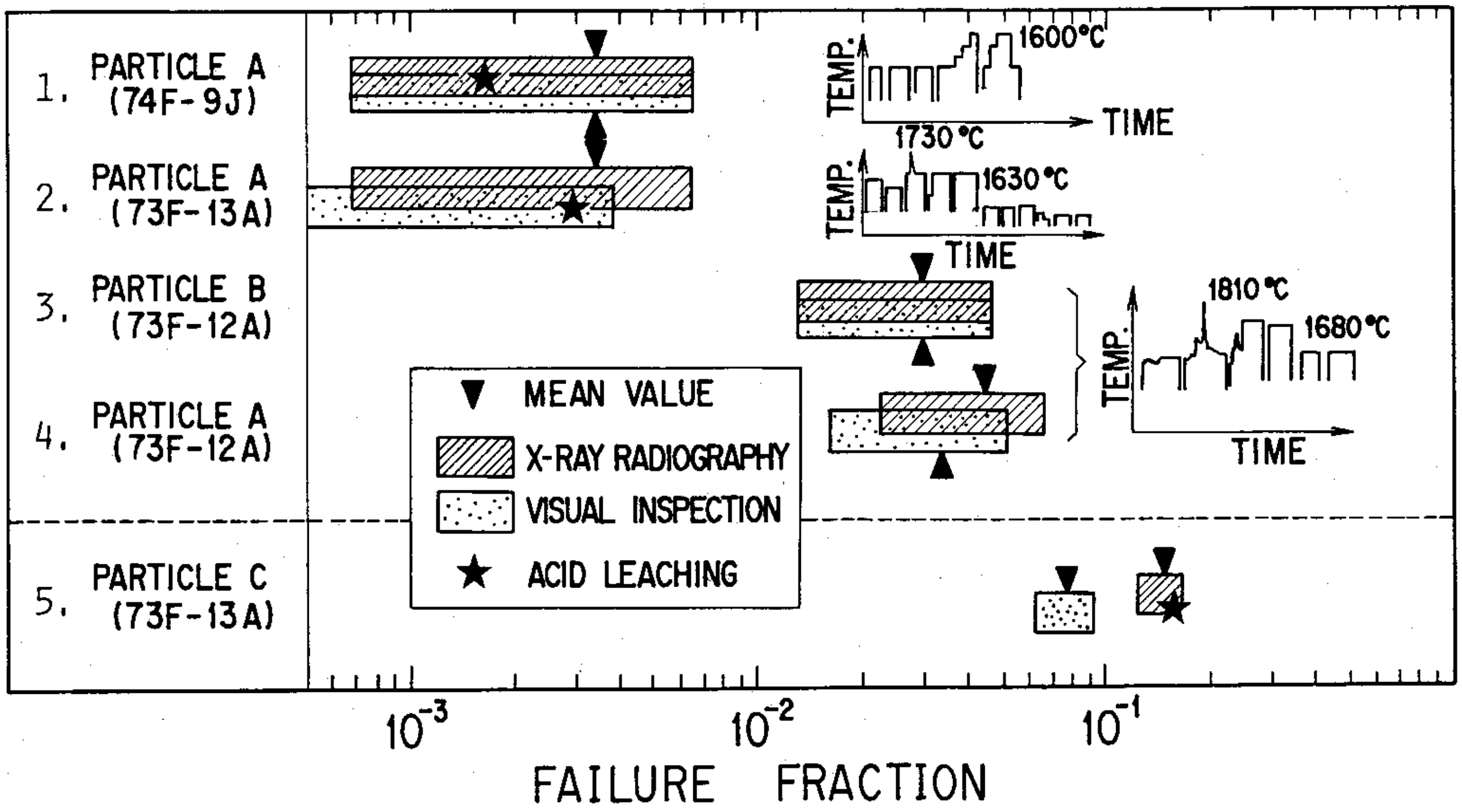
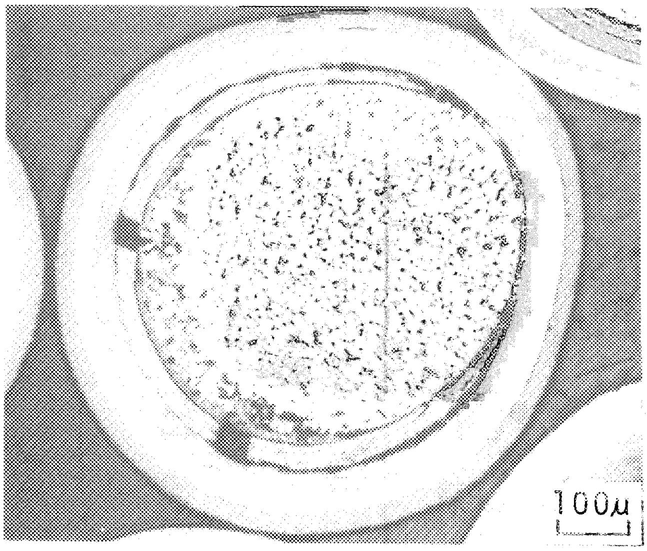
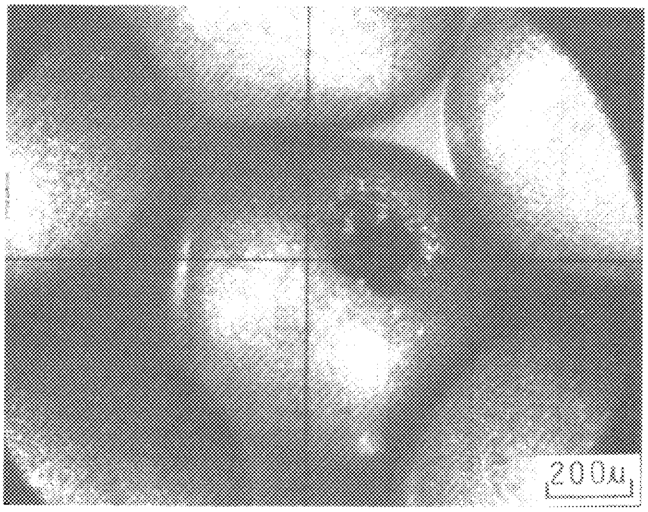


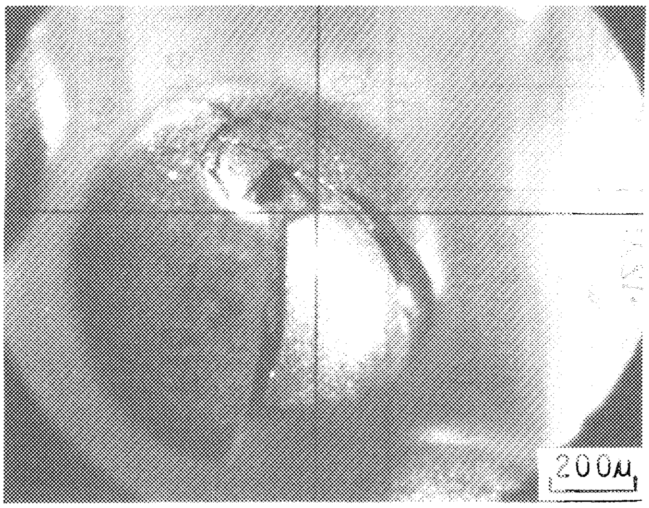
Fig. 1 Failure fraction of coated particles irradiated above 1600°C.



Kernel migration
(73F-12A, Particle B)



Kernel migration
suffering outer layer
(73F-12A, Particle B)



Coating cracks by
mechanical interaction
between particles
(73F-12A, Particle A)

Fig. 2 Post-irradiation examination of the coated particles irradiated for simulation of abnormal transient of reactor operation.

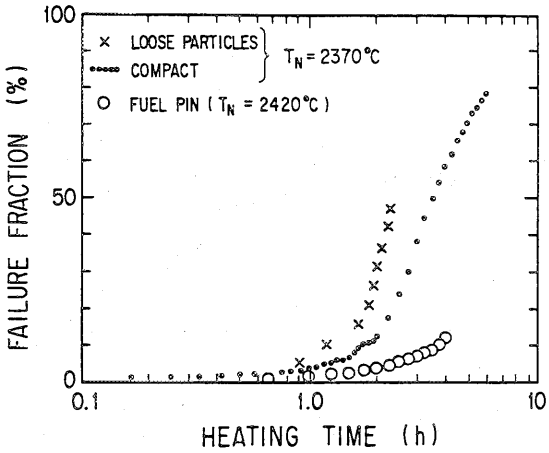


Fig. 3 Failure fraction of non-irradiated fuels heated at 2400°C.

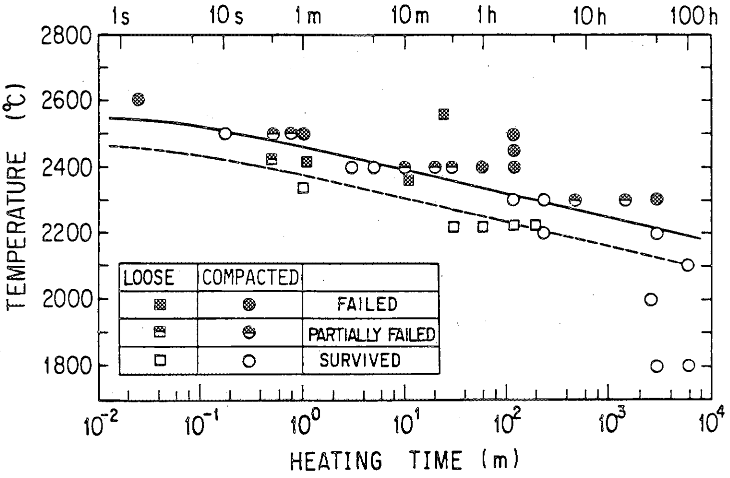


Fig. 5 Time dependent survival temperature limit.

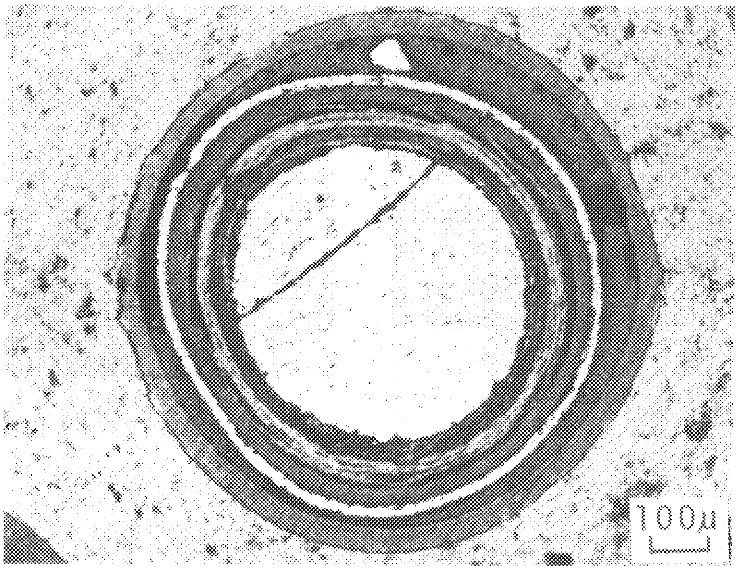
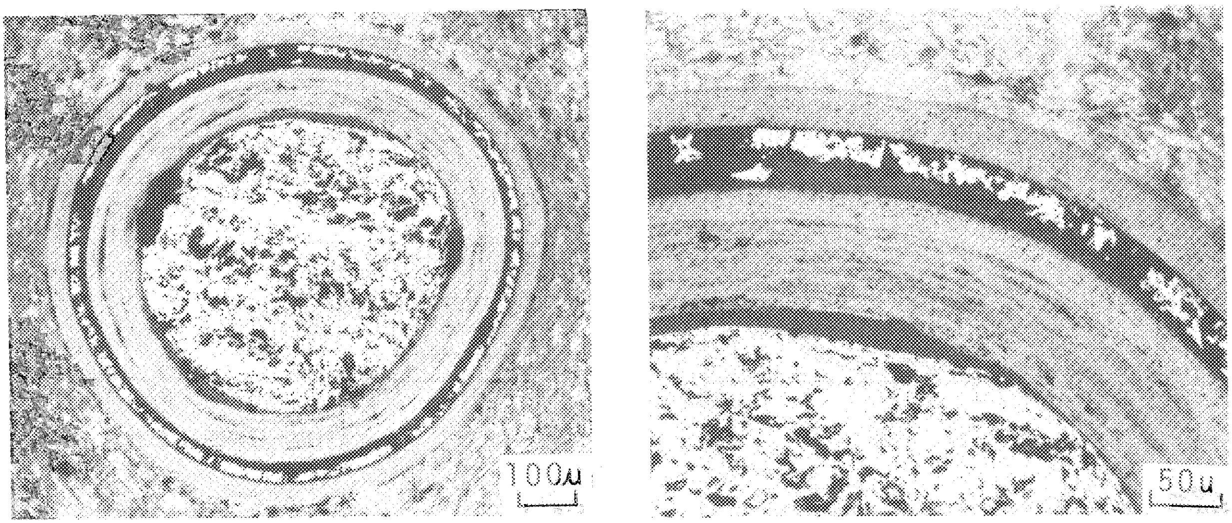
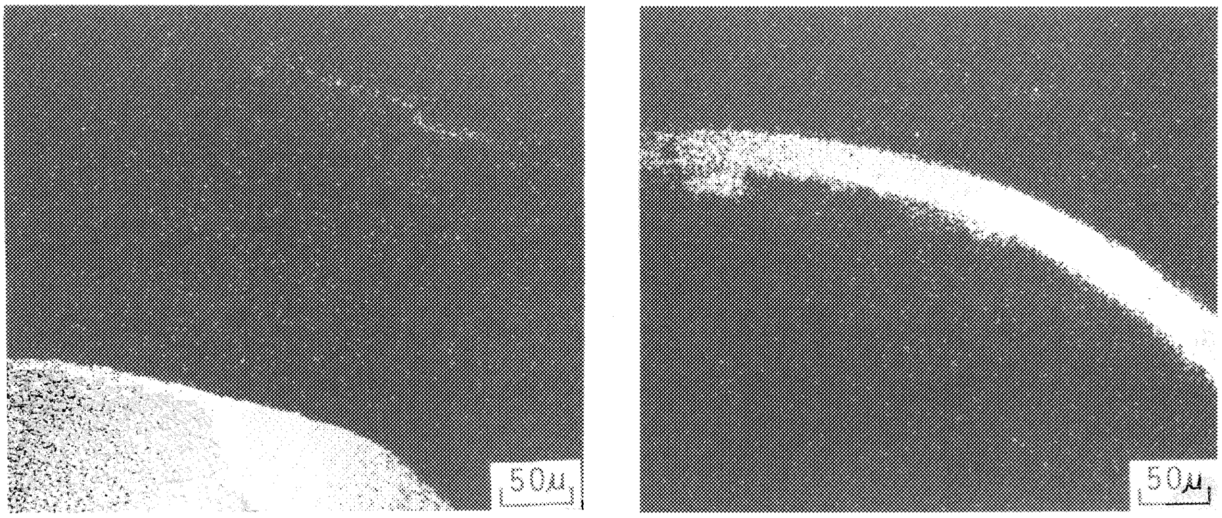


Fig. 4 Ballooning of the coated particle heated up to 2550°C.

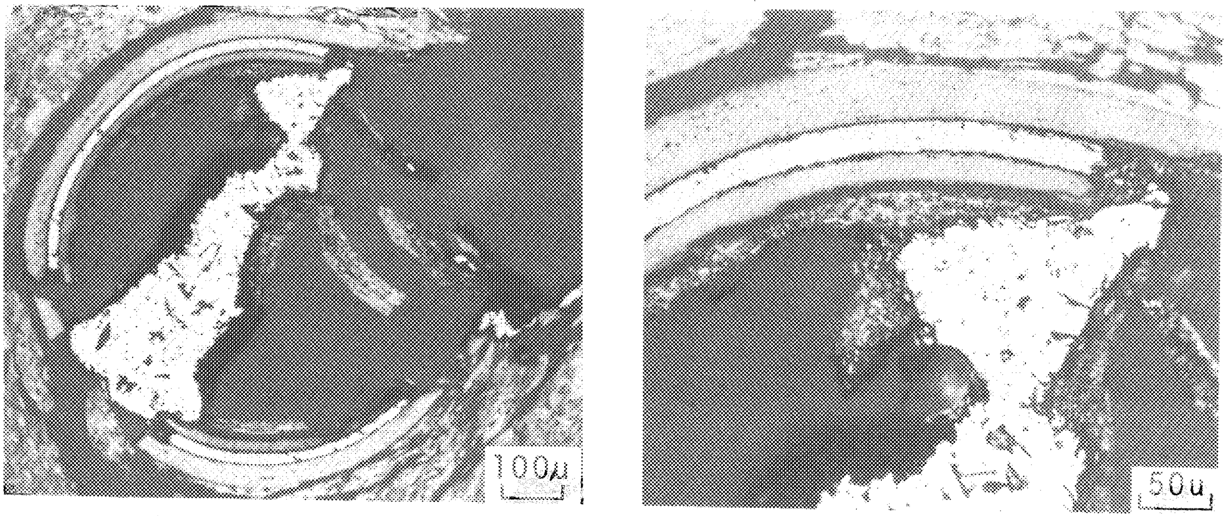


Ceramography Ceramography

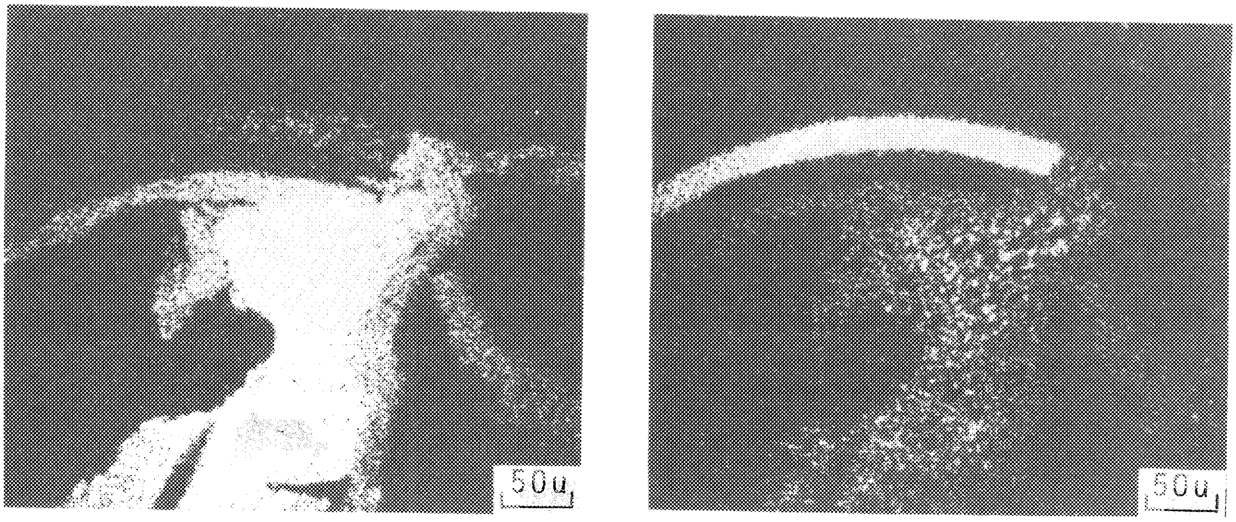


EPMA(U-Mα) EPMA(Si-Kα)

Fig. 6(A) Ceramography and EPMA of the coated particles heated at 2400°C for 5min.

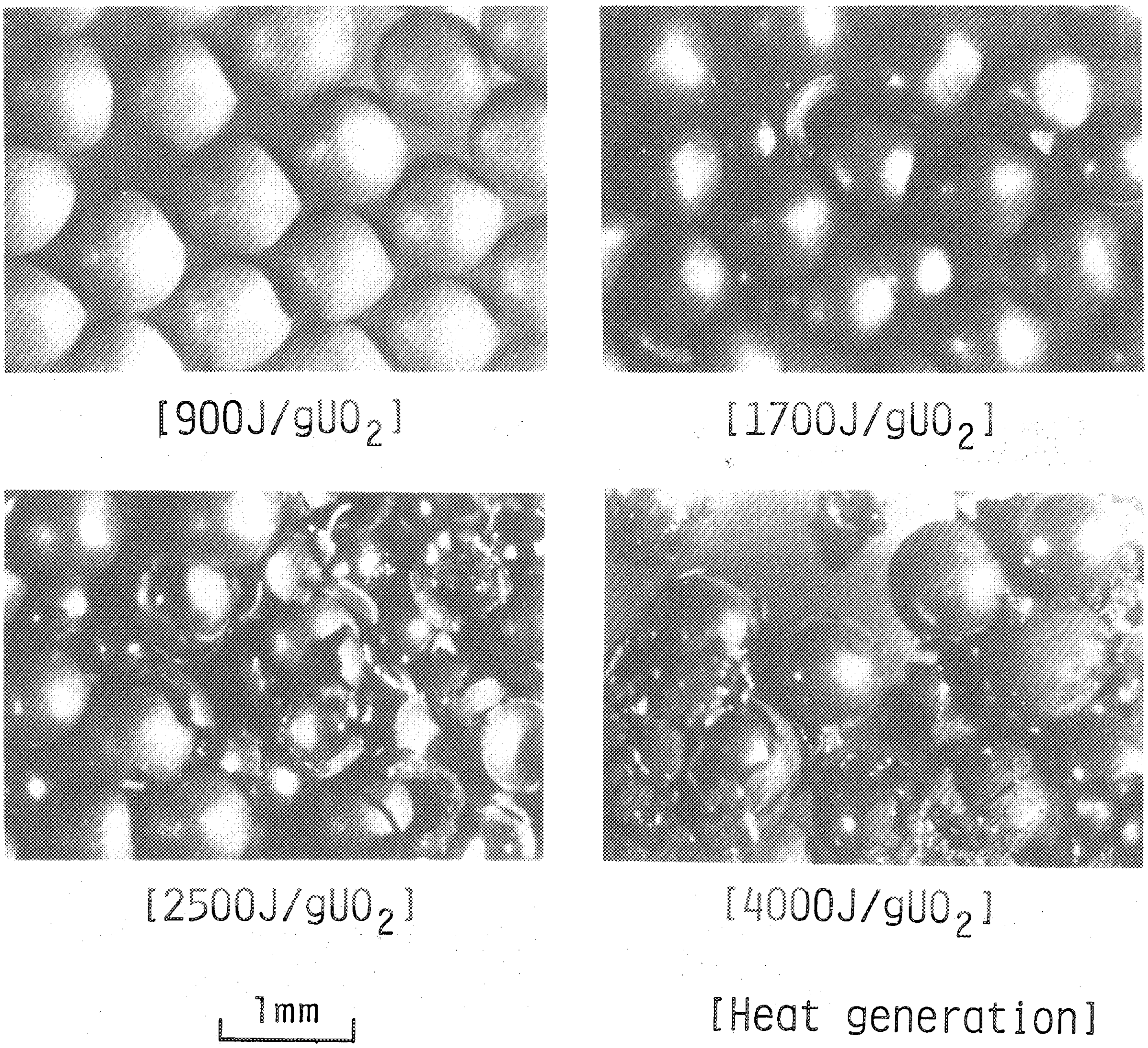


Ceramography Ceramography



EPMA(U-Mα) EPMA(Si-Kα)

Fig. 6(B) Ceramography and EPMA of the coated particles heated at 2600°C for 1.5sec.



[900J/gUO$_2$]

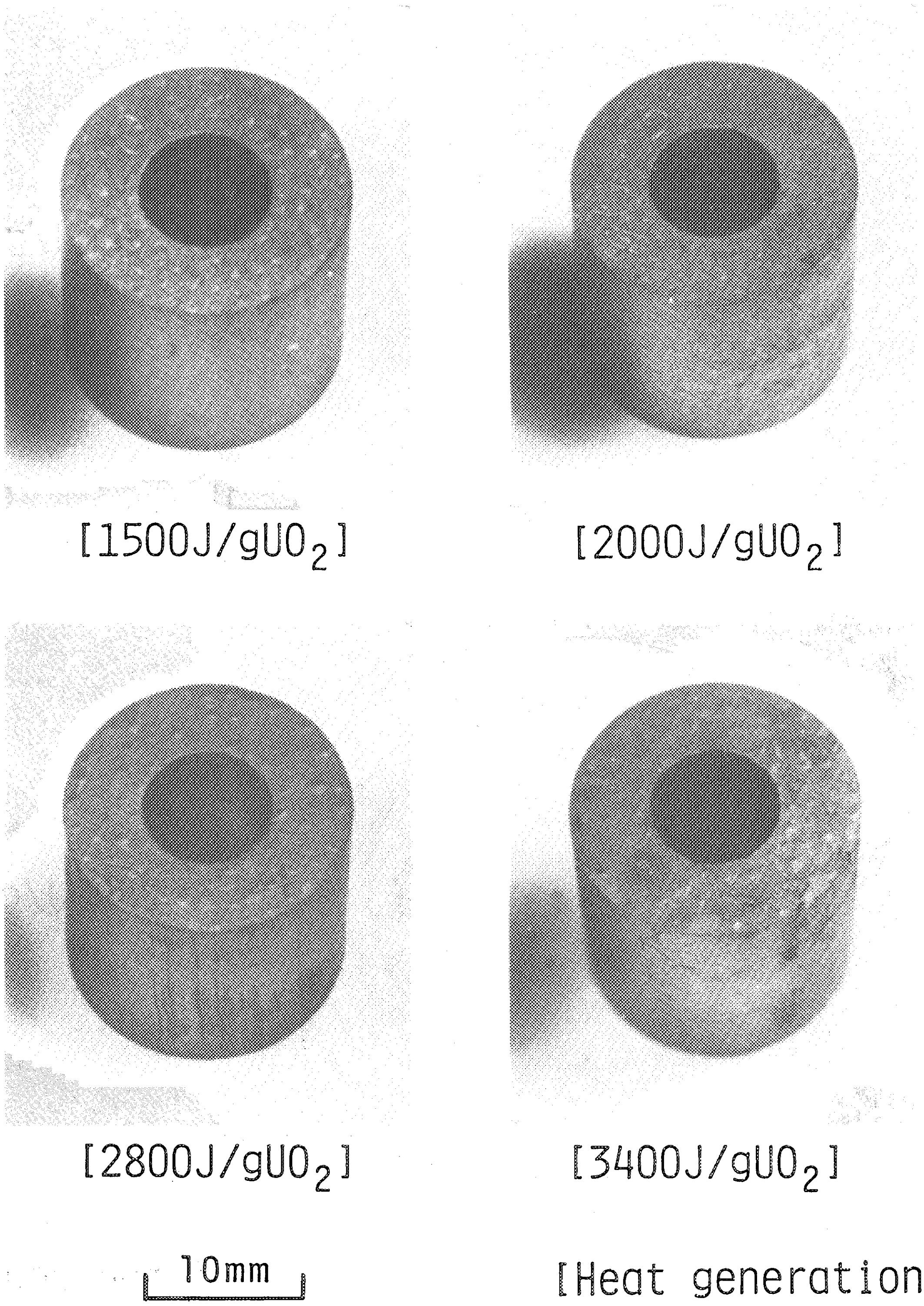
[1700J/gUO$_2$]

[2500J/gUO$_2$]

[4000J/gUO$_2$]

[Heat generation]

Fig. 7 Appearance of the loose coated particles irradiated in NSRR.



[$1500 J/gUO_2$]

[$2000 J/gUO_2$]

[$2800 J/gUO_2$]

[$3400 J/gUO_2$]

[Heat generation]

Fig. 8 Appearance of the fuel compacts irradiated in NSRR.

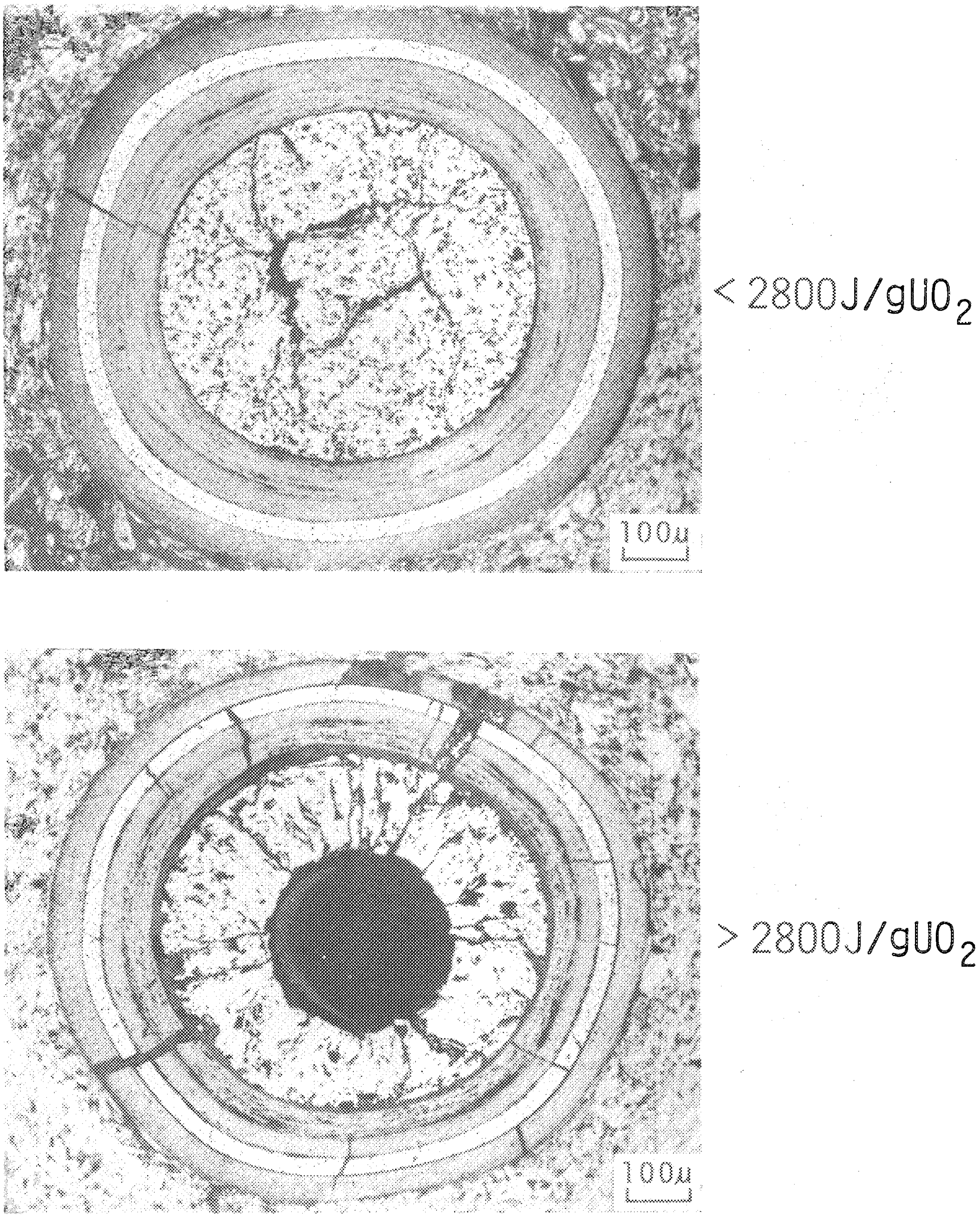
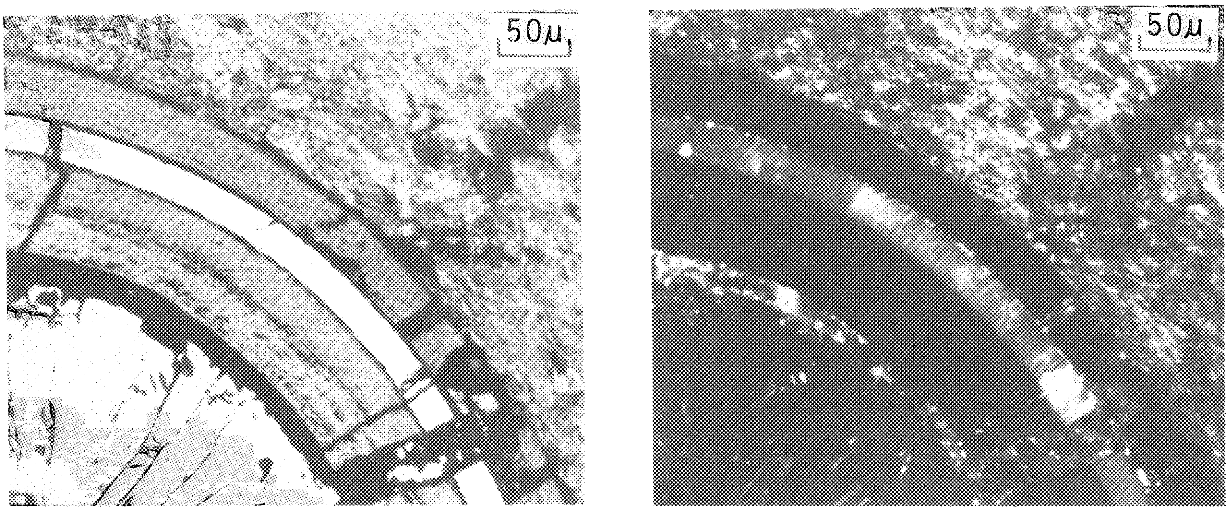
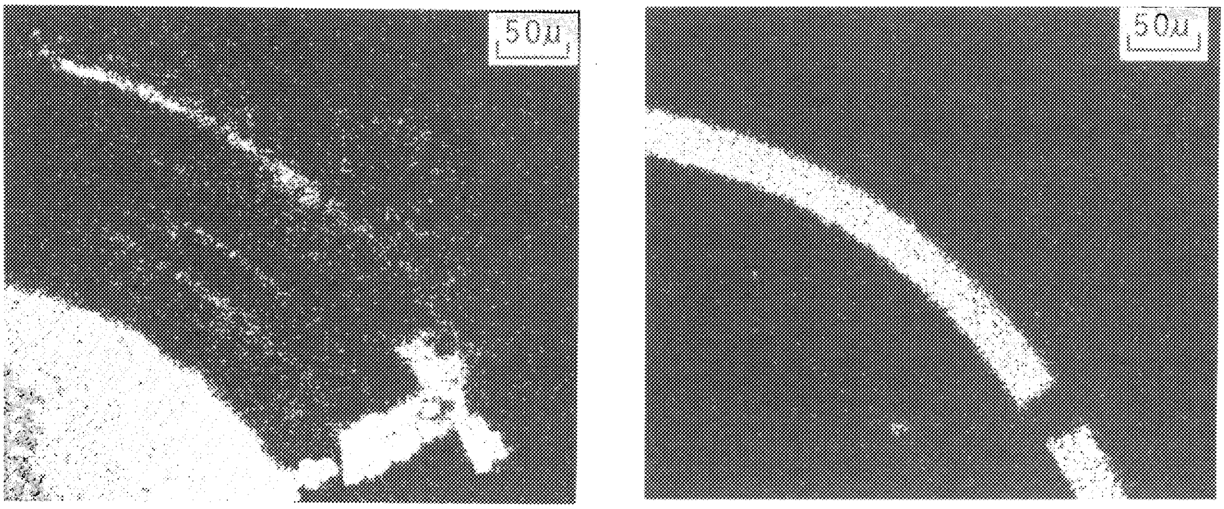


Fig. 9 Comparison of behavior of the coated particles irradiated at different heat generations.



Ceramography
(Bright light)

Ceramography
(Polarized light)



EPMA(U-Mα)

EPMA(Si-Kα)

Fig. 10 Ceramography and EPMA of the coated particles within compact irradiated in NSRR.