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SURVIVAL TEMPERATURE LIMIT OF UNIRRADIATED
COATED FUEL PARTICLES

June 1985

Satoru KASHIMURA and Katsuichi IKAWA

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Survival Temperature Limit of Unirradiated Coated Fuel Particles

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As a part of the study related to the hypothetical accidents of the experimental VHTR, a series of out-of-pile heating experiments has been carried out to determine the survival temperature limit of unirradiated TRISO-coated UO_2 particles. After heating, observations were made by X-ray radiograph, and optical microphotograph of polished cross-section. X-ray diffraction analysis were also used.

When heating time is short, survival temperature limit was found to be 2500°C for consolidated particles and 2400°C for loose particles. Plot of failure fraction as a function of heating time appears to be S-shaped. SiC degradation at relatively low temperatures (2200°C) started with thinning, whereas at high temperatures (2500°C) it started with micropore formation. At an extremely high temperature, total particle failure occurred physically before severe SiC degradation. At somewhat lower temperatures, total particle failure proceeded chemically after severe SiC degradation.

Keywords: Coated Fuel Particle, Silicon Carbide, Failure,
Very High Temperature, Temperature Dependence, VHTR,
Survival Temperature Limit,

未照射被覆燃料粒子の耐熱限界

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高温ガス実験炉 VHTR の仮想事故関連研究のひとつとして、未照射 TRISO- UO_2 粒子の耐熱限界を求めるための一連の炉外加熱実験を行った。加熱後、X線ラジオグラフィと研磨面の光学顕微鏡観察を行った。また、X線回折による分析も行った。

加熱時間が短い場合の耐熱限界は、成形粒子では 2500 °C、未成形粒子では 2400 °C であった。粒子破損率を加熱時間に対してプロットすると、S字型になった。SiC劣化は比較的低温 (2200 °C) では厚さ減少で始まったのに対し、比較的高温 (2500 °C) では気孔生成で始まった。極度の高温では、粒子の完全破損が激しい SiC劣化の前に物理的に起こったのに対し、若干低温では、粒子の完全破損は激しい SiC劣化の後に化学的に起こった。

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1. Introduction

Coated particle behavior and fission product release under accident conditions have been investigated in GAT⁽¹⁻³⁾, KFA⁽⁴⁻⁶⁾, JAERI⁽⁷⁻⁹⁾ and LA⁽¹⁰⁾. Data from US and FRG, however, are not directly applicable for safety evaluation of the JAERI's experimental VHTR, because kernel material and coating configuration or thickness are different from those of JAERI specification.

Survival temperature limit obtained by the previous JAERI experiment⁽⁷⁾ covers only 5-35min. SiC degradation was also investigated but only in a limited time range⁽⁸⁾. The purpose of the present experiment is to generate a set of data of time dependent survival temperature limit covering a much wider time range and to observe SiC degradation under a wide variety of temperature-time conditions.

2. Experimental

2.1 Specimen

Unirradiated TRISO-coated UO_2 particles were used either in loose or consolidated state. The dimensions and densities of the components of the particles are shown in Table 1. Well developed processes were used for particle fabrication, namely sol-gel process for kernel and fluidized CVD from acetylene, propylene and methyltrichlorosilane for low density carbon, high density carbon and SiC, respectively.

The process for consolidation of coated particles is shown in Fig.1. The particles are overcoated with graphite powder in a rotating flask. Five overcoated particles were pressed together

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to from a mini-size green compact, which is then heat treated at 900°C for 2h and at 1800°C for 1h. The consolidated specimen thus prepared is 2mm in diameter and 6mm in length. The external appearance and X-ray radiograph of the specimen were shown in Photo.1.

2.2 Procedure

Fig.2 shows the experimental apparatus. Specimens are heated by electric current through the specimen holder. The atmosphere is maintained inert by argon stream. The right half of Fig.2 shows a comparison of the two different arrangement of particles. Thirty particles in loose state are contained in the specimen holder, whereas in consolidated state two minicompacts containing five particles each are inserted in the holder. The holder is 4mm in outer diameter and 2mm in inner diameter and has a uniform temperature region of about 15mm in length.

For long time heating, the temperature was increased relatively slowly, but for short time heating, the temperature increasing rate was first slow then rapid, because the thermal effect in the particles in the course of temperature increase must not be too significant in comparison with that at the highest temperature. In the case of 2600°C heating, for example, temperature was increased relatively slowly up to about 1300°C which is the design fuel temperature in the experimental VHTR and then up to the highest temperature within 30-40sec. Temperature was measured using an optical pyrometer through a small hole drilled through the wall of the specimen holder (see Fig.2).

3. Results and discussion

The results are summarised in Fig.3. Particle denotes the loose state and compact denotes the consolidated state. When the outermost layer of a particle was found to be intact on the X-ray radiograph, the particle was defined to be intact, even if there were some reactions in SiC or on the kernel on the microphotograph of polished cross-section. The term "partially failed" was given to the case that at least one particle remained intact. The ratio of black area to white area in the symbols corresponds to the ratio of the number of failed particles to that of intact particles.

Fig.3 indicates that survival temperature limit line for consolidated state, the solid line, is about 100°C higher than that for loose state, the chain line. The survival temperature limit in the consolidated state was found to be 2500°C. At this temperature, however, particles failed within 1min. Time dependence can be made more clear by plotting failure fraction as a function of heating time as shown in Fig.4. It was found to be S-shaped. The fact that it takes a certain length of time to fail implies that the failure is, at least partly, chemical in nature.

In the case of consolidated particles, not only X-ray radiography to determine failure fraction but also optical microscope observation on polished cross-sections was made to obtain information on SiC degradation. X-ray diffraction analysis was also made to confirm chemical reactions.

Photo.2 shows the results of 100h heating at 2100°C. There is no change in either in X-ray radiograph or in microphotograph. X-ray radiograph after 51h heating at 2200°C also showed no change but microphotographs clearly indicate SiC degradation to a

significant extent as shown in the right photo of Photo.3. Burn leach method may not detect this extent of degradation but, without doubt, release of some fission products such as cesium and barium will be enhanced significantly. Photo.4 (2500°C, 10sec) is another but different example of all survived in X-ray and SiC degraded in microphoto. The difference is in the mode of SiC degradation. Photo.3 shows thinning, whereas Photo.4 shows micropore formation. SiC in Photo.4 seems to be more transparent to gasses than that in Photo.3, because the gap between the kernel and the buffer layer shown in the former Photo is wider than that in the latter Photo implying that CO generated by the reaction between the kernel and the buffer layer escaped more easily in the former case. It seems to be difficult to explain the wider gap by larger shrinkage of the kernel or of the buffer layer considering that the density of the kernel is already 96%TD before heating and that the ratio of the thickness of the buffer layer to that of the inner high density carbon layer appears to be similar to the ratio of those before heating. The explanation of the gap formation by the chemical reaction rather than by the physical shrinkage is supported by the presence of shining UC_2 deposit on the inner surface of the buffer layer (see Photo.4, right).

At 2600°C, the highest temperature in the present experiment, the length of time at the temperature was not accurate because electric current was cut immediately after the temperature was confirmed. It is surprising to note that SiC layer still existed to a considerable extent as shown in Photo.5. The micropore formation appears to be even less than in the case of 2500°C, 10sec, indicating that SiC degradation of this kind takes time.

Therefore failure at 2600°C is considered to be a physical phenomenon such as explosion due to internal CO pressure, although compact explosion as was observed in the previous experiment⁽⁷⁾ did not occur in the present experiment, probably because the compact was encased in the graphite holder. X-ray diffraction pattern (see Fig.5) indicates that UO_2 and UC still exist which means that the reaction between the kernel and the carbon layers was not complete. Fig.5 also shows the existence of SiC. This agrees with the above mentioned microphotograph observation in Photo.5.

At lower temperatures, chemical effect appears to prevail in the failure mechanism. Photo.6 shows a microphotograph of a particle after 2h heating at 2400°C. The particle failed completely and SiC disappeared completely. X-ray diffraction analysis indicates neither UO_2 nor SiC as shown in Fig.6. Only a trace amount of UC (an evidence of incomplete reaction) could be detected. Complete disappearance of SiC was already observed after 30min (see Photo.7). Even 5min was found to be too long for SiC at this temperature (see Photo.8).

Comparison between Photo.7 and Photo.8 indicates that drastic failure occurs between 5min and 30min at 2400°C. This comparison, however, is not enough to know whether total failure occurs immediately after SiC disappearance, because the time span between the two is too long. For 2500°C heating, two convenient microphotographs were taken. One was taken after 45sec, when SiC still existed to a large extent (see Photo.9) and the other was taken after 1min, when the particles failed completely (see Photo.10). Comparison between these two Photos indicates that

total failure occurs immediately after SiC degradation, although we must be careful to apply this observation directly to lower temperature cases when chemical reaction proceeds more slowly.

4. Conclusion

- (1) Survival temperature limit is 2500°C for consolidated particles and 2400°C for loose particles.
- (2) Failure at high temperatures occurs physically before severe SiC degradation, whereas at low temperatures it proceeds chemically after severe SiC degradation.

Acknowledgement

The authors are grateful to Dr.K.Iwamoto, the director of the authors department, for his support to the present experiment.

References

- (1) C.L.Smith, GA-A 12971 (1974)
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- (5) W.Schenk, JUL-1490 (1978)
- (6) W.Schenk, JUL-1883 (1983)
- (7) K.Ikawa, et al., J.Nucl.Sci.Technol.,15 (1978) 774
- (8) Y.Kurata, et al., J.Nucl.Mater., 92 (1980) 351
- (9) Y.Kurata, et al., J.Nucl.Mater., 89 (1981) 107
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Table 1 Dimensions and densities of the coated particle components

Component	Thickness (microns)	Density (g/cm ³)
UO ₂ kernel	576(diam)	10.5
Low density carbon	67	1.14
Inner high density carbon	26	1.85
Silicon carbide	22	3.21
Outer high density carbon	41	1.83

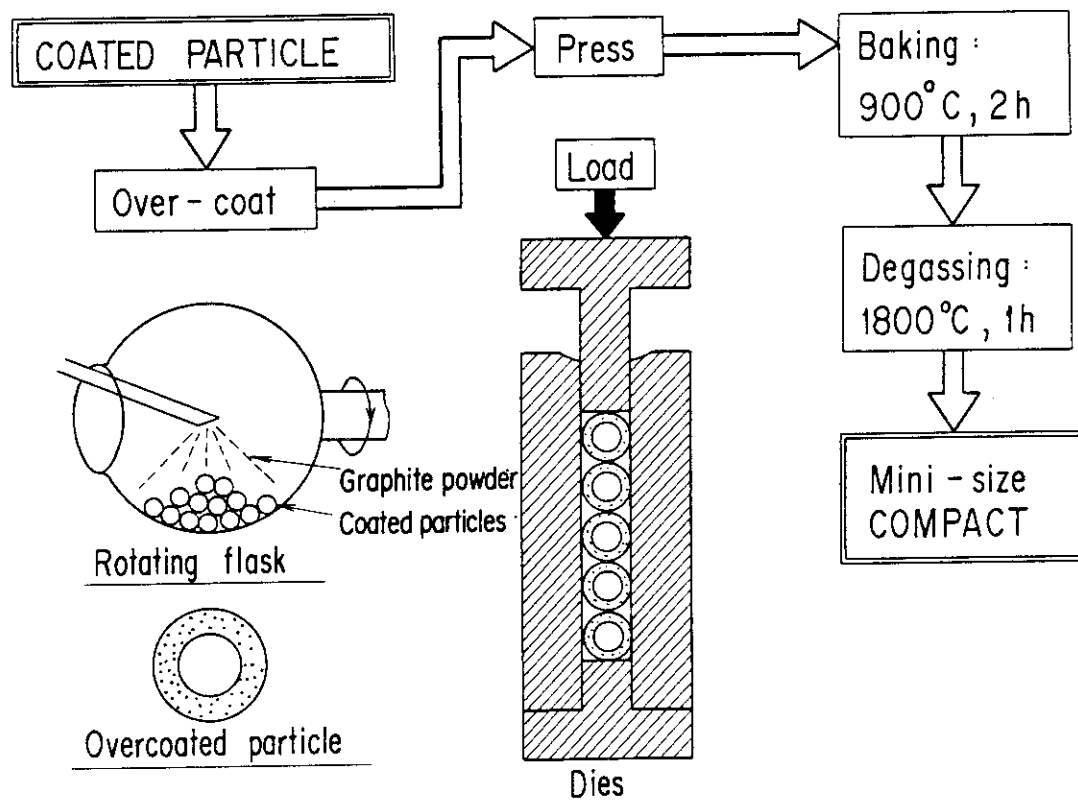


Fig.1 Consolidation of coated particles

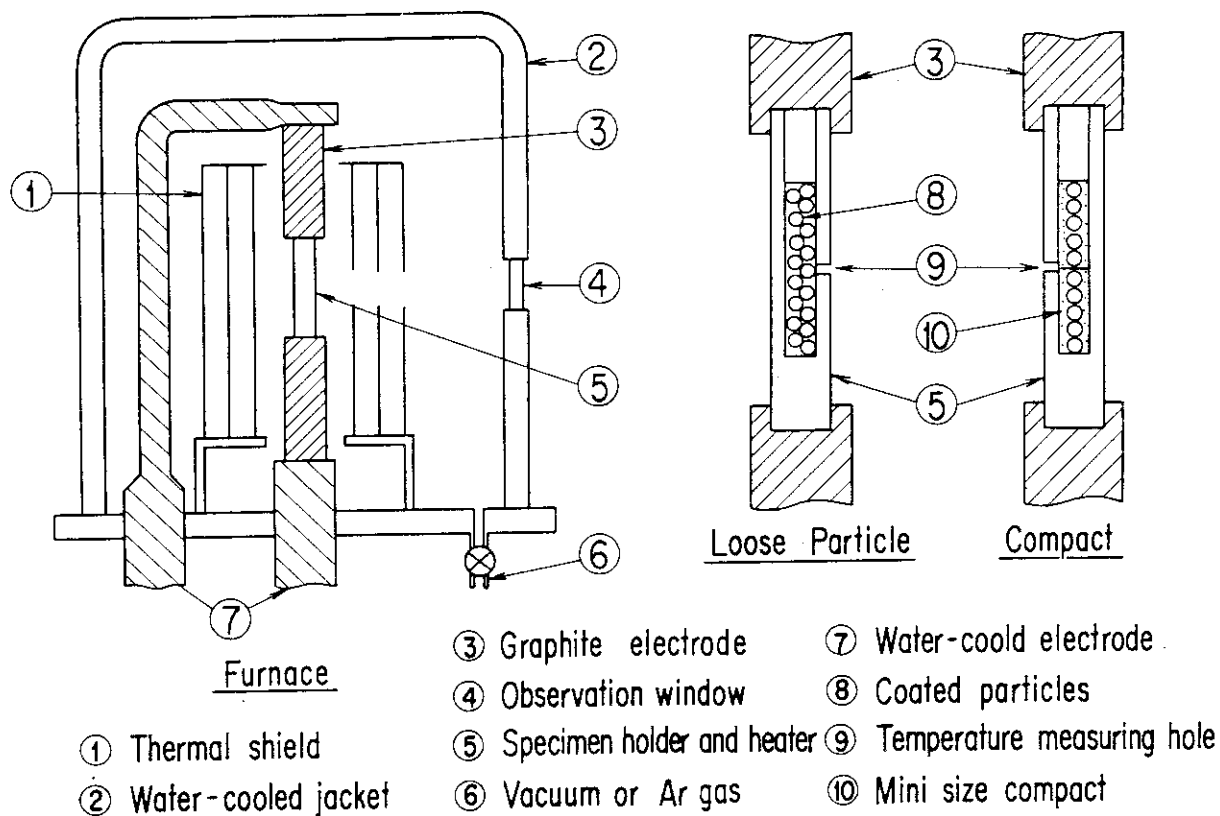


Fig.2 Experimental apparatus

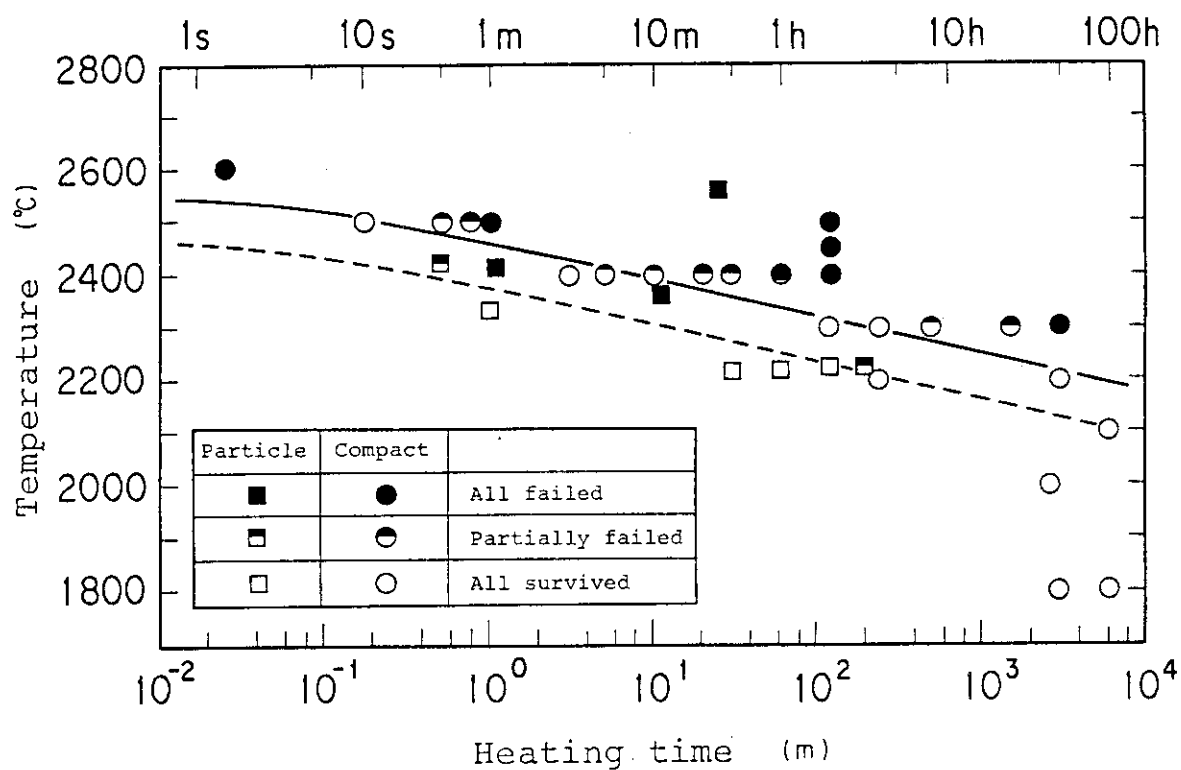


Fig.3 Time dependent survival temperature limit

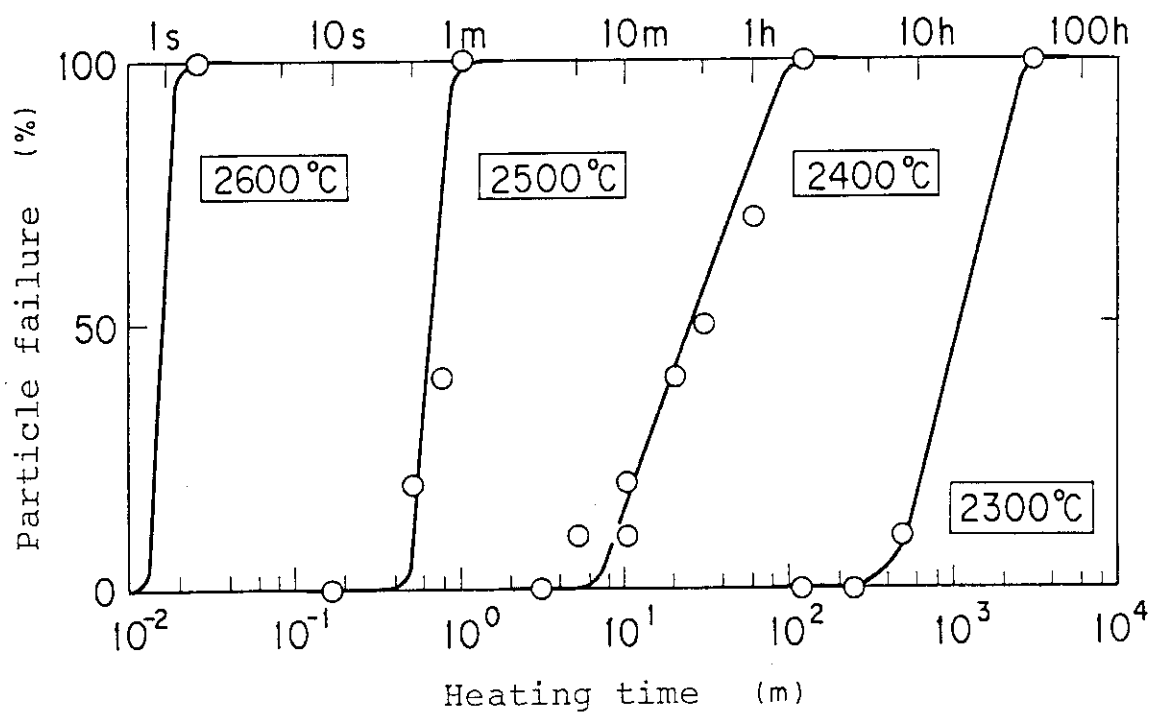


Fig.4 Heating time dependence of failure of particles in the consolidated state

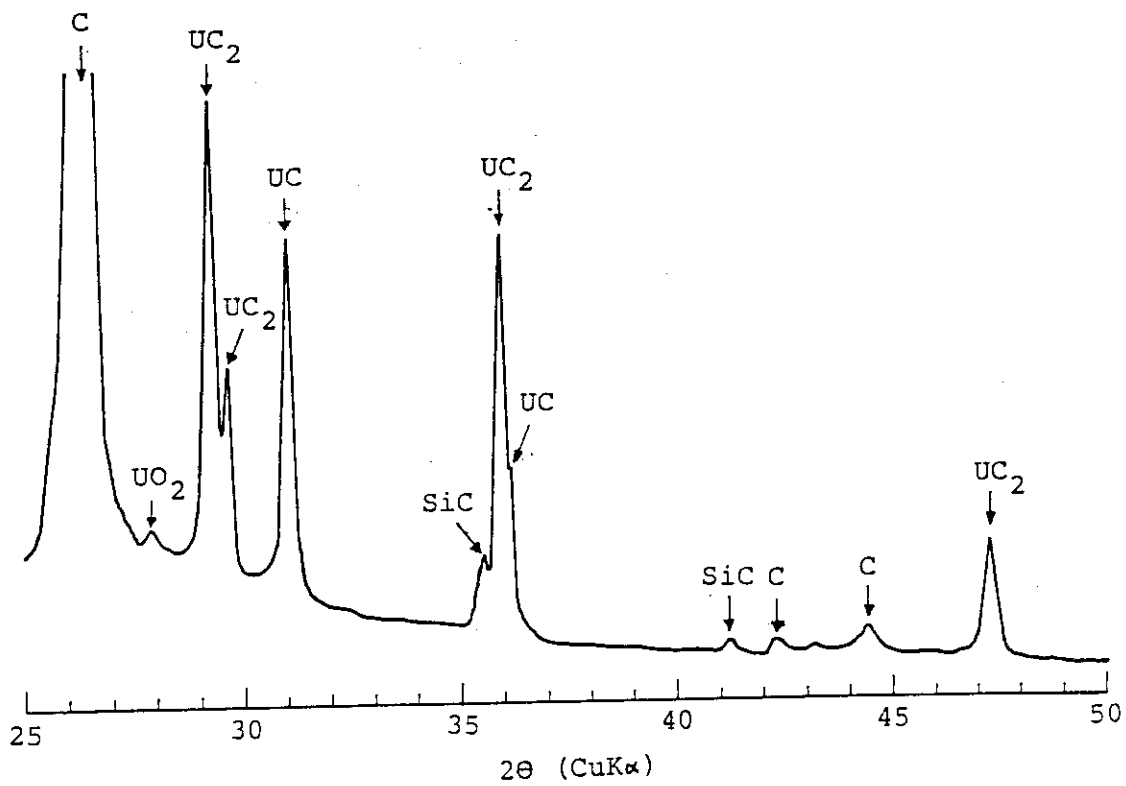


Fig.5 X-ray diffraction analysis of particles after 1.5sec at 2600°C

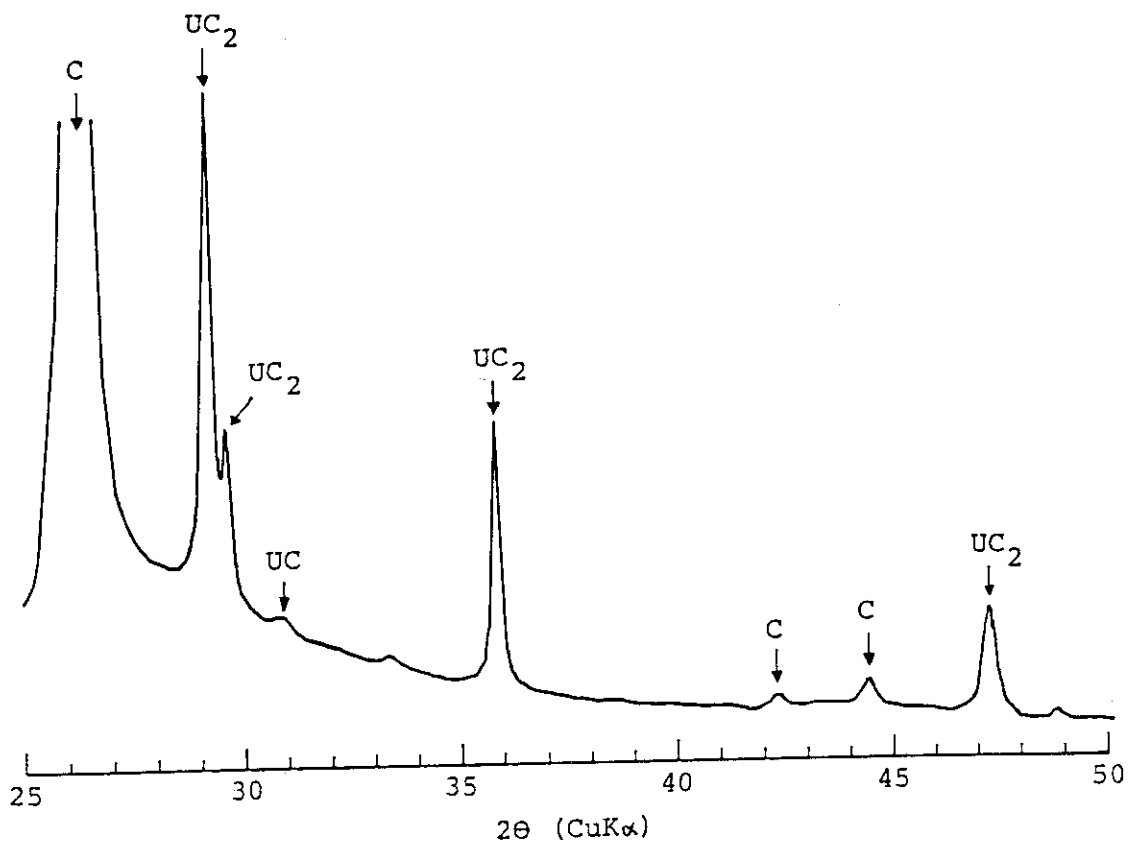
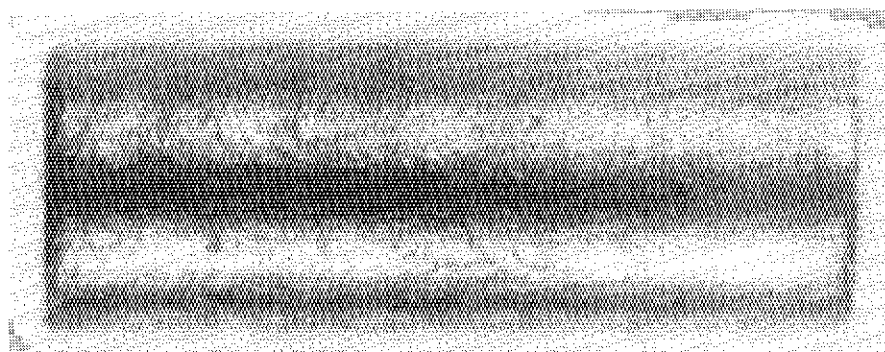
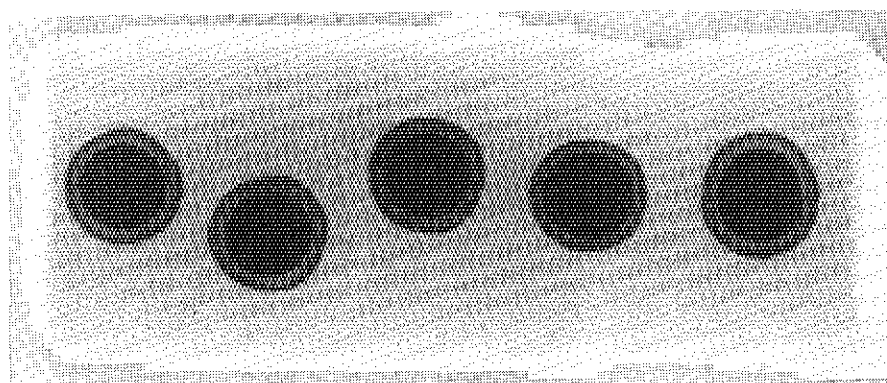


Fig.6 X-ray diffraction analysis of particles after 2h at 2400°C



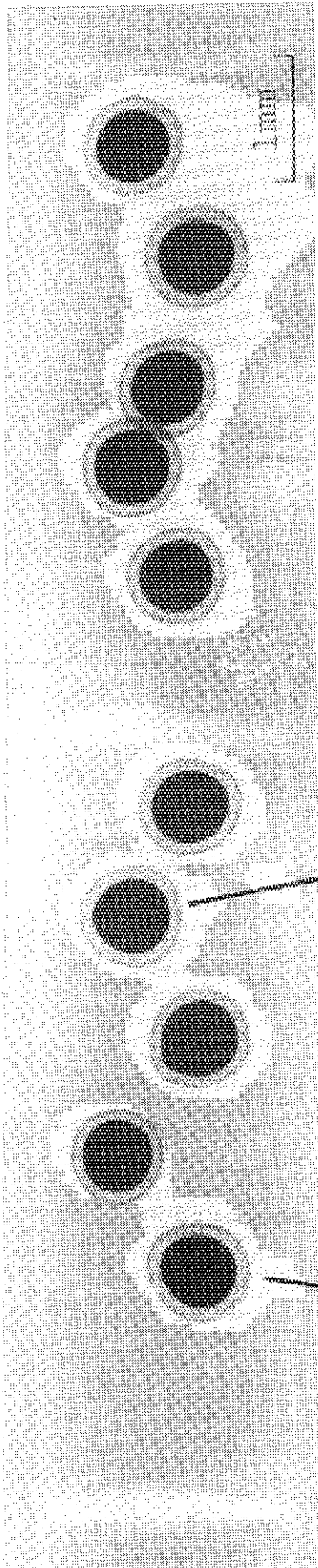
External appearance

1mm

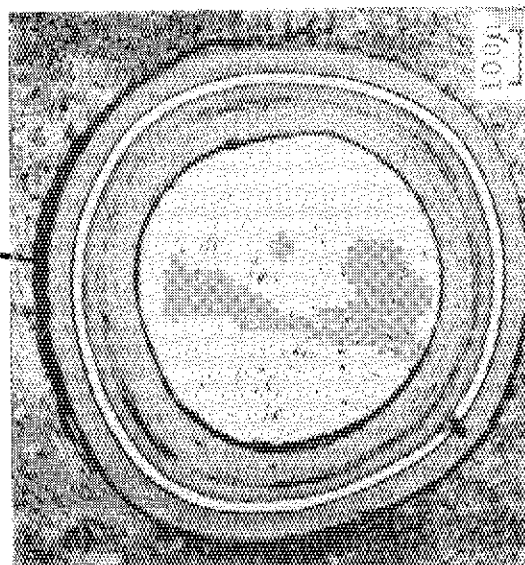
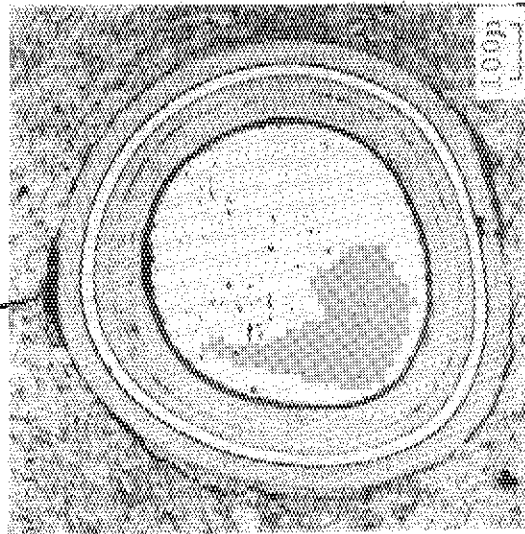
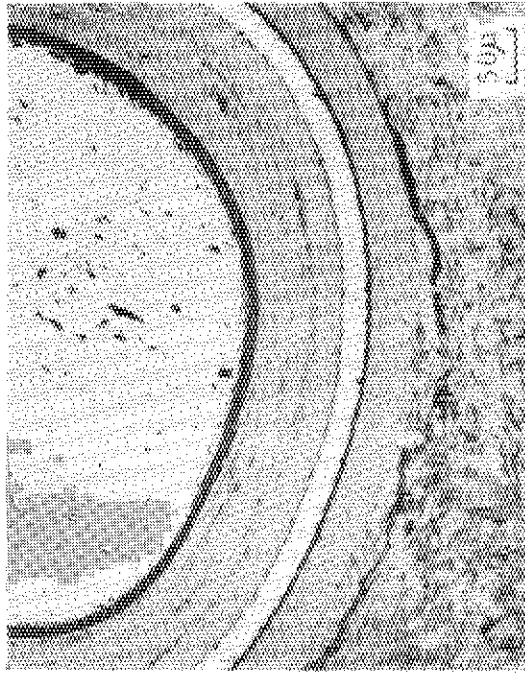


X-ray radiograph

Photo.1 External appearance and X-ray radiograph
of compact specimen

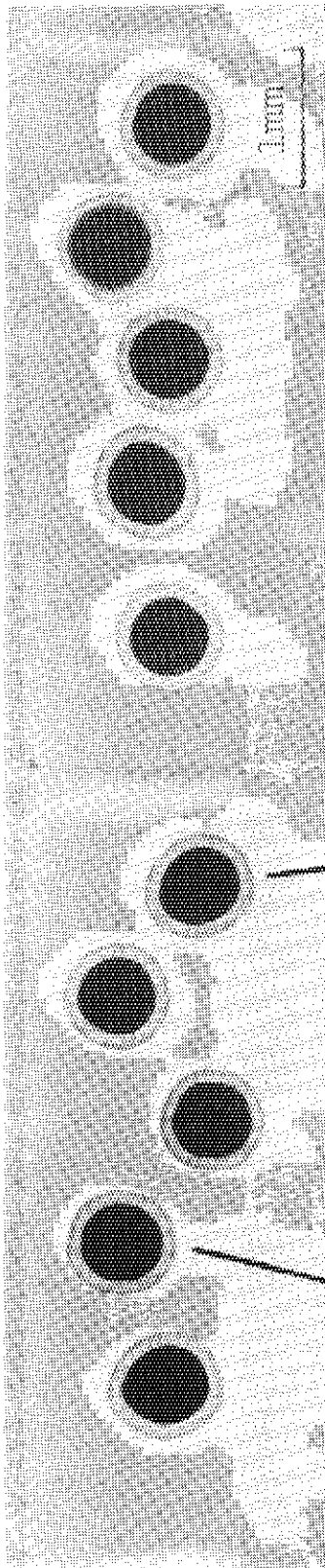


X-ray radiograph

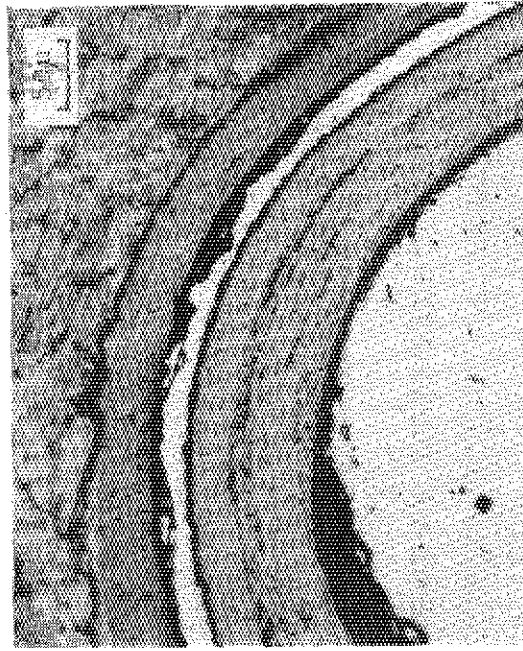


Microphotograph

Photo.2 X-ray radiograph and microphotograph of consolidated particles after 100h at 2100°C



X-ray radiograph



Microphotograph

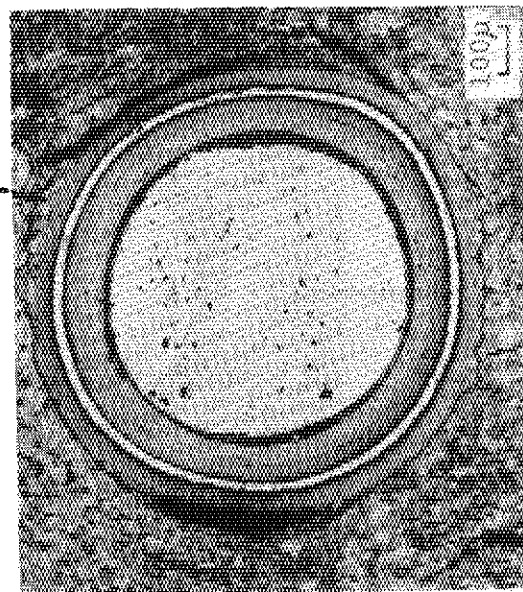
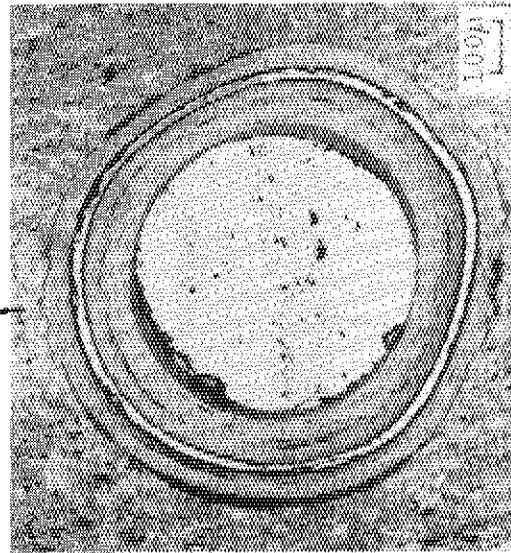
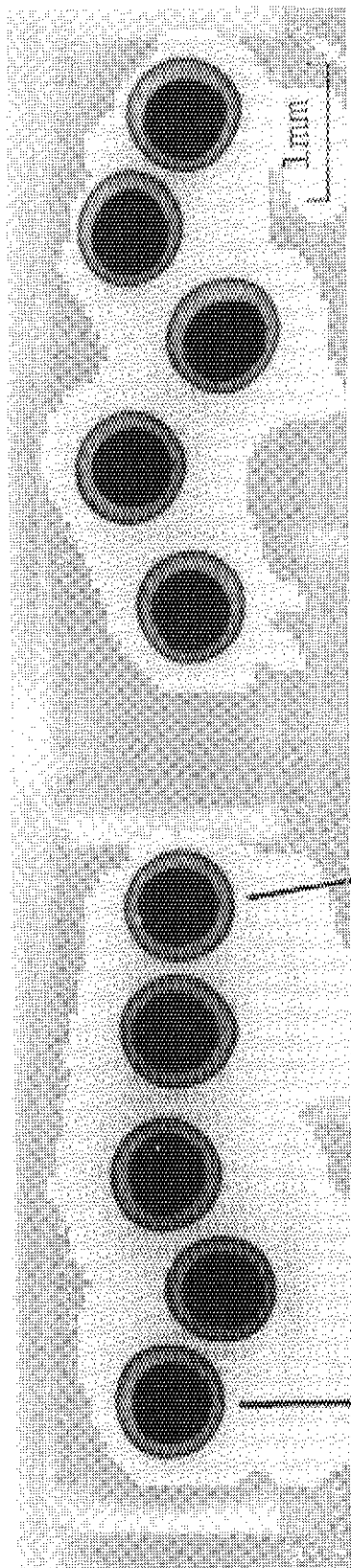
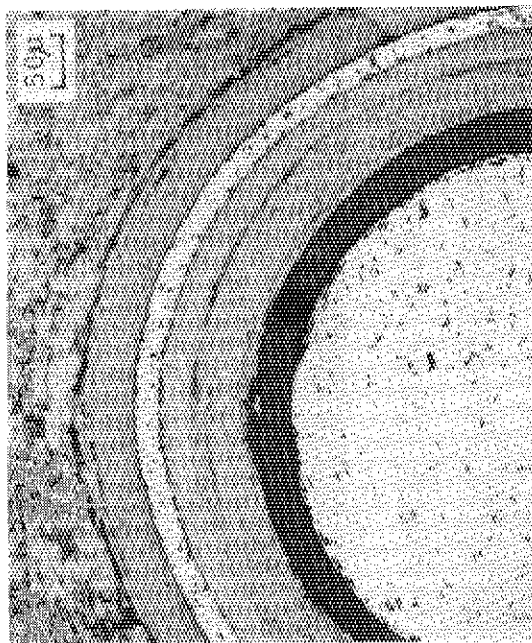


Photo. 3 X-ray radiograph and microphotograph of consolidated particles after 51h at 2200°C



X-ray radiograph



Microphotograph

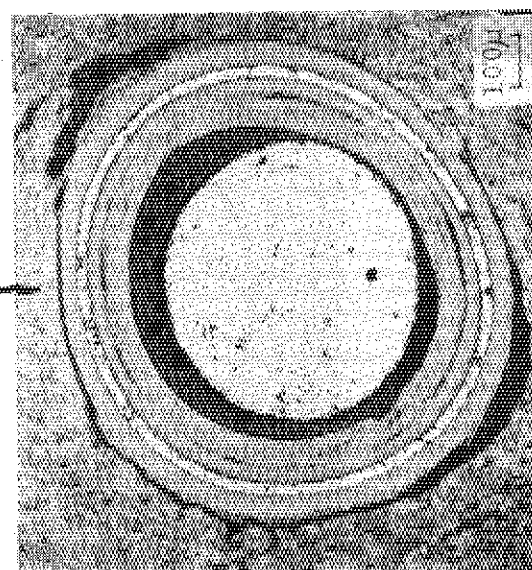
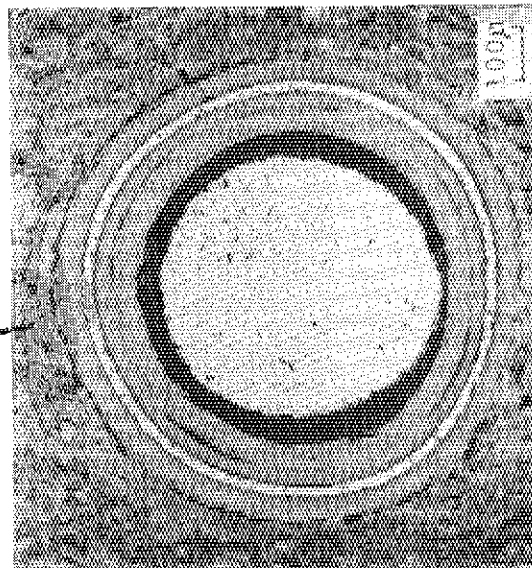
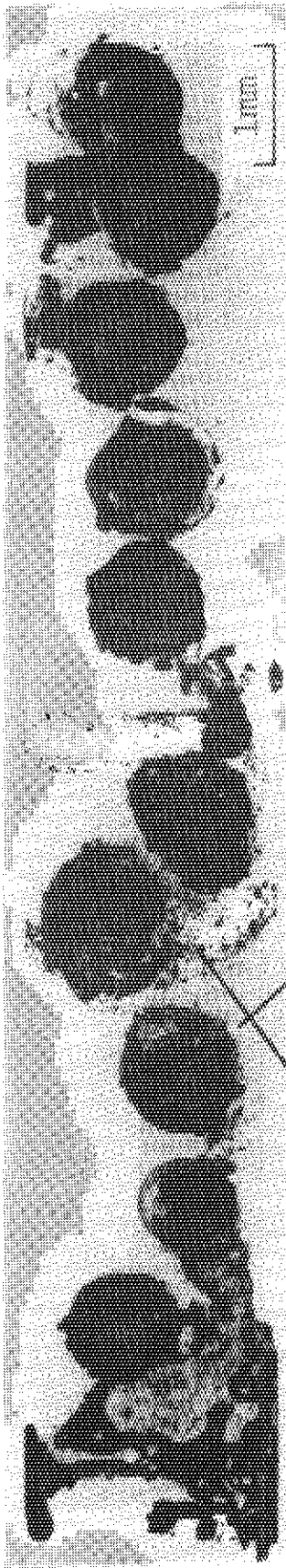


Photo.4 X-ray radiograph and microphotograph of consolidated particles after 10sec at 2500°C



X-ray radiograph



Microphotograph

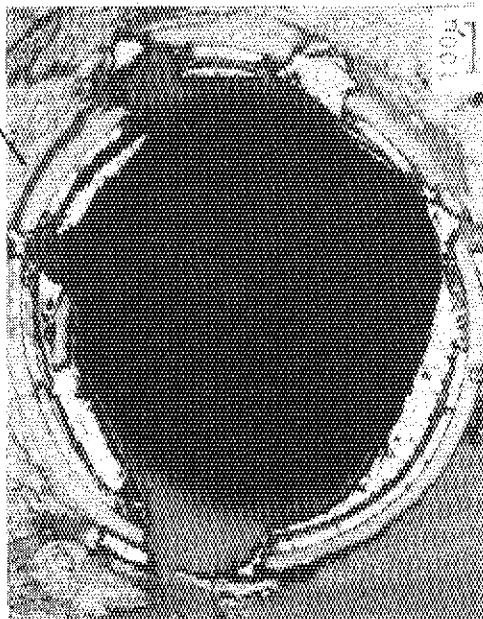
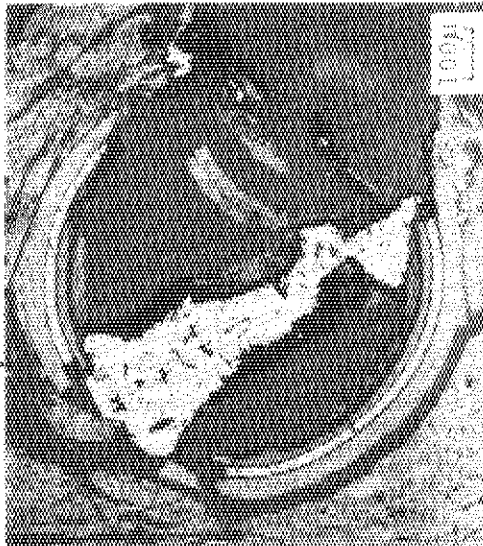


Photo.5 X-ray radiograph and microphotograph of consolidated particles after 1.5sec at 2600°C

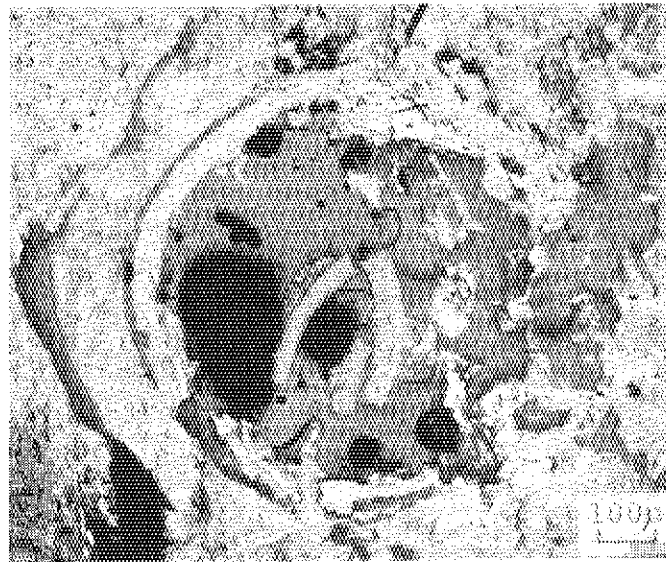


Photo.6 Microphotograph of a consolidated particle after 2h at 2400°C

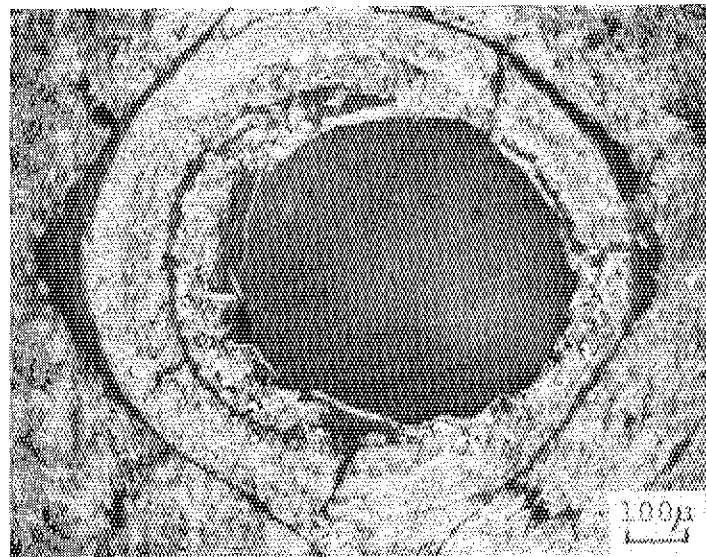


Photo.7 Microphotograph of a consolidated particle after 30min at 2400°C

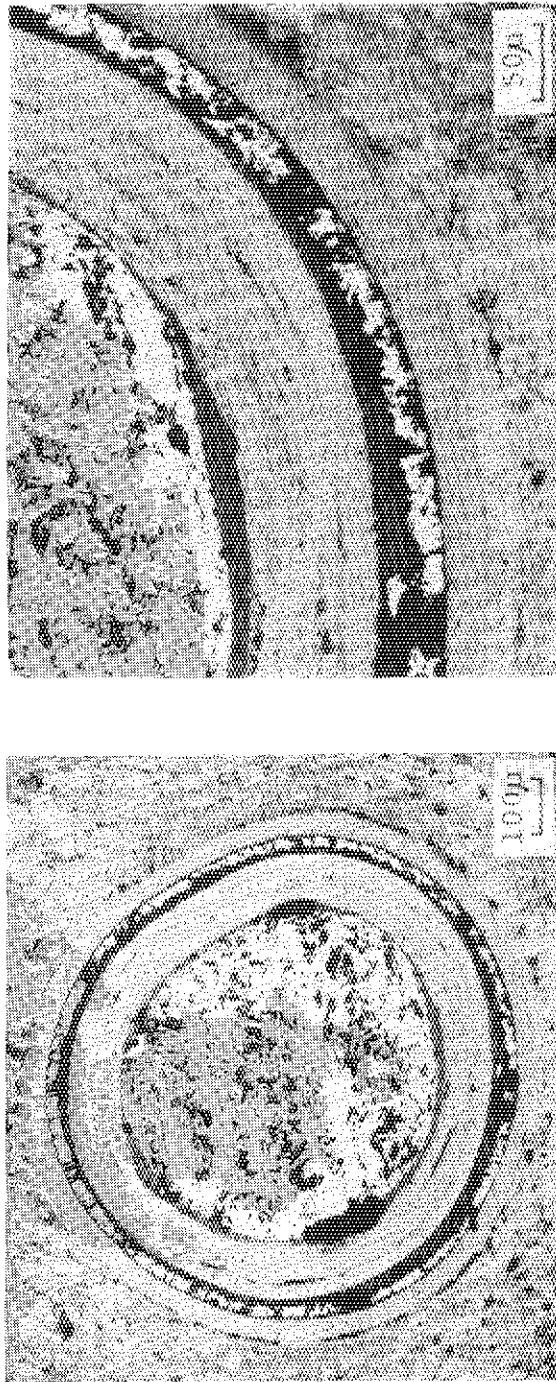


Photo.8 Microphotograph of consolidated particles after 5min at 2400°C

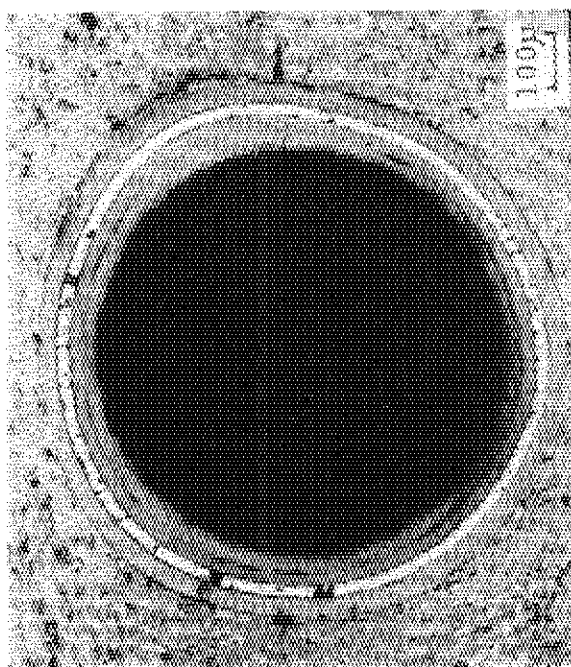
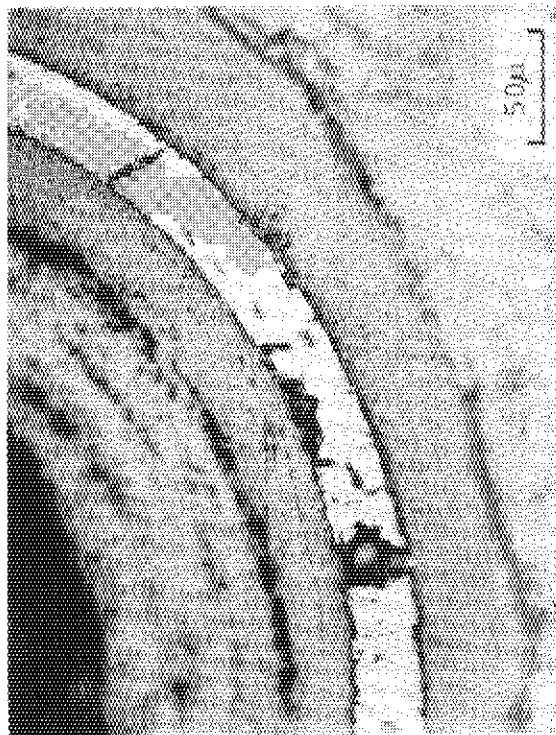


Photo.9 Microphotograph of consolidated particles after 45sec at 2500°C

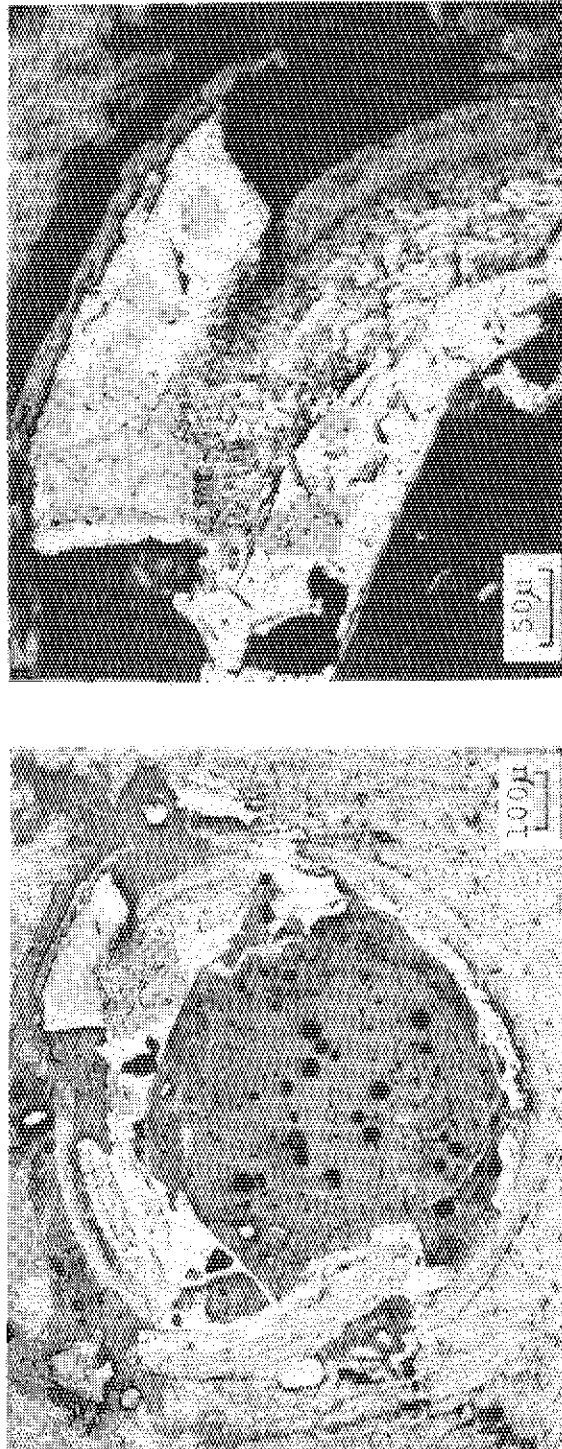


Photo.10 Microphotograph of consolidated particles after 1min at 2500°C