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EXPERIMENTAL OBSERVATIONS ON UO_2 KERNEL / PyC COATING INTERACTION

by

A. DRAGO

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Joint Nuclear Research Centre
Petten Establishent - Netherlands

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ABSTRACT

The heat treatment induced movement of UO_2 kernels through the pyrocarbon coatings of unirradiated coated particles was observed by a contact-microradiographic technique. A difference in the development of the phenomenon was observed in particles with visibly damaged coatings.

KEYWORDS

HEAT TREATMENTS
COATED FUEL PARTICLES
URANIUM DIOXIDE
PYROLYTIC CARBON
PELLETS
SILICON CARBIDES
MOTION
DEFECTS
DIFFUSION
RADIOGRAPHY
MICROSCOPY

Introduction.

The kernel/coating interaction was observed by means of a non-destructive technique on unirradiated particles having UO_2 kernels with the coating sequence : buffer-layer-high density pyrocarbon-silicon carbide - high density pyrocarbon. Both monolayer and massive annular compactswere tested, particles at both top and bottom ends of the latter being examined.In the case of the massive specimens, the radiographs show considerable damage to the particle coatings and a destructive examination of the sample confirmed this. Fig. 1 shows the image observed at the microscope of a coated particle after a very light grinding operation which only removed the matrix and outer pyrocarbon layer of the coating. In the picture it is possible to observe the seriously damaged silicon carbide layer.

The non-destructive test utilized to single out the presence of the kernel/coating interaction and to follow movements of the kernel was a contact-microradiographic technique utilizing an X-ray generator capable of working in the low kilovolt range.

Experimental observations.

During the examination of radiographic images of monolayer compacts of coated particles it was observed that in one of these, a few particles showed some kernel/coating interaction.

In figs. 2 and 3, two of these particles appear as radiographic images. These show the silicon carbide layer and everything enclosed by it. The outer pyrocarbon layer of the particle and the matrix of the monolayer compact are seen as a uniformly black region because the exposure time, and energy, were chosen to reveal the maximum detail in the region inside the silicon carbide shell.

In figures 2 and 3 it is possible to observe the gap existing on one side between the coating and the free surface of the kernels, whereas on the opposite side the region of attack is evident. Behind the free surface of the kernel is a track whose profile exactly reproduces that of the kernel. This track shows the approximate position occupied by the kernel when the interaction between the kernel and coating began. The track is considered to reveal the presence, in such a region, of material with an atomic number higher than silicon. There are reasons for thinking that the track is caused by the presence of uranium carbide, generated at the start of the process. The maximum distance between the track and the kernel profile is not considered to exactly represent the deplacement of the kernel but rather to combine kernel movement with a diffusion process tending to shift the track in the opposite direction.

To observe the dependence of the phenomenon on temperature, the monolayer compact was heat treated in a graphite box which was filled with graphite powder to avoid large temperature gradients. The temperature chosen for heat treatment was 1800°C, the temperature to which the compact had been exposed during manufacture.

The heat treatment was carried out in a number of increasing time intervals, the monolayer compact being radiographied to record the movement of the kernels through the coating at each step.

Figures 4 and 5 show the new position of the kernels for the same two particles after 7 hours at 1800°C and 1 hour at 1900°C.

The final heat treatment step consisted of two hours at 1900°C and then a rapid increase of temperature to 2100°C followed by immediate rapid cooling. One of the two particles (the one in which the phenomenon was the more pronounced) was completely destroyed by the reaction during the final treatment, uranium migration being observed in the matrix surrounding the particle. This can be seen on comparing figs. 6 and 7 which show at low magnification, before and after heat treatment,

the monolayer compact containing these two particles, (marked Aa) B).

Fig. 8 shows the other particle (see figs. 3 and 5), which was located in the monolayer compact close to the destroyed particle. The large and shapeless white region is due to uranium migration from the destroyed particle.

The movement of the kernel was also observed in all the other particles present in the monolayer for which the phenomenon had been evident before the start of the several heat treatments. All other particles, which were intact at the beginning, remained unchanged at the end of all the heat treatments.

Fig.. 9 shows one of the particles present in the full-size compact. In this picture it is possible to observe both the kernel attack and two large cracks in the coating. It will be noticed that the region between the kernel profile and its track looks like a "fog". Such "fogging" is always present in all the particles showing kernel attack with heavy damage to the coating. Fig. 10 shows the reaction between kernel and coating at an advanced stage.

Considerations.

On surveying the radiographs of the monolayer compact it can be observed that the kernel attack appears to have no preferential orientation but is randomly orientated among the particles. From this it is concluded that the phenomenon was not generated by the presence of a general temperature gradient through the monolayer compact, but perhaps by local temperature gradients. From all the photographic evidence so far accumulated, it appears that whatever the reason for starting, once begun the reaction, continues to proceed preferentially in the same region thus causing the kernel to appear to move in the same direction. In cases where the coating is seriously damaged, a "fog" effect is always present in the region opposite to that through which the kernel is moving. Such a "fogging" in the picture indicates the presence of heavy metal on the internal wall of the coating ; for instance, uranium carbide produced as a consequence of CO release through the damaged coating.

Conclusions.

The present work does not pretend to give an interpretation of the kernel/coating interaction, but aims to show the validity of contact-microradiography as a non-destructive test for studying and following the evolution of the phenomenon.

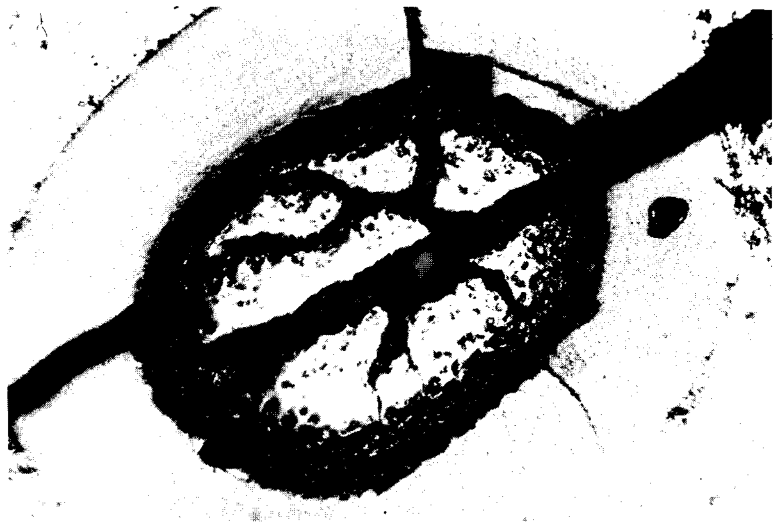

Fig. 1 Particle after very light grinding showing damage caused to SiC layer during consolidation of the compact.

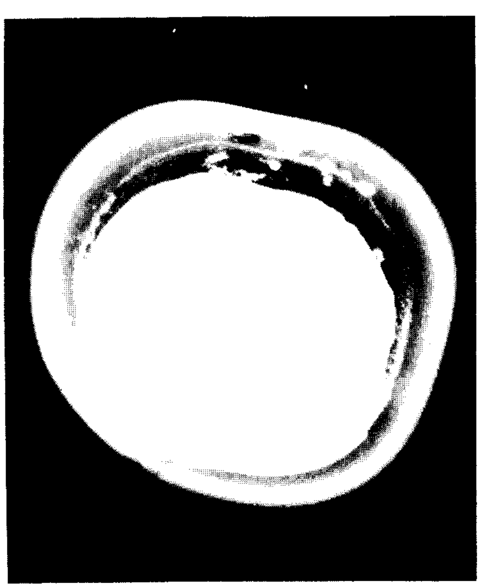

Fig. 2 Particle A before heat treatment

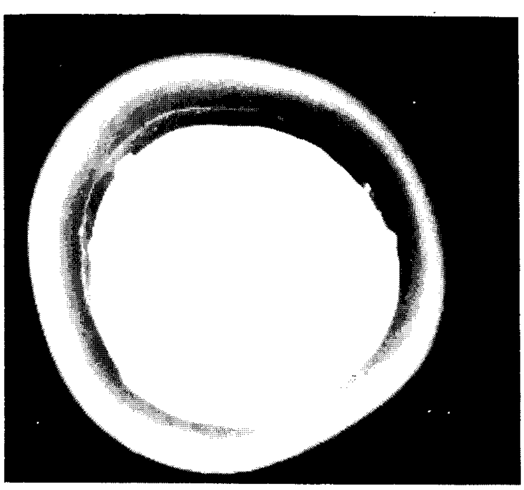

Fig. 3 Particle B before heat treatment

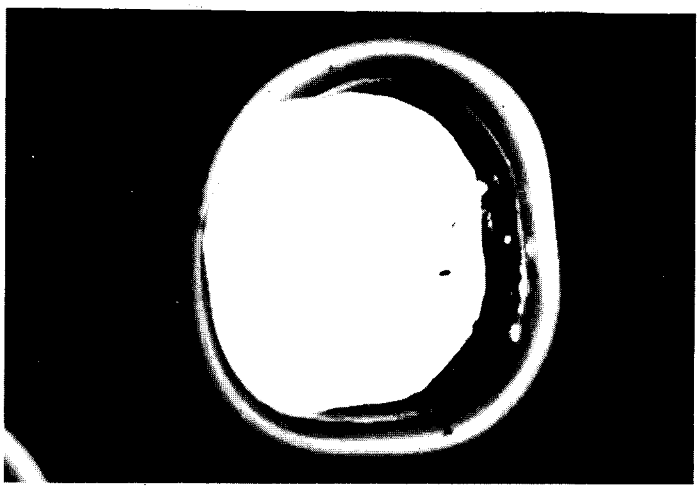

Fig. 4 Particle A after 7 hours at 1800°C and 1 hour at 1900°C

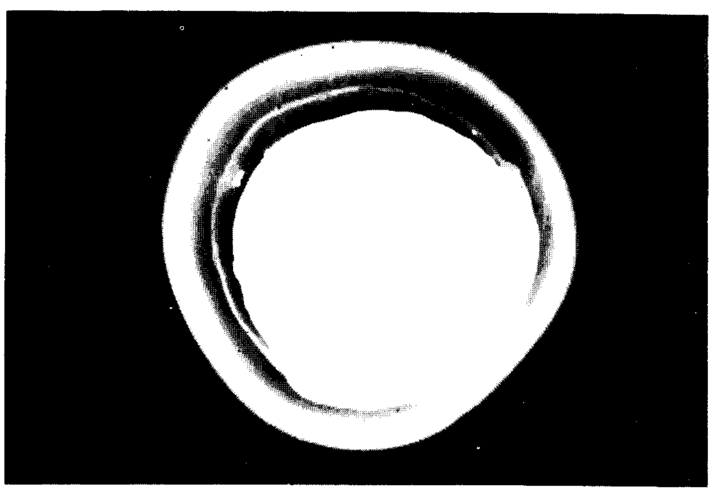

Fig. 5 Particle B after 7 hours at 1800°C and 1 hour at 1900°C

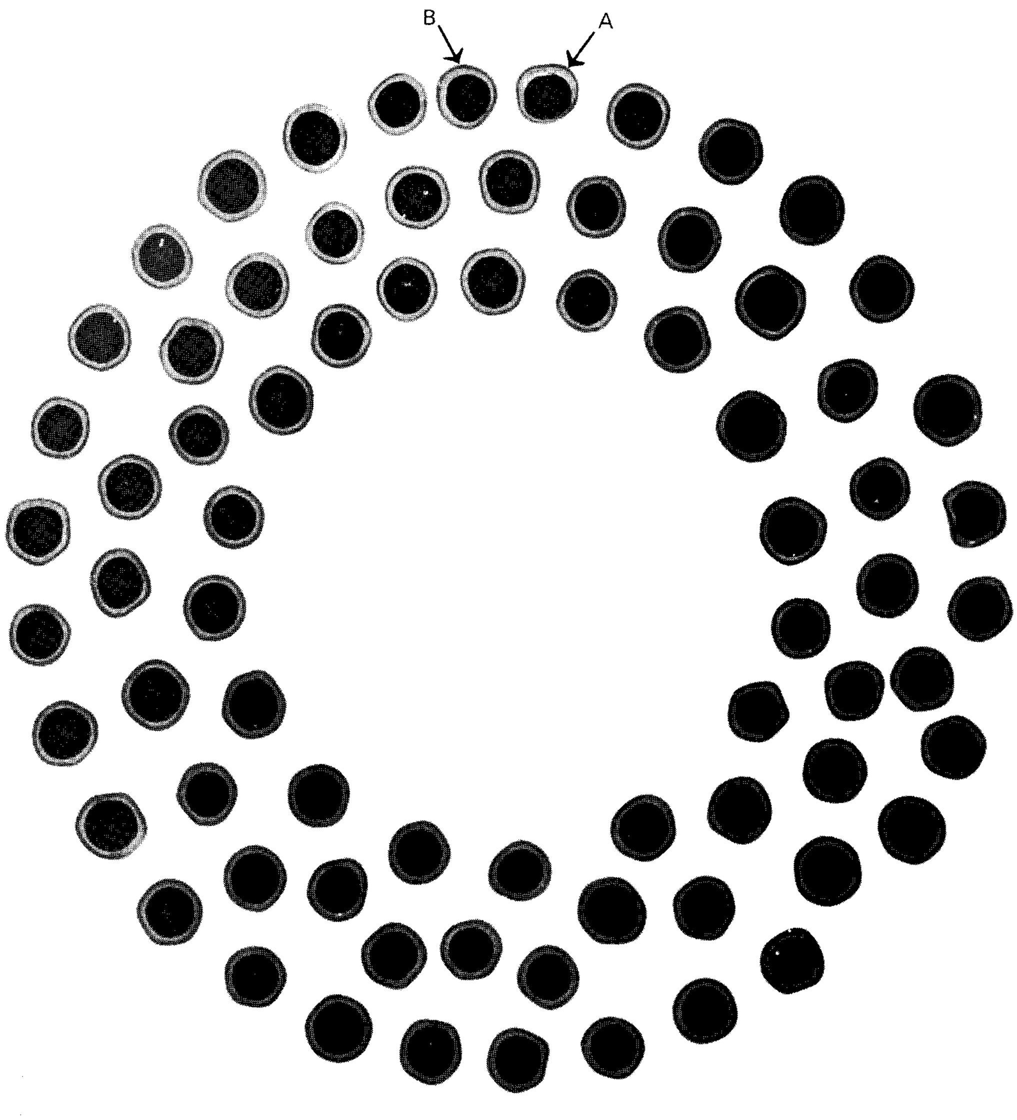

Fig. 6 Monolayer compact containing particles A and B before the heat treatment.

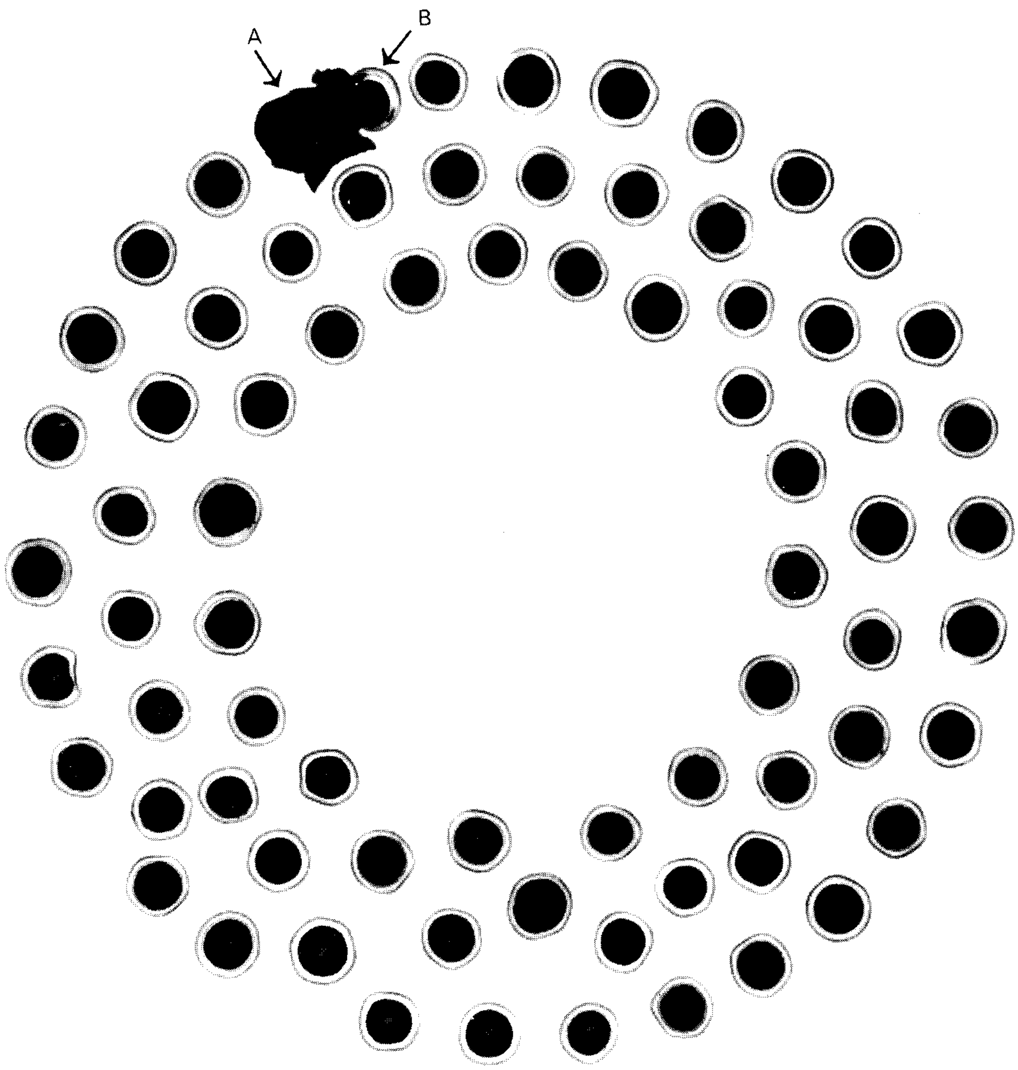

Fig. 7 Monolayer compact containing particles A and B after final heat treatment.

Fig. 8 Particle B after the final heat treatment [partly obscured by heavy metal diffused from particle A]

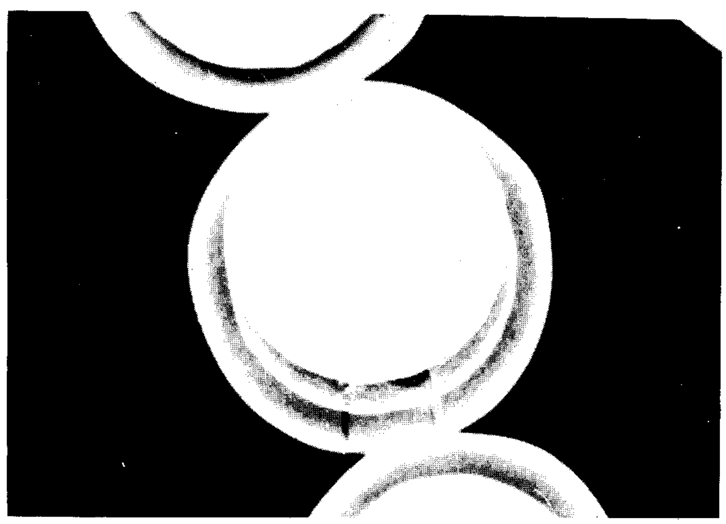

Fig. 9 Particle from full-size compact showing kemel attack and cracked coating.

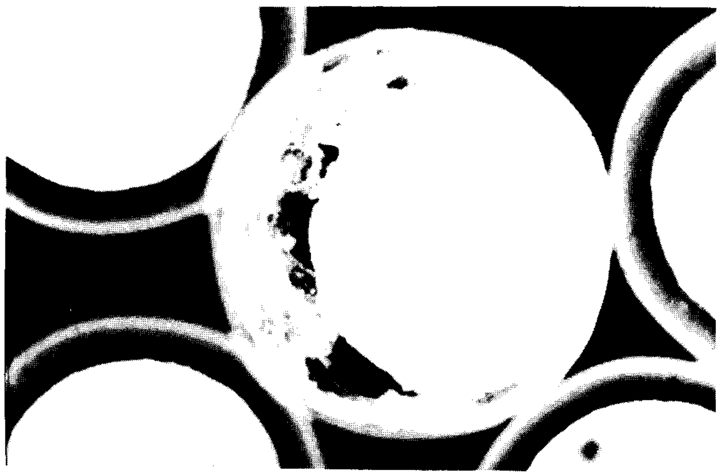

Fig. 10 Kemel/coating reaction at advanced stage.

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Alfred Nobel

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