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CRUSHING BEHAVIOUR OF PYROCARBON COATED FUEL PARTICLES

by

A. DRAGO and W. HUBER

1970

Joint Nuclear Research Center
Petten Establishment — Netherlands

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ABSTRACT

In this report the importance of the optical analysis of the mode of crushing of stressed coated particles, for the characterization of mechanical behaviour of the coating, is exposed. Experimental results are presented and discussed.

KEYWORDS

OPTICAL PROPERTIES
ANALYSIS
CRUSHING
STRESSES
FUELS
PARTICLES
COATING
MECHANICAL PROPERTIES

CRUSHING BEHAVIOUR OF PYROCARBON COATED FUEL PARTICLES *)

INTRODUCTION

There are several possible mechanisms by which fuel materials and fission products can diffuse through a graphite or carbon matrix at high temperature. The only acceptable situation for H.T.G.R. is one in which the migration can be contained within the coated particles and is controlled by a very slow process.

At the moment a fuel element for H.T.G.R. is generally composed of an external graphite shell lapped by the cooling gas. Inside there is a fuel compact which consists of coated fuel particles dispersed in a carbon matrix.

In such a fuel element the coating has an important role as a pressure vessel for gaseous fission products, as a diffusion barrier for fuel materials and for fission products. In such an optic the failure of the coatings have catastrophic consequences on the fractional release level.

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*) Manuscript received on 15 April 1970

During the whole life of a fuel element different factors can threaten the mechanical integrity of the coating, as for instance:

1) mechanical shocks during coated particle manipulations,
2) mechanical and thermal shocks during consolidation of carbon matrix,
3) thermal stress during irradiation,
4) radiation damage effects,
5) swelling of kernels and shrinkage of carbon matrix during irradiation,

However, the relevance of the mechanical behaviour of the coating may be seen in a different optic when its behaviour, when dispersed in a graphite matrix, is considered; in this respect this work should be considered as a first approach to the study of the irradiation behaviour of fuelled compacts. New diffusion barriers which have to be combined with pyrocarbon are under consideration in several laboratories for the purpose of increasing the retention of fission products. According to the current design of coated particles for thermal reactors, this implies a multi-layer structure; in some cases (i.e. PyC - SiC mixed structures) the different irradiation behaviour and elastic properties of the materials involved, is giving rise to peculiar levels and states of stress. The final aim being to minimize the overall fission product release, a balance should be made, taking care of both diffusion properties and mechanical behaviour.

From the previous considerations we feel that attention should be directed to the mechanical behaviour of coatings before and after irradiation. Particularly it is our intention to correlate the maximum crushing force and the crushing modality with some manufacturing parameters and to find parameters and criteria to predict by physical analysis before and after irradiation, the behaviour under irradiation.

1. EXPERIMENTAL APPROACH

To study the mechanical behaviour of the coating we have realized a crushing strength device, presented in fig. 1, that permits optical and photographic examination of coated particles before, during and after crushing. The device allows the evaluation of the maximum crushing force in coated particles and to record simultaneously the elastic deformation under stress; the particle diameter can also be recorded. We consider the optical and photographic analysis very important in the characterization of pyrocarbon coating macro structures.

In our preliminary work several coated particles of three different batches have been examined, in order to check the validity of our supposition on the importance of the optical examination of the crushing modality. In table I we summarize some manufacturing parameters for the coated particles considered.

COATED PARTICLE TYPE	KERNEL	COATING	THICKNESS OF HIGH DENSITY COATING
A	UC-10	Pyrocarbon + Buffer layer	104 μ mm
B	UO_2	Pyrocarbon + Buffer layer	97 μ m
C	Glassy-Carbon	Pyrocarbon	100 μ m

The coated particles type A and C are predominantly spherical whereas the B type are generally polyhedrics.

For the determination of the crushing stress we have utilized the approximate expression:

$$\sigma = \frac{Fe}{\pi (b^2 - a^2)} \qquad (1)$$

proposed by W. Delle et al [1]. The expression (1) is derived with the same considerations from the more general relation:

$$\sigma - \sigma_P = - \frac{Fe - Fi}{\pi (b^2 - a^2)}$$

where

σ = crushing stress
σ_p = internal stress resulting from manufacture
Fe = crushing force measured
Fi = reaction force between kernel and coating
a = internal radius of coating excluding the sacrificial layer
b = external radius of coating

The authors arrive at the expression (1) from the following consideration:

a. Between the kernel and the exterior there is a porous buffer layer. The strength of this buffer layer is not very high, consequently it is possible to compress the buffer heavily without much force. There will be no important support of the kernel if the print depth is significantly smaller than the depth of the buffer layer. In this case it can be stated approximately that:

$$Fi = 0$$

b. A support of the kernel cannot be effected if the Young's modulus of the exterior coating is considerably smaller in a radial direction than in a tangential direction. This phenomena can be described by the ''arc effect'' which is much more efficient than the support of the kernel. In this case too

$$Fi = 0$$

c. The only remaining stress in the coating before the crushing test derives from the fabrication process. Hence the only stresses before the crushing test will be those coming from the anisotropic thermal expansion during cooling from coating temperature to room temperature. Estimations show that the highest amounts of stress occur at the inner and exterior coating surface and that their amounts are between 1 and 1.5 kg/mm^2 at usual coating temperatures. The mean value across the coating thickness however is considerably smaller than the values mentioned above since the stresses change their sign at the middle of the coating. In the frame of the applied theory of shells it can be put $\sigma_i = 0$.

In table II we give the mean crushing stresses determined on 24 coated particles for each considered type.

TABLE II

COATED PARTICLE TYPE	A	B	C
$\bar{\sigma}$ (Kg/mm^2)	8,79	11,66	21,60
$s = \sqrt{\frac{(\sigma_i - \bar{\sigma})^2}{n(n-1)}}$	± 0,26	± 0,18	± 1,31

2. CRUSHING BEHAVIOUR

2.1 Coated particles type A

A typical elastic deformation under stress of a coated particle type A is shown in fig. 2. In all our measurements the particles were stressed until evident breakage occurred. Examining the curve shown in fig. 2 a certain number of fluctuations or peaks can be seen before the particle breaks down. At the same time the region with the peaks, shows a variation of the slope. The last observation could indicate that plastic deformations have taken place. In order to explain these phenomena more accurate analyses were necessary.

Optical control during stress revealed the absence of external cracks when the peaks on the recorder chart appear. On the other side when loading the same particle two or three times just before final crushing occurs (fig. 3), the peaks present in the first curve disappear in the second and third curve. Furthermore the variation of the slope noted in first curve has disappeared together with the peaks, and a more elastic behaviour of the particle can be deduced from the smaller slope of the second curve. However the second and third curve are identical. Twenty-four coated particles which were stressed until peaks were observed, microscopic analyses of the external surface were performed. Once more no cracks could be found. Metallographic examination of the 24 particle sections made it obvious that delamination processes have taken place, see fig. 4 and 5.

The conclusion of the previous observations is that the peaks are associated with ungluing phenomena taking place in the coating.

On the other hand it could be observed that the broken particles show evident delamination processes, see fig, 6. More strikingly this effect can be demonstrated in fig. 7a and 7b, where the same particle is photographed stressed and unstressed.

Finally an agreement between the number of peaks and the number of delaminations were found. The rupture process of the particle type A happens by ''collapse'' (*) as shown in fig. 8. As far as particle type A is concerned, the kernel is always fractured together with the coating, fig. 9 and 10.

(*) The term ''collapse'' indicates a process characterized by high deformation of the particle before rupture and complete fall-in of broken fragments.

2.2 Coated particles type B

A typical elastic deformation under stress of a particle type B is shown in fig. 11. With this type of particle peaks are also present before the crushing force is attained. However no change in the elasticity can be observed. The peaks in this type of particle are not ascribable to ungluing phenomena but they are related to the polyhedrial geometry. In fact when the peaks of the particle under compression are recorded chips of the coating are leaving the particle near the contact point. The chips jumping away are a consequence of a superficial explosion.

In fig. 12 it is possible to observe the chip effect. The breakage of this type of particles happens by ''explosion''(*) The optical analysis of particles after crushing show that the fracture surfaces present a ''flight of stairs'' configuration, see fig. 13.

This ''flight of stairs'' points out the shell structure of the coating, but no ungluing phenomena are evident. For this type of particle entire kernels are generally present, see fig. 14

2.3 Coated particles type C

A typical deformation under stress of a particle type C is shown in fig. 15. No peaks are presented during stress until rupture of the particle occurs, when the particle explodes. If the contact surfaces of particles are the poles,

(*) The term 'explosion' indicates a process characterized by low deformation of the particle before rupture and a complete dispersion of broken fragments.

optical analysis shows that the fracture surfaces are clean cuts along longitudinal lines. Near the poles where the highest stress concentrations are built up, the fracture surfaces are conchoids, and never a ''flight of stairs'' formation, see fig. 16.

o For this type of particle entire kernels are only present from time to time. It is necessary to remember that the particles type C have carbon-glass kernels and no buffer-layers.

3. Discussion

The question can be raised as to whether this crushing behaviour is typical of the behaviour of all coated particles, and then if it is of possible use for quality control and characterization.

For example, whereas the particles type A and B are from industrial or pilot scale production, the particles type C are from a laboratory coater, where special care has been taken to carefully control all fluidization parameters.

The examination of the crushing modality and of the fracture surface, may reveal some intrinsic characteristics of pyrocarbon coatings which are derived from the manufacturing process, thus giving rise, in some cases, to stratification of the deposits.

Such a stratified structure is demonstrated by stressing the coating by compression.

It would be of interest to correlate the number of layers to the coating ''story''; it is to be noticed that the number of layers is not necessarily identical to the number of existing ones, because of the peculiar distribution of stress created by the crushing procedure.

In UO_2 coated particles the fact that the kernel remains whole after crushing, can be ascribed to the combined effect of the presence of the buffer layer and of the free volume generally present between buffer layer and kernel. Typical crushing behaviour of a UO_2 kernel coated only with a buffer layer is shown in fig. 17. It has already been observed [1] that the fragments of coatings are biangular and if the stressed points of particles are considered as poles, fracture occurs along longitudinal lines.

4. CONCLUSIONS

The experimental results at present observed demonstrated the very important contribution of optical analysis for coating characterization: hence it is our intention to utilize optical analysis as a criterium for this purpose. At the present stage of investigation , a highly isotropic coating seems to assure a better mechanical behaviour before irradiation. But it is our opinion that improvements can be made in the deposition of pyrolytic carbon coatings, especially in the realization of a well improved and controlled fluidization process. In this way it will be useful to compare data of mechanical behaviour before and after irradiation of lots manufactured with different furnaces and techniques.

5. ACKNOWLEDGEMENTS

We wish to acknowledge the photographic work of J. Dubois and C. Agace and the technical assistance of N. Wächter in executing the crush tests is recognized.

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[1] W. DELLE - K. DRITTLER - G. HAAG und H. SCHIFFERS, ''Abschätzung der Spannungen in der Hülle gedrückter beschichteter Partikeln'', (KFA) Jül-569-RW, Jan. 1969.

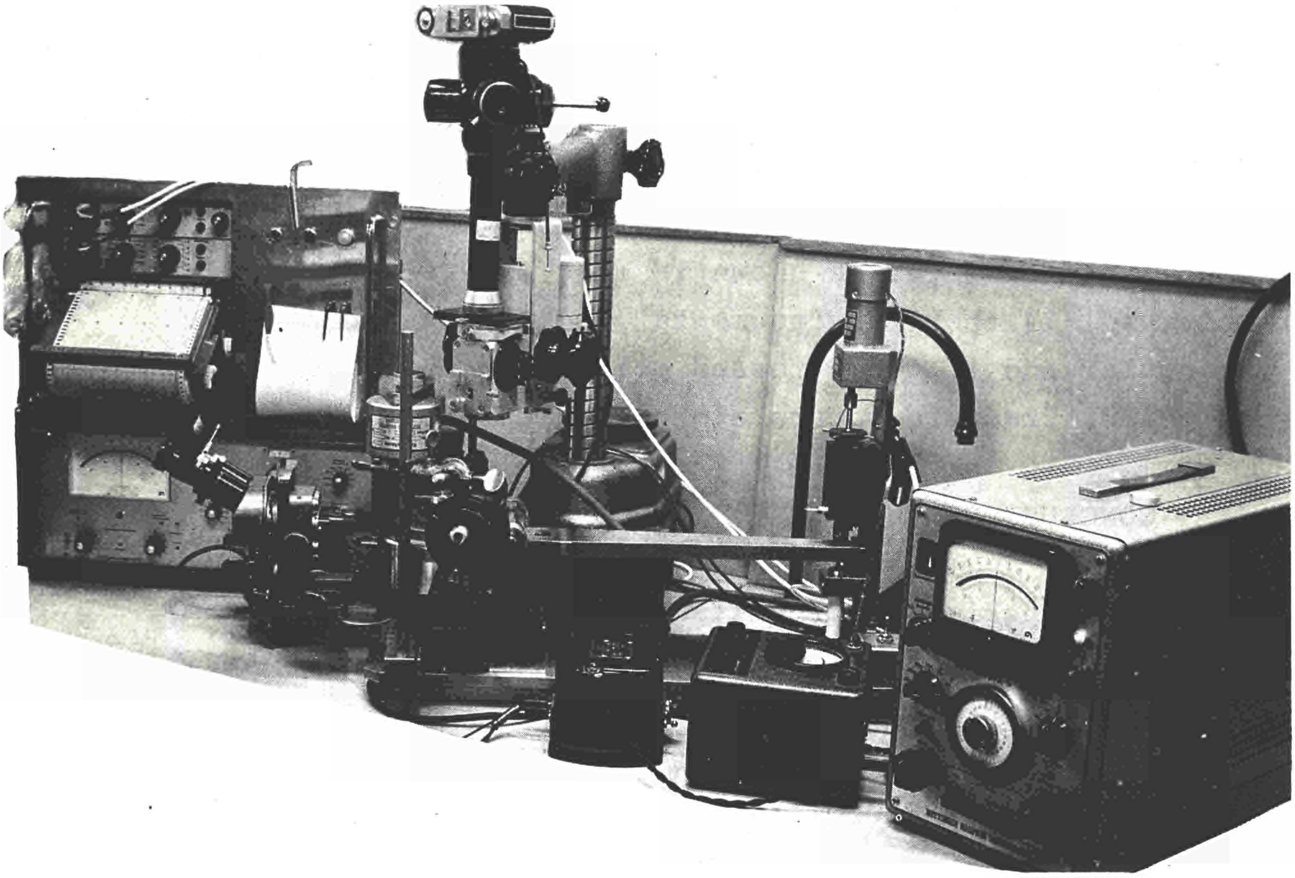


Fig. 1 View of the crushing strength assembling.

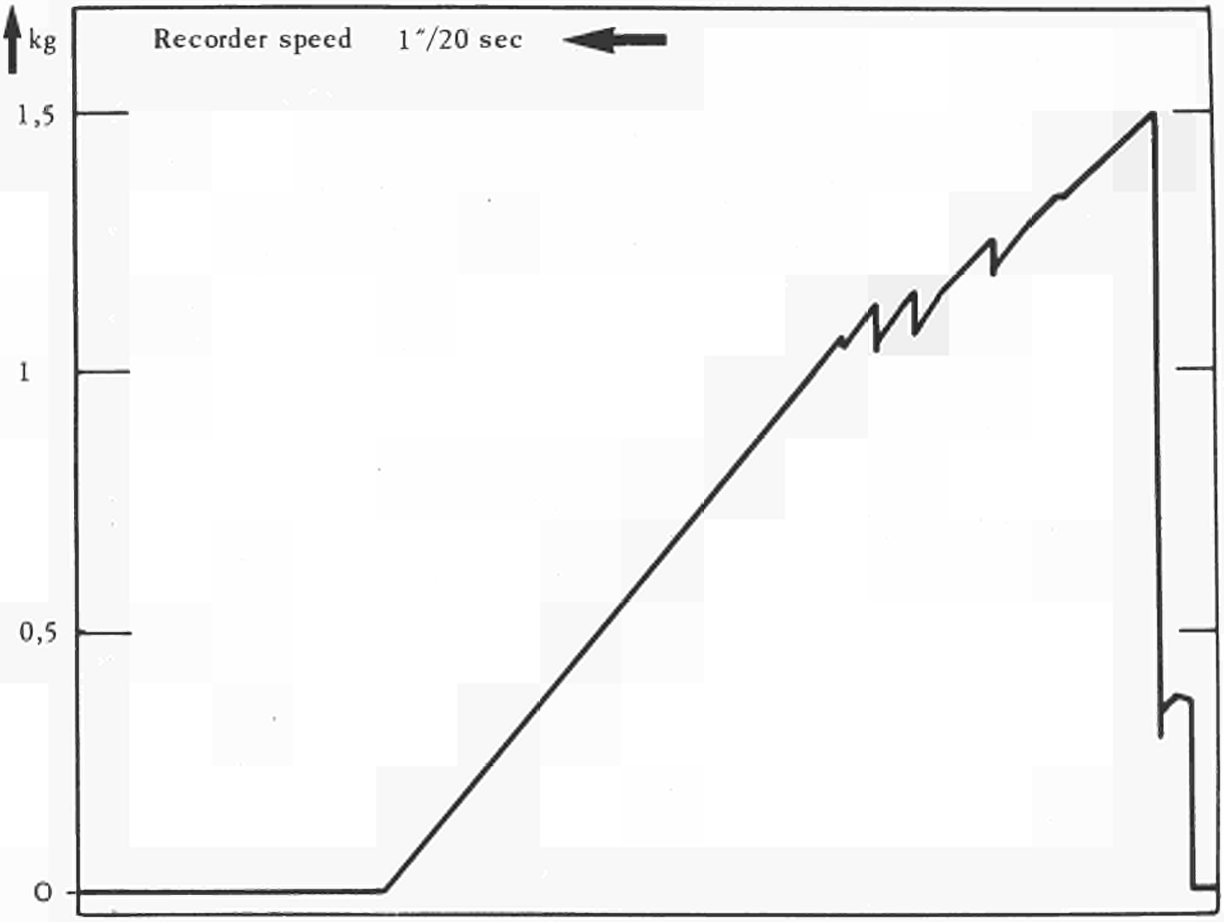


Fig. 2 Typical elastic deformation under stress of a coated particle type A.

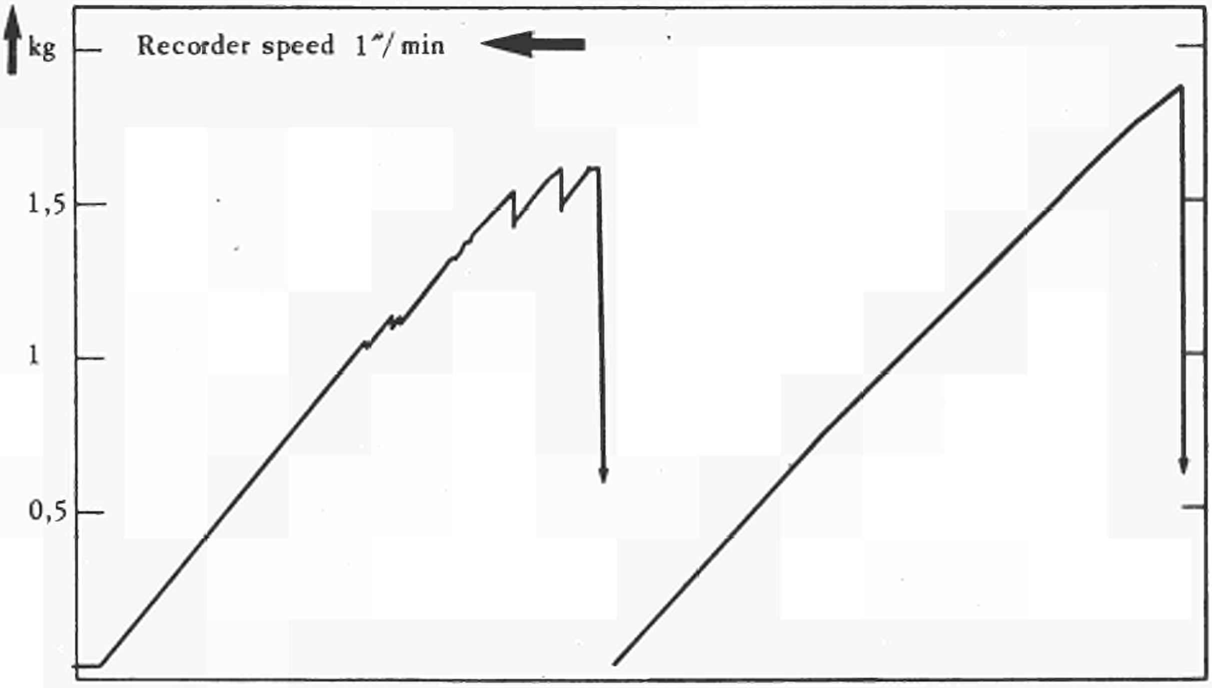


Fig. 3 Repeated stress test.

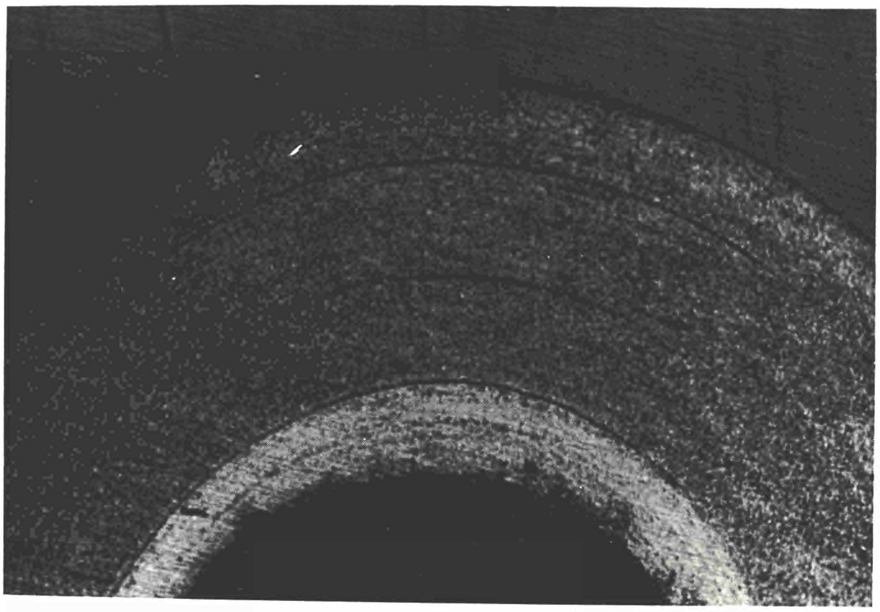


Fig. 4 Micrograph of a particle section.

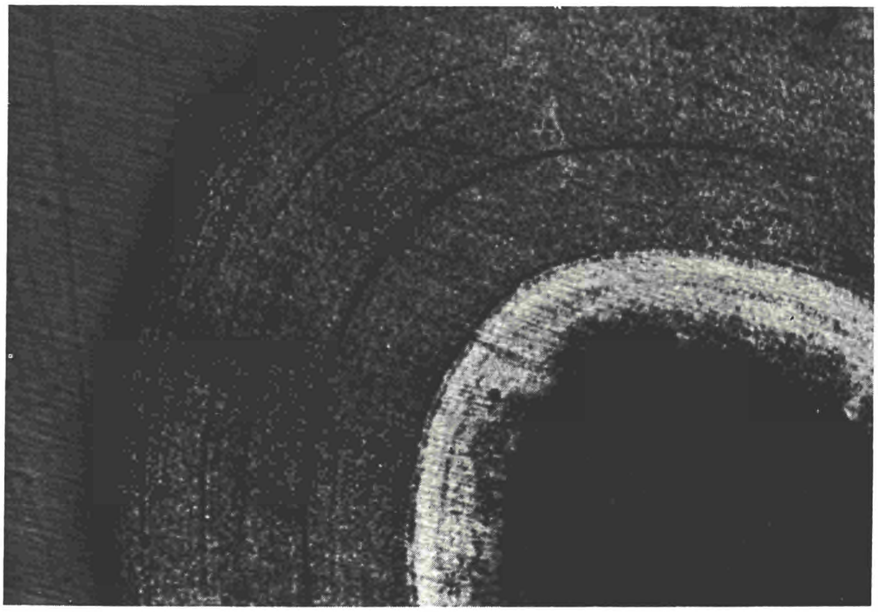


Fig. 5 Micrograph of a particle section.

Fig. 6 Micrograph of crushed particles type A showing delamination processes.

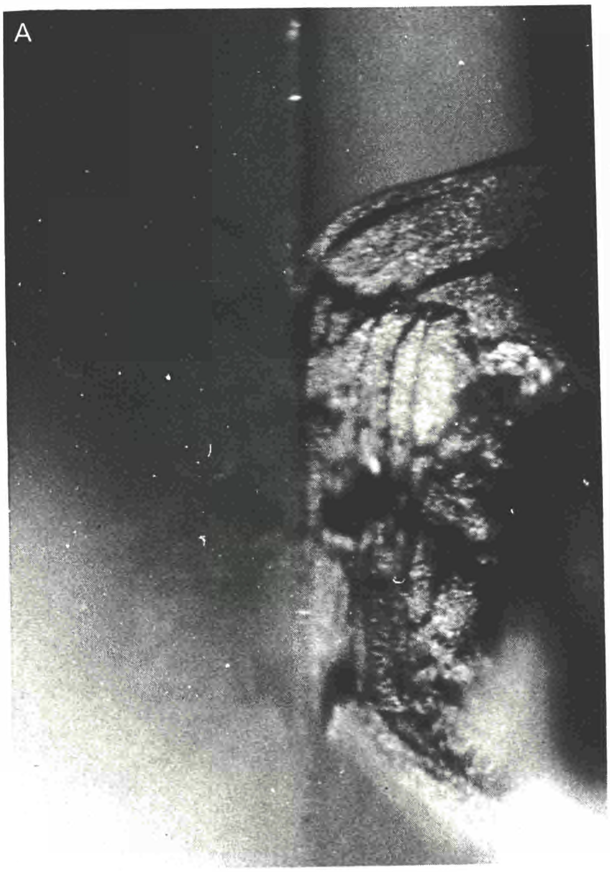
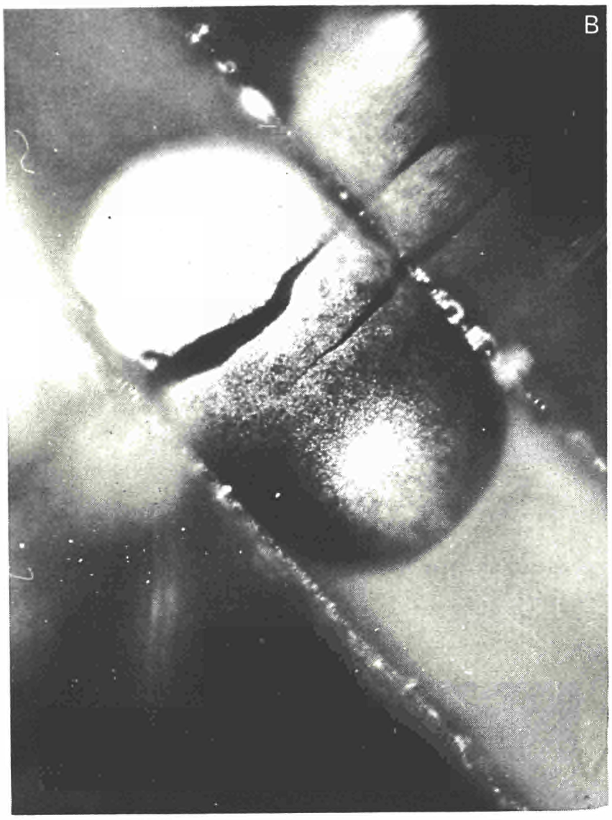
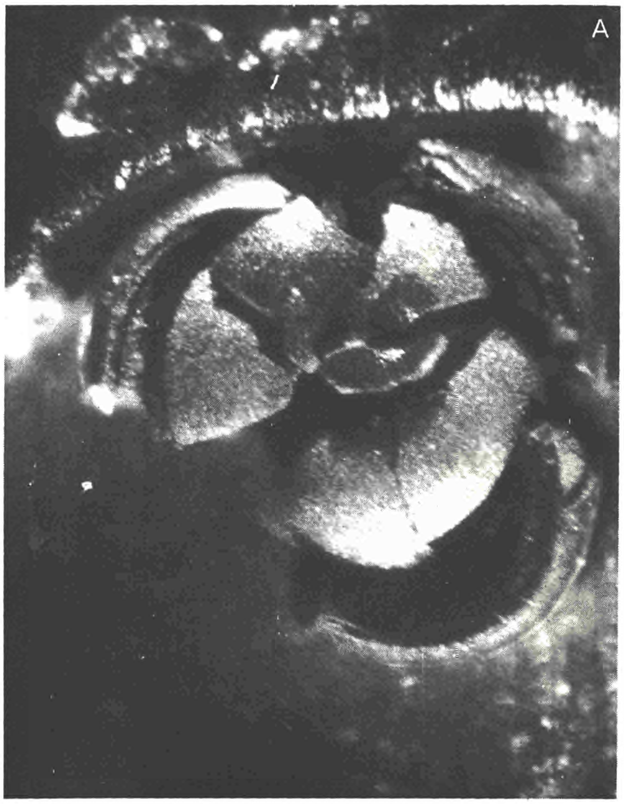


Fig. 7a Crushed particle under stress.

Fig. 7b Crushed particle after discharge .

Fig. 8 a) Collapsed coated particle,
b) Deformation of a stressed particle at rupture.





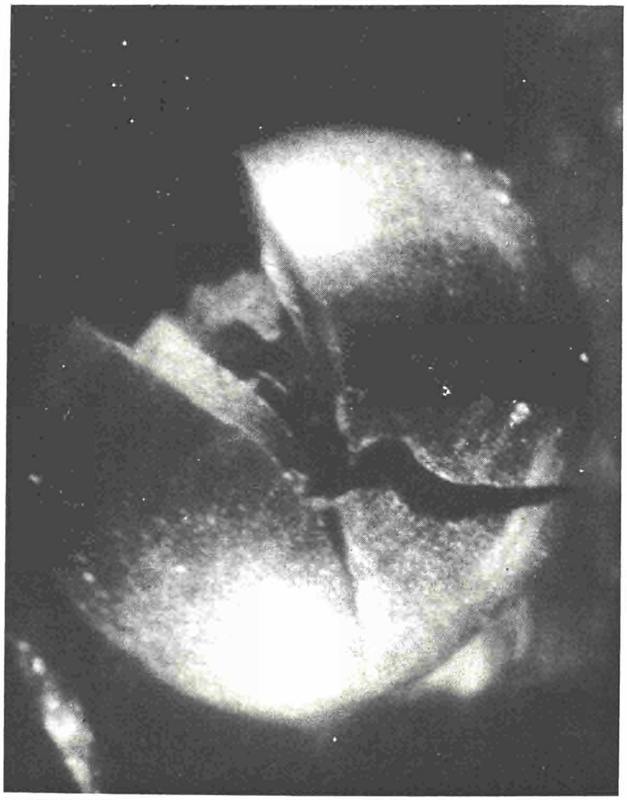


Fig. 10 Coated particle type A crushed.

Fig. 9 Coated particle type A crushed.

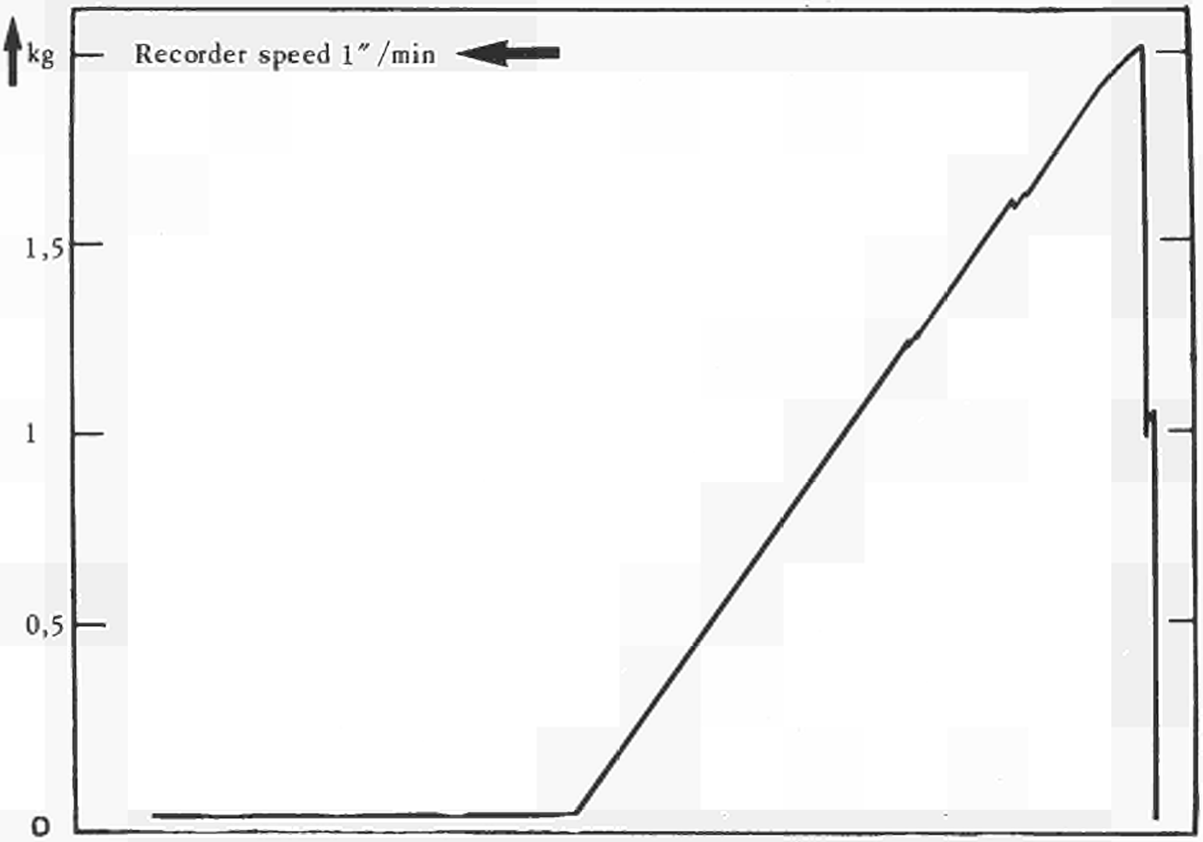
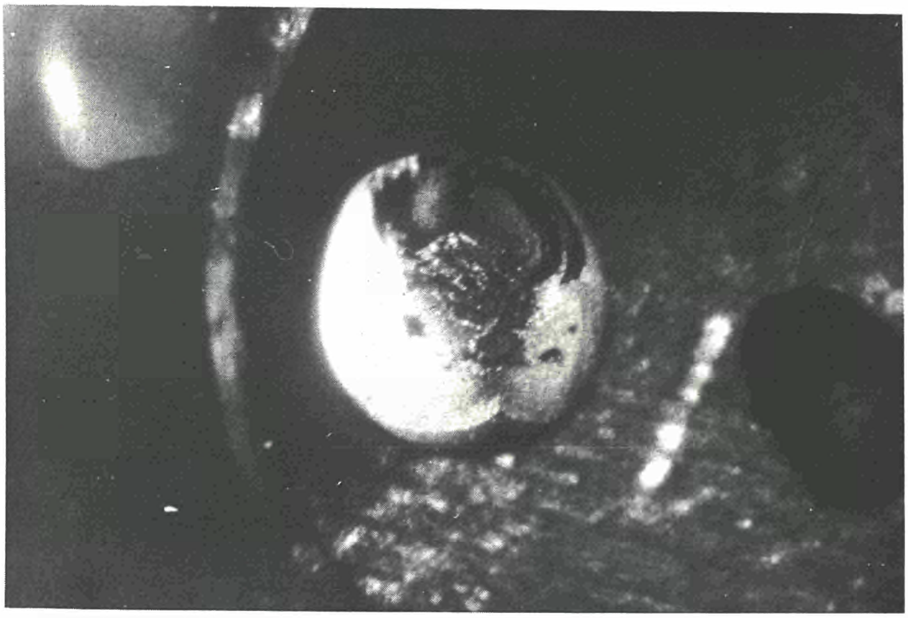


Fig. 11 Typical elastic deformation under stress of a coated particle type B.



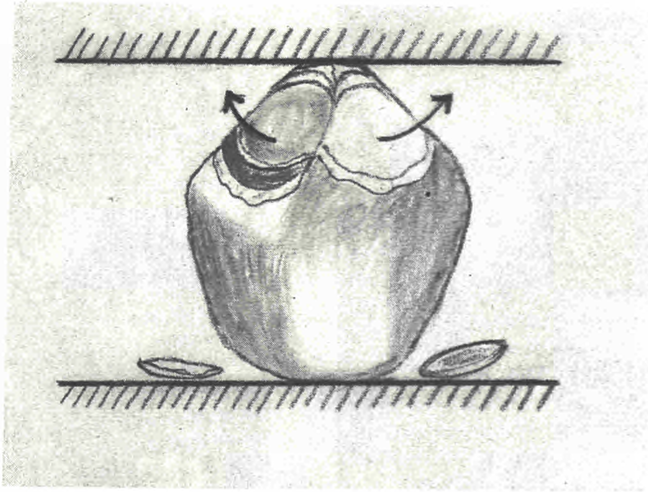


Fig. 12 Presentation of chip effect in B type particle.

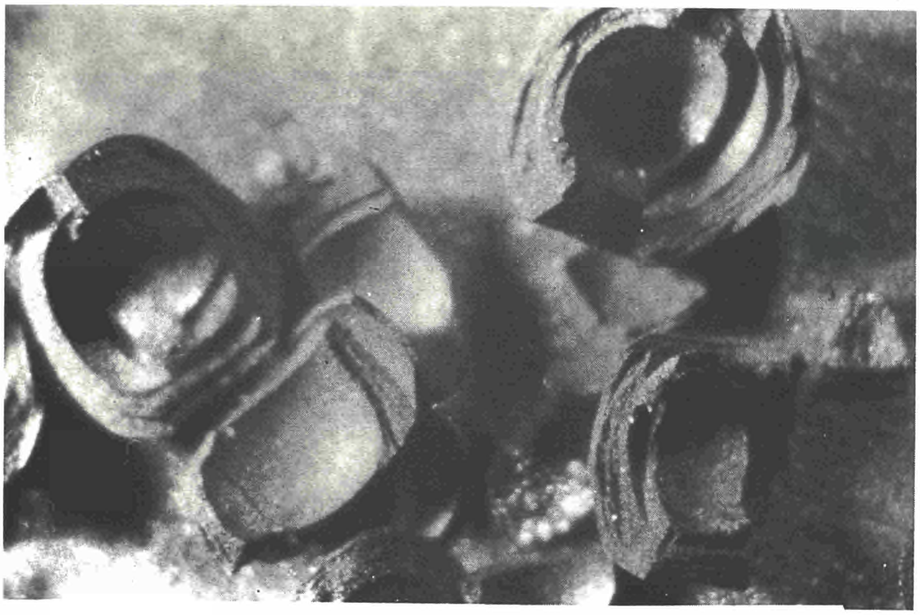


Fig. 13 Flight of stairs present in fracture of B type particle.

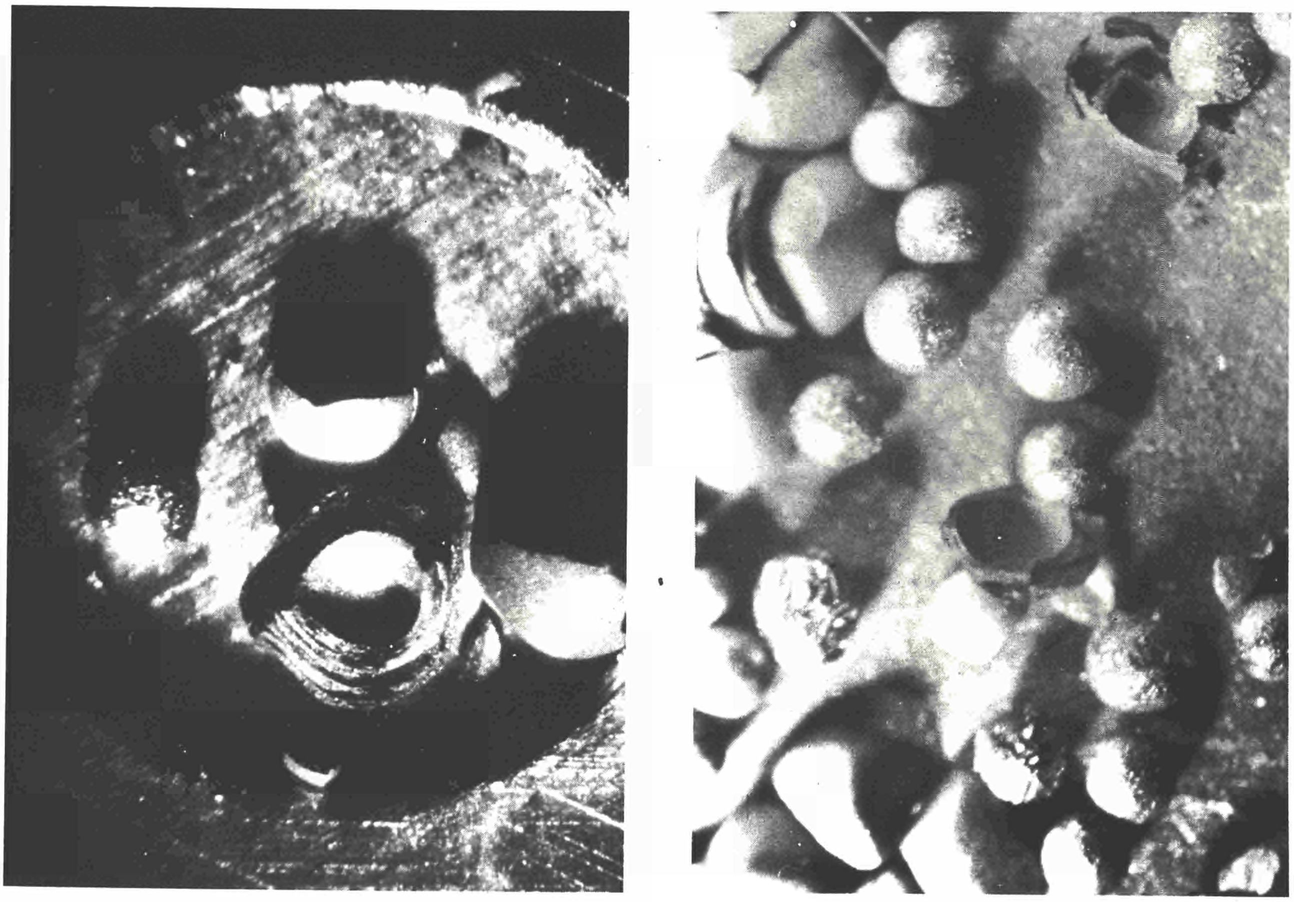


Fig. 14 View of B type Kernel present after crushing.

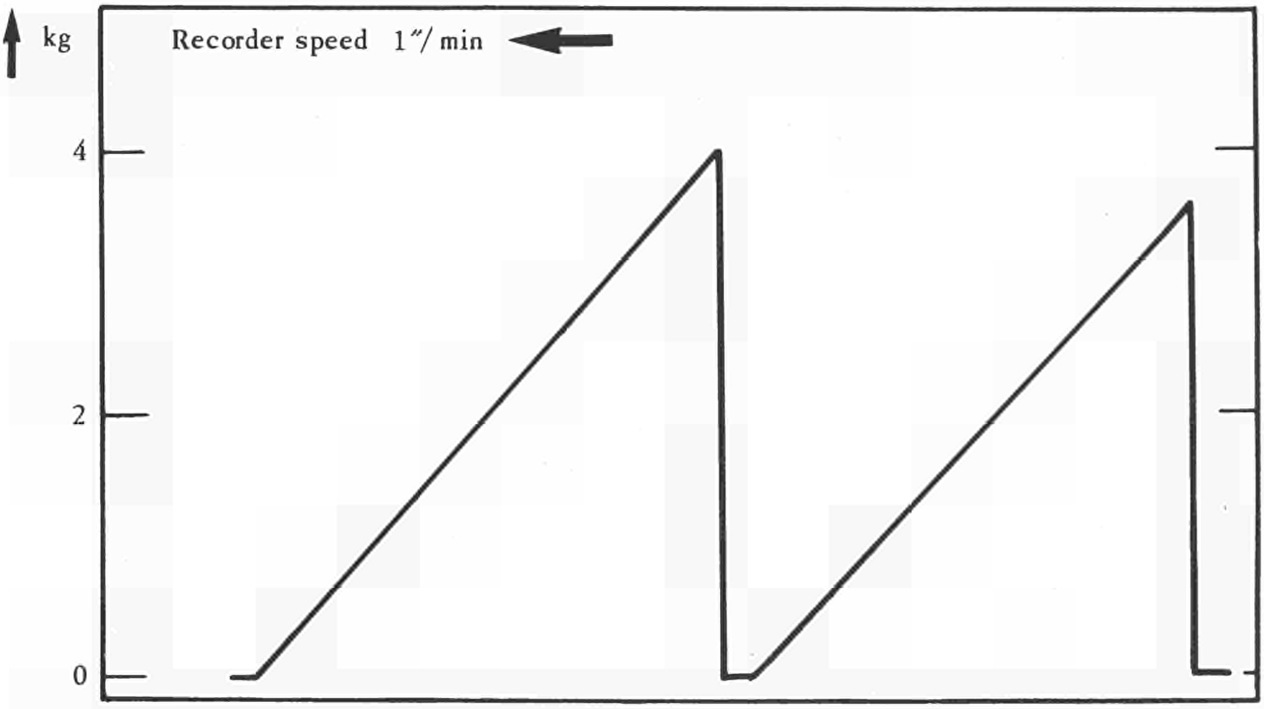


Fig. 15 Typical elastic deformation under stress of two coated particles type C.

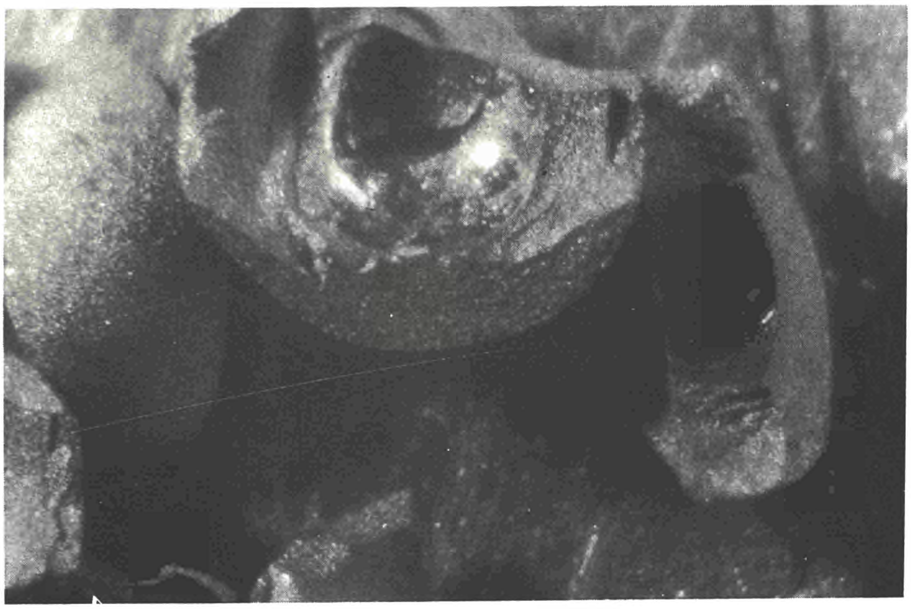


Fig. 16 Micrograph of a crushed C type coated particle.

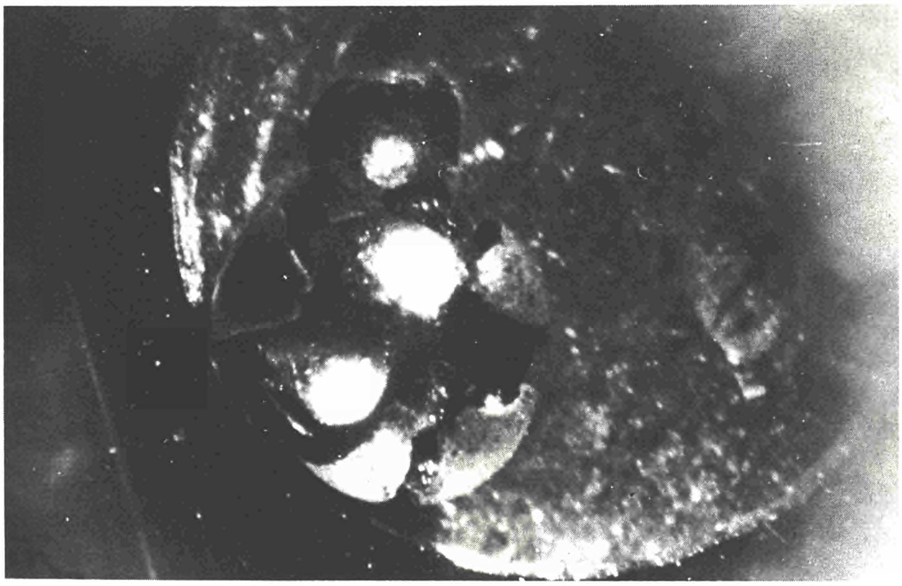


Fig. 17 Crushing behaviour of a UO_2 Kernel coated only with a buffer layer.

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>
> Alfred Nobel

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