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URANIUM DISTRIBUTION AND URANIUM DIFFUSION THROUGH THE PYROCARBON COATING IN UO_2 COATED PARTICLES

by

G. BUSCA, A. DRAGO, P. FENICI and W. HUBER

1970

Joint Nuclear Research Centre
Petten Establishment — Netherlands

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Actually a uniform distribution of uranium through the coating except for the buffer layer has been observed.

Both experimental results indicate that the uranium diffusion for the range of temperature and type of coated fuel particles considered, is entirely controlled by the kernel composition.

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ABSTRACT

The uranium diffusion through the pyrocarbon coating of a UO_2 kernel has been studied in the temperature range of 1600-1900°C. The experimental data fit an equation of the form $D = 4.77 \times 10^{-4} \exp(-86000/RT)$. The uranium distribution through the coating has been evaluated by means of a micro-grinding technique carried out in our laboratory.

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KEYWORDS

URANIUM
DIFFUSION
CARBON
URANIUM OXIDES
TEMPERATURE
KERNELS
COATING
BUFFERS

INTRODUCTION

For an evaluation of the initial release of fission products of coated fuel particles it is necessary to establish the amount and the distribution of uranium in particle coatings.

The alpha-counting method and the recoil release measurement which are generally employed are able to give information of coating contamination up to a limited depth. Furthermore, the alpha counting result depends considerably on the thorium content.

In the study of fission product release during irradiation it is important to know the uranium released from coated fuel particles as a function of irradiation temperature, if the contribution of the different phenomena to fission product release is to be determined.

Out of these considerations (and for the program of research established in our laboratory) the uranium distribution through the coating of UO_2 coated particles and the uranium diffusion through the coating has been investigated in the 1600-1900^{o}C temperature range. The range of temperature was imposed by other programmes of investigation.

1. URANIUM DISTRIBUTION THROUGH THE COATING *)

The coated fuel particles which have been tested consisted of UO_2 kernels on which three pyrocarbon coatings have been applied: first, the buffer layer, obtained by pyrolysis of C_2H_2 at 1380°C and with a deposition rate of 386 µ/h, secondly a thin pyrocarbon layer obtained from CH_4 decomposition at a coating temperature of 1400°C and a deposition rate of 38 µ/h and finally a high density layer obtained with CH_4 at a coating temperature of 1950°C and a deposition rate of 40 µ/h.

The whole batch was monitored by alpha counting to control the uniformity of the superficial contamination level.

A certain amount of the tested coated particles has been used for the evaluation of uranium distribution and the rest for the study of uranium diffusion. For the evaluation of the uranium distribution through the coating we have utilized a method developped in our laboratory.

The main feature of this method is a micro-grinding device which is described in the following. The micro-grinding machine presented in fig. 1 consists of a pivoted arm (T) with a supporting block (A) fixed at one of the extremities. A cylindrical sample holder (B) can be connected to the supporting block. A horizontal slide (C) having a standard surface (D) can be moved under the sample holder. The surfaces of the slide of the sample holder and of the supporting block are parallel to each other. Finally, an inductive displacement transducer (K) is rigidly fixed to the support of the device. The mouvable part of the instrument (K) is kept in contact with the bottom surface of the block (A).

*) Manuscript received on 15 April 1970

The coated particle to be analysed is stuck at the top surface of the sample holder (B).

When the sample holder is connected with (A) a measurement of the diameter of the coated particles is done on the standard surface (D).

On the slide a strip of emery paper is fixed. When the coated particle is put on the paper the slide is moved and leaves a track. The normal force exerted on the coated particles can be varied by moving a weight (P) along the ax of the pivoted arm (T). When one track is left on the emery paper the ground coated particle is placed again on (D) and the variation of height produced by the grinding operation is read.

Before the emery paper is taken away it is covered with scotch tape to protect the track. The coated particles to be examined are irradiated before the start of the micro-grinding operation, in order to have a sufficient concentration of fission products in the tracks left on the emery paper. The examination is performed by gamma-analysis. From fission product concentrations the amount of existing uranium is determined.

Before the gamma activity levels in the tracks can be related to uranium concentration through the coating, it is necessary to do some considerations concerning the geometry of the sample and the experimental procedure adopted.

It is assumed that the coating is composed by a series of shells 1, 2, 3,....each of them with a different concentration C_1, C_2, C_3.... of fissile materials, but constant within every shell. With reference to fig. 2 the track produced on the grinding-paper from an abrasion depth h_1 contains only material with concentration C_1, in the second step for the abrasion depth h_2 in the track we have material with concentration C_1 and C_2.

If the h_i values and the diameter of the particles are known it is possible by geometrical considerations to determine the mean concentrations C_i. With our grinding machine it is possible to control 1 µm of depth.

The La-140 amount was determined in all tracks by gamma counting. For this determination it was necessary to await equilibrium between Ba140 - La140, before the grinding operation. The gamma analysis of tracks was carried out with a 3"x3" NaI(Tl) crystal connected to a 4000 channel pulse height analyser. On 50 coated particles analyzed the uranium distribution is uniform through the coating, but changes markedly in the buffer layer range. E. Selleck (1) with an other experimental technique and with different types of coated particles observed that the contamination gradient is uniform from the particle surface up to about 70% of the pyrolytic carbon coating

Because of the experimental technique used by Selleck it was not possible to remove the coating completely down to the buffer layer. Since the particles were not precisely identical, removal of minute amounts of buffer would nullify results.

It is believed the difference in uranium distribution in the buffer is due to chemical reactions between the kernel and the buffer layer during the deposition process and to the fact that the buffer layer acts as a sink for fission products during irradiation. The fraction of uranium observed in the high density pyrocarbon coating of our coated particles was 4×10^{-4} of the total amount. In fig. 3 the uranium distribution of four coated particles is shown.

The same distribution of uranium was observed when the mica technique has been used (2). In fig. 4 and 5 the uranium concentration observed with both methods on a special batch of coated particles is presented, where an external uranium contamination was produced artificially.

2. URANIUM DIFFUSION THROUGH THE COATING

The amount of coated particles for the tests was divided into lots of about 2 grams. Each lot was introduced with graphite powder into a graphite sample holder and hermetically sealed in an inert atmosphere. The graphite powder was added to obtain a better uniform distribution of temperature during the heat treatment. The temperature chosen for the treatments was 1600°C, 1700°C, 1800°C and 1900°C.

After the heat treatment the coated particles have been separated from powder by sieving and cleaned by ultra-sound technique. No change was observed in the superficial contamination level by alpha counting measurements. The uranium distribution through the coating has been controlled on several coated particles which received a thermal treatment at 1900°C for 100 hours. No change in the level and in the distribution of uranium through the coating was observed. Each sample holder and the corresponding graphite powder was irradiated in the reactor. After a suitable cooling time the amount of La140 was determined by gamma analysis. The experimental data fits very well the equation

$$D = 4.77 \times 10^{-4} \exp\left(-\frac{86.000 \pm 8000}{RT}\right)$$

as can be seen in fig. 6. The value found for the activation energy is in good agreement with the value of 85 ± 7 Kcal/mol reported by J.F. Marin and P. Contamin (3) for the self-diffusion of uranium in UO_2.

3. CONCLUSIONS

All the experimental results obtained from uranium diffusion and distribution through the coating indicate that the uranium migration is entirely controlled by the nature of the kernel in the case of UO_2 coated particles.

The agreement between our results and those of Selleck concerning the uranium distribution through the coating obtained with different types of particles suggest that simpler methods than previously described can be applied for the determination of uranium concentrations in the coatings.

A practical method could be for instance a catcher foil method using coated particles individually wrapped in Al-foil.

4. ACKNOWLEDGEMENT

We wish to acknowledge the technical assistance of N. Wächter in executing the nuclear measurements.

5. REFERENCES

(1) E. Selleck, GAMD-8325 (15 May, 1968), "Coating contamination in fuel particles".

(2) B. Chinaglia D.P. Report 536 (April 1968), "Method for measuring the uranium contamination on fuel particle coatings".

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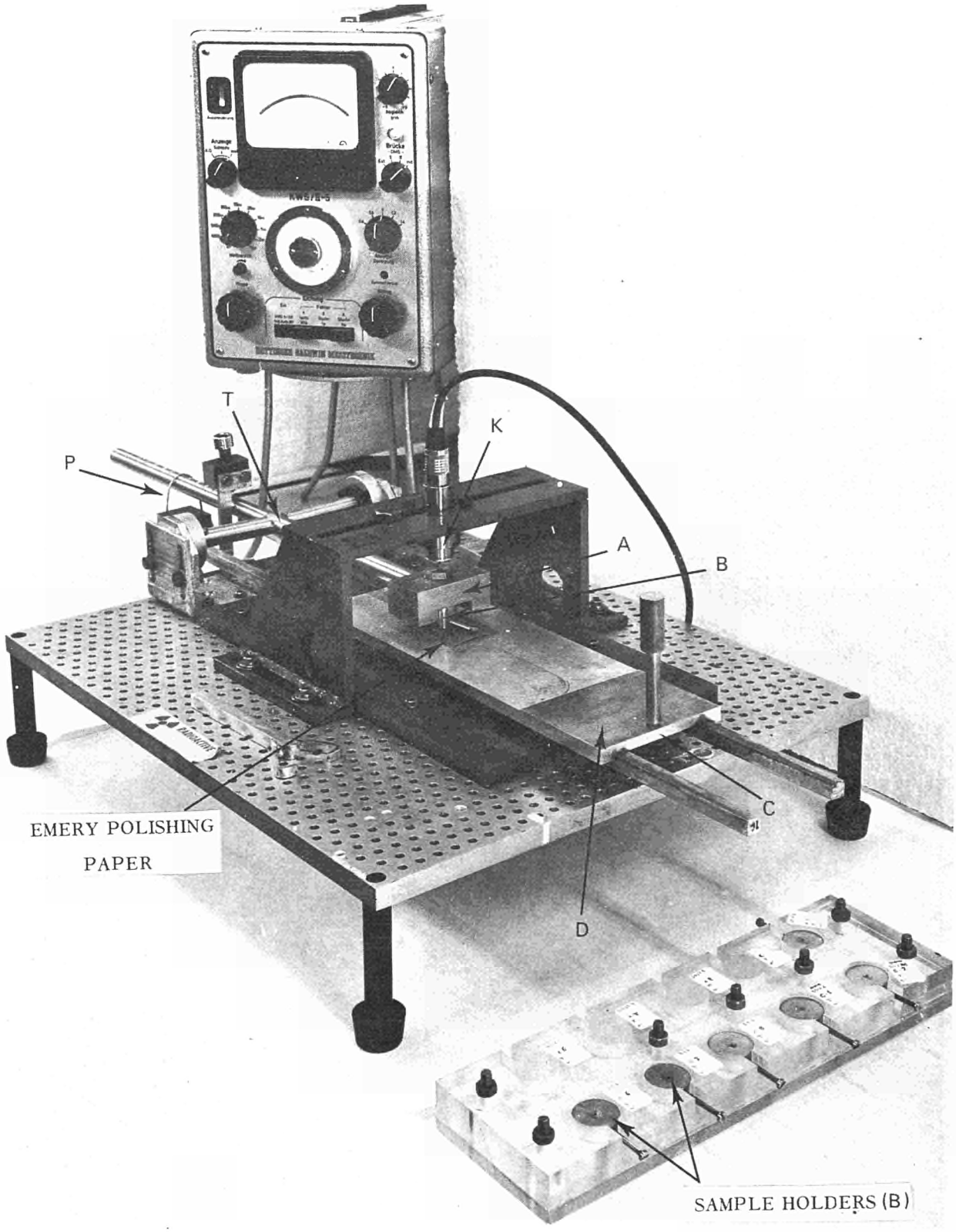

Fig. 1 Micro grinding device

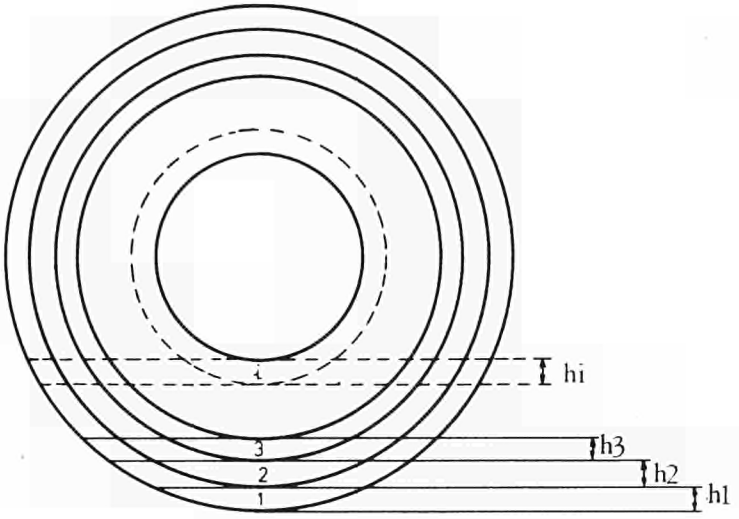

Fig. 2 Schematic cross-section of a coated particle.

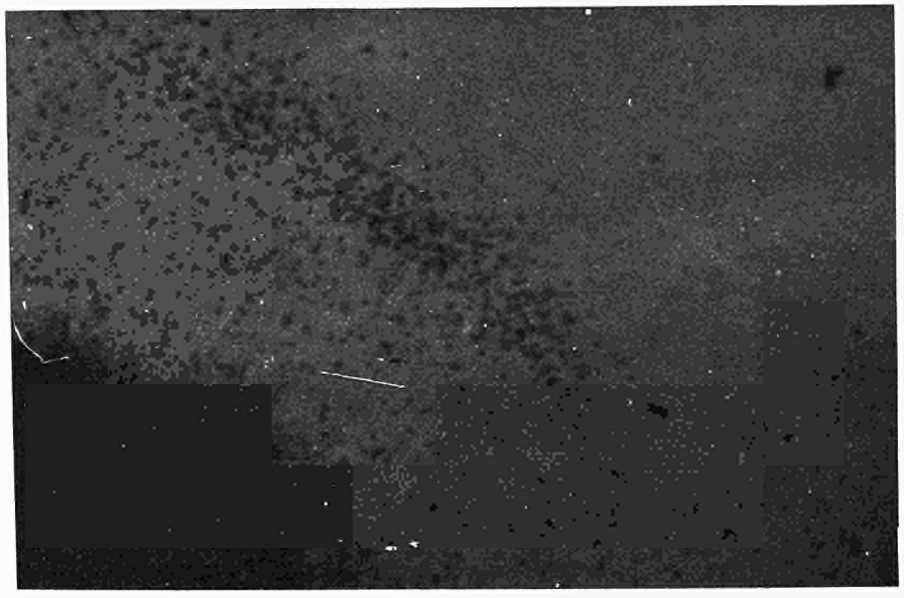

Fig. 4 Uranium concentration observed with the quartz technique.

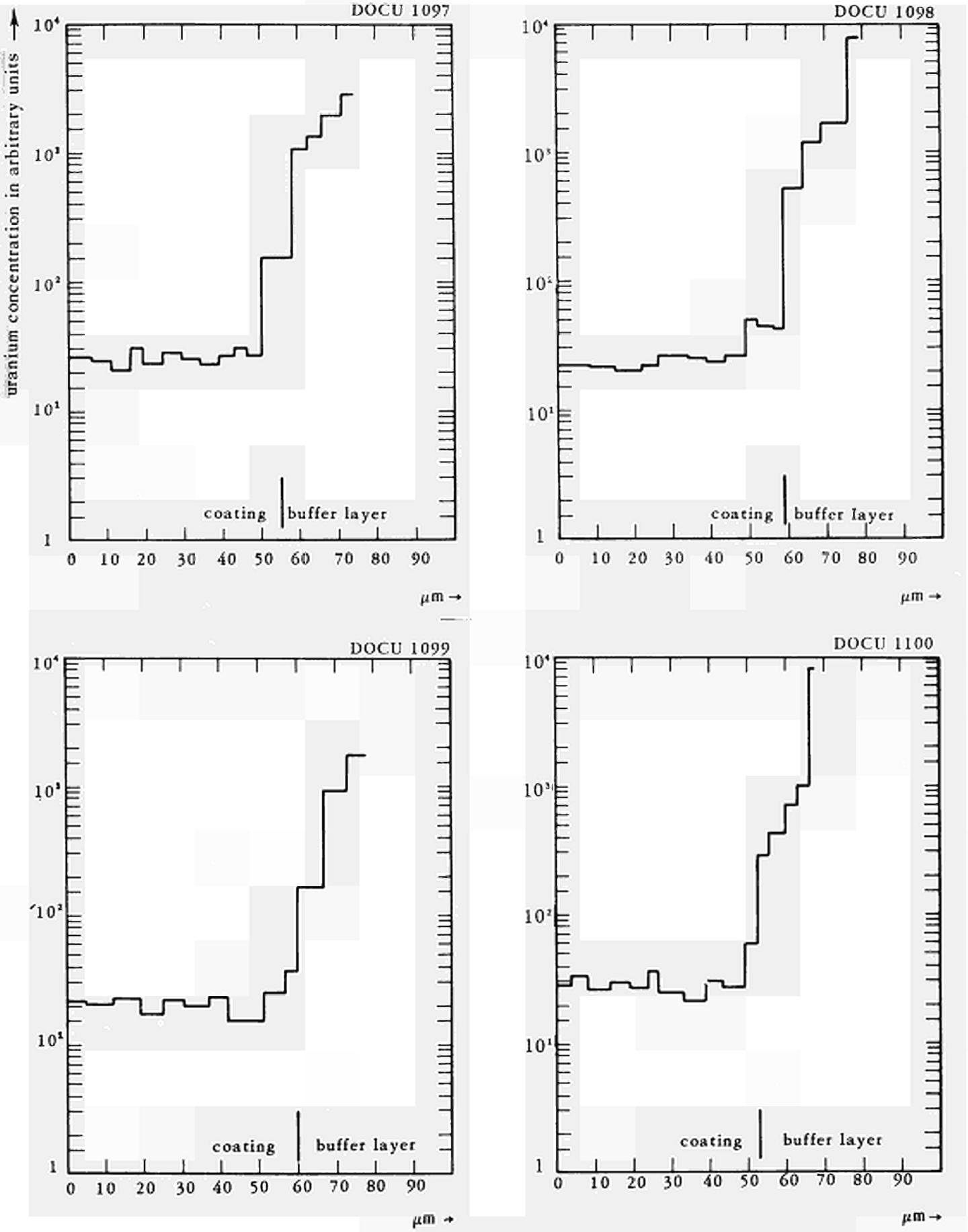

Fig. 3 Typical Uranium distribution observed into the coatings of four coated particles.

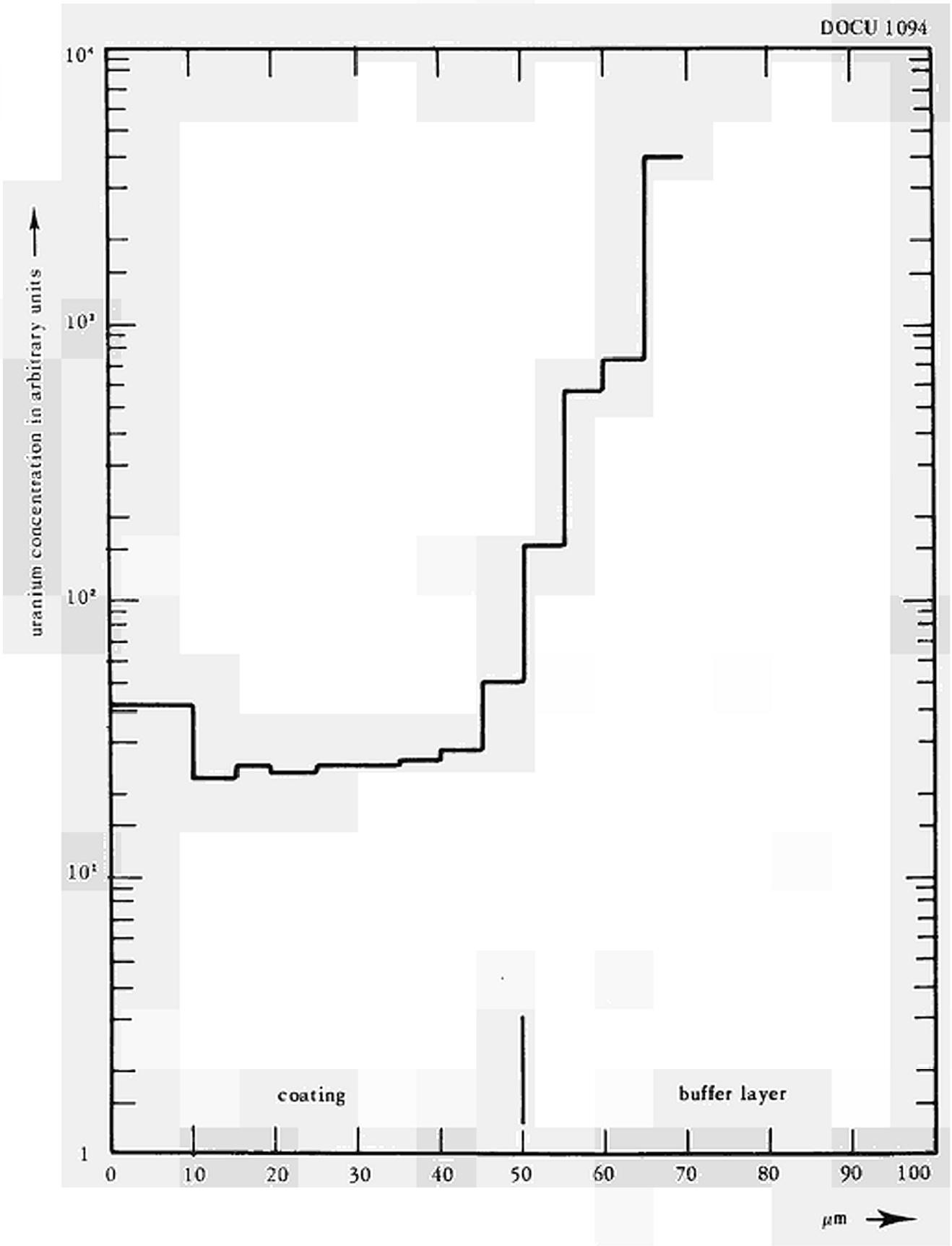

Fig. 5 Uranium concentration observed with the micro-grinding technique.

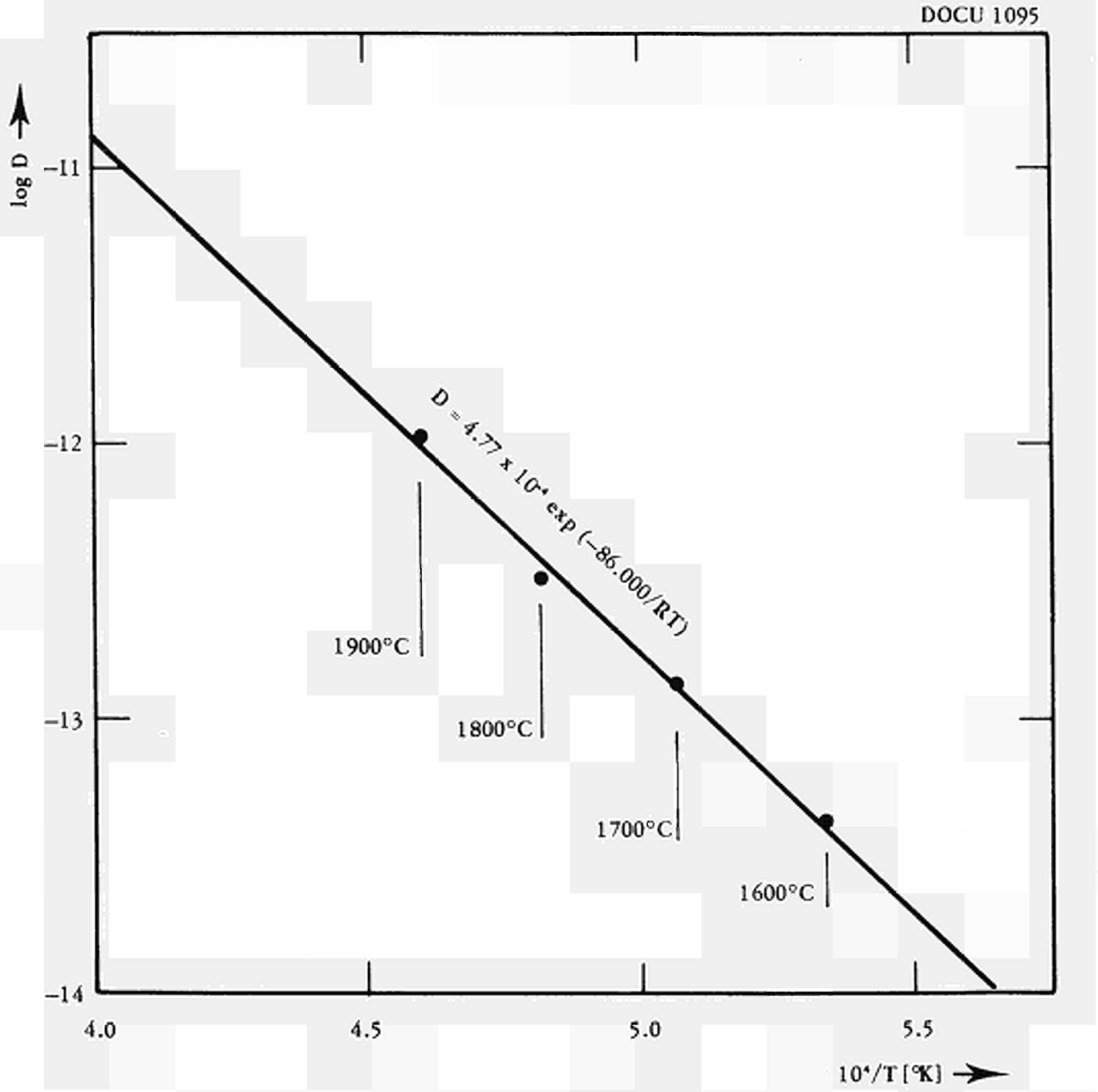

Fig. 6 Arrhenius-plot of uranium diffusion in UO_2 pyrocarbon coated particles.

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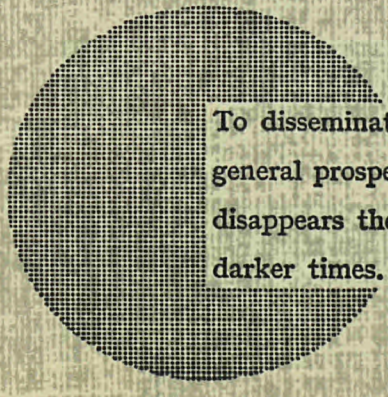

To disseminate knowledge is to disseminate prosperity — I mean general prosperity and not individual riches — and with prosperity disappears the greater part of the evil which is our heritage from darker times.

Alfred Nobel

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