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RESEARCH ESTABLISHMENT

LUCAS HEIGHTS

H.T.G.C. FUEL AND MODERATOR MATERIAL IRRADIATION PROGRAMME

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B.S. Hickman

Sydney, January, 1958.

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Abstract

Probable mechanisms of irradiation damage in fuel and moderating materials of interest to the H.T.G.C. Reactor programme are discussed. A programme is outlined for investigations into irradiation damage in H.T.G.C. fuels and moderators. The experimental methods to be used are briefly outlined.

CONTENTS

1. INTRODUCTION

The H.T.G.C. Reactor research programme depends to a large extent on the development of a fuel element which will withstand high burnup. The programme described in this paper is aimed at the investigation of the effects of irradiation on various possible types of fuel elements with the final object of specifying one or more fuel element types which would appear to satisfy the requirements of the design group. These fuels can then be tested further on a larger scale in the proposed gas cooled loop in HIFAR.

The present conception of the H.T.G.C. system involves the dispersion of the fissile and fertile materials through the moderator. Systems based on beryllium metal, beryllium oxide and graphite as moderators have been considered. The following are the composition ranges of interest.

(i) Beryllium moderator – U: Th: Be: : 1: 30: 2000–4000

(ii) Beryllia moderator – UO_2: ThO_2: BeO: : 1: 30: 2000–3000

(iii) Graphite moderator – U: Th: C: : 1: 30: 4000–6000

Various combinations of these systems are possible. For instance a UO_2: ThO_2: BeO fuel could be canned in a beryllium or graphite case, or a U: Th: Be fuel could be used in conjunction with a BeO moderator.

To reduce the number of variables which have to be considered, the preliminary irradiation programme will concentrate on the testing of possible fuel element components rather than the testing of small scale fuel elements.

The various types of systems will now be considered in more detail together with the irradiation programme associated with each.

2. BERYLLIUM MODERATED SYSTEMS

2.1 General

Fuel elements based on beryllium metal are likely to consist of a dispersion of the intermetallic compounds UBe_{13} and $ThBe_{13}$ in beryllium metal, integrally canned in beryllium metal.

The main advantages of this system are that fabrication techniques are fairly well established and the fission products should be retained in the fuel, thus enabling a clean gas circuit to be maintained.

The disadvantage of the use of beryllium apart from irradiation damage aspects are mainly limitations on temperature due to: –

(i) High vapour pressure of beryllium at elevated temperatures which could cause mass transfer.

(ii) Compatability problems if CO_2 is used as a coolant.

2.2 Irradiation Damage in Beryllium Based Fuels

Break down of the fuel under irradiation could occur by several mechanisms, the most important of which are considered to be the following: –

2.2.1 Swelling

Uranium metal has been found to increase in volume under irradiation due to the accumulation of fission product gases in cavities which then expand. This effect can be very serious at elevated temperatures, and volume increases of over 100% have been observed (7). A similar effect would be expected to occur in a beryllium based fuel, not only due to the fission product gases but also due to helium and tritium produced by the following nuclear reactions in beryllium:-

(1) $Be^9(n, \alpha)\ Li^6\ (n, \gamma)\ Li^7 \rightarrow He^4 + H^3$

Threshold = 0.71 MEV

(2) $Be^9\ (n, 2n)\ Be^8 \rightarrow 2\ He^4$

Threshold = 1.85 MEV

Cross sections for these fast neutron reactions are not accurately known but experiments by Richman (1) have indicated that approximately one atom of gas will be produced by these reactions per uranium fission for dispersion compositions in the range under consideration. This can be compared with 0.25 atoms of fission product gas which are produced per uranium fission.

The gaseous fission products will escape from the fissile particles into the matrix to an extent determined by the fissile particle size (See figure I.) It has been shown that at the temperatures under consideration diffusion is unimportant and that escape occurs substantially by recoil (8). Preliminary experiments indicate that it may be difficult to produce dense UBe_{13} particles of diameter greater than 30-50 μ, at which size a considerable proportion of the fission product gases will escape into the matrix.

Alder (2) has carried out some calculations on the swelling of beryllium metal under a fast neutron flux, which indicated that the effect could be serious at temperatures above 600°C. Similar calculations have been carried out by the author for a dispersion type fuel using improved creep data (3) and an improved treatment developed by Foreman (4). (See Appendix I.) The results are shown in Figure 2 and indicate that for 100% burnup of the original fissile investment swelling will not become appreciable (i.e., of the order of 1%) until 650 - 700°C. However, as this is near to the proposed operating temperature, this problem will need careful investigation.

2.2.2 Thermal Stress Effects

It is not expected that thermal stresses will result in failure of a beryllium metal fuel element. At the temperatures under consideration the stresses should be relieved by creep if they become excessive. The effects of irradiation on thermal stress will be considered in more detail in connection with the UO_2.ThO_2.BeO system (see section 3.2.1).

2.2.3 Effect of Solid Fission Products

This effect will depend on the fissile particle size which will determine the amount of fission products which will escape into the matrix. It is possible that they could cause embrittlement of the matrix but even at the high burn ups anticipated in these fuels, the concentration of fission products will be low and it is not expected that the effect will be serious.

2.3 Irradiation Programme

It is not possible to take account of all possible variables in the initial tests and hence the samples have been chosen to give information on those variables which are considered most important, bearing in mind the most likely effects of irradiation as discussed in the preceding section. The variables covered are:-

(i) Composition (i.e. heavy metal content)

(ii) Rate of burn up and hence thermal stress

(iii) Fissile particle size

(iv) Temperature

(v) Effect of restraint

All samples will be irradiated in duplicate and accelerated damage over that expected in a reactor will be obtained by replacement of some thorium atoms with uranium atoms. (N.B. This should not affect the physical and mechanical properties of the samples,) and by irradiation in a higher effective flux. This will enable fission product damage equivalent to 100% burn up of the original fissile investment in an actual fuel element to be obtained in 3 - 4 months. Initially tests will be carried out in a 4V position in HIFAR where the fast flux is low ($2 \times 10^{}$) and hence helium production in the beryllium will be much lower than in the reactor. However, as soon as hollow fuel element facilities are available in HIFAR, some or all of the tests will be repeated in these facilities where the rate of helium production should be comparable with the fission product gas production.

The samples for the initial tests are outlined in Table I. Samples 1-8 cover the composition range likely to be of interest, i.e., from about 10 volume % to 50 volume % of heavy metal compounds which corresponds to atom ratios of 1: 30: 4000 to 1: 30: 1000. Samples 7 and 8 have the maximum concentration of intermetallic compounds which can be incorporated in a dispersion. Samples 3 and 4 have been chosen as reference samples and samples 9-18 all have the same composition as 3 and 4. Samples 9 and 10 have a higher fissile content which will give a higher burn up rate and thermal stresses. Samples 11-14 are designed to investigate the effect of temperature. Samples 15 and 16 have a coarser fissile particle size than the reference samples. Samples 17, 18 are designed to test the effect of a thick beryllium can on restraining the swelling (if any) of the fuel core.

3. BERYLLIA MODERATED SYSTEMS

3.1 General

In this system the fuel and fertile material will be dispersed as oxides through a beryllium oxide matrix. At present the main interest in this system is for use as a fuel material inside a beryllium or impermeable graphite can. However, it may also be possible to develop an impervious BeO can for use with this fuel.

3.2 Irradiation Damage in Beryllia Based Fuels

Considerable interest is being shown in ceramic fuels at the present time due to their excellent stability under irradiation. UO_2 and $UO_2.ThO_2$ samples have been irradiated to 1.5% burn up of metal atoms with little or no damage occurring (Cf 0.15% metal atom burn up in an H.T.G.C. system.) Little data of any significance has been obtained on the irradiation behaviour of the $UO_2.ThO_2.BeO$ system. In some early work at Argonne (5), samples of $UO_2.BeO$ containing 2 and 10 wt% UO_2 were irradiated at pile temperature ($< 100^{o}C$)

for 24 and 63 day periods. Maximum burn up was 0.03% of uranium. The crushing strength of the samples was approximately doubled, the Youngs Modulus reduced by 1% to 2% and the density decreased by 0.5 – 1.0%. The major part of these changes had occurred after 24 days irradiation. The most striking change was in thermal conductivity which was reduced to 25% of the initial value after 24 days and to 20% after 63 days. Annealing at 675°C in vacuo removed only 15% of the induced change.

Some samples of 12.5 wt% UO_2 in BeO were irradiated by Harwell in Windscale in liquid sodium to 1.25% burn up of uranium atoms. All these samples disintegrated during irradiation but it is thought that this was due to the fact that the specimens were porous and infiltration of sodium occurred, causing the disintegration. (A similar effect has been observed with UO_2).

Possible causes of irradiation damage in ceramic fuels are discussed in the following sections.

3.2.1 Thermal Stress Effects

It is considered that the most likely cause of failure in ceramic fuels is cracking and disintegration due to thermal stresses. Fuel ratings in the proposed system have been chosen to give thermal stresses at the surface of the order of half of the ultimate tensile strength at the operating temperature. However, these have been calculated assuming the thermal conductivity of the fuel material is the same as unirradiated beryllia. Although the presence of discrete UO_2, ThO_2 particles should only affect the thermal conductivity by 10–15% (6) the effect of irradiation may cause a large change as is indicated by the results quoted above. Changes in the other properties which determine the thermal stress are expected to be small compared with the changes in thermal conductivity. The changes in thermal conductivity could be due to two causes.

(i) Damage to the crystal lattice by fast neutron and fission fragment bombardment

(ii) Distortion of the lattice by fission products.

The first of these effects may anneal out at least to a certain extent at the operating temperatures although the Argonne results indicate that little annealing occurs at 675°C. The effect will depend to a certain extent on fissile particle size. With a large fissile particle size most of the fission fragment bombardment effects and fission product accumulation effects will be limited to the particles but with a fine fissile particle size, the beryllia matrix will be affected.

Thermal stress effects are likely to be more serious with ceramic fuels than metallic fuels because there is little chance of relief of the stresses by creep. This will only occur in the case of beryllia at temperatures above 950°C.

3.2.2 Effect of Solid Fission Products

Effects due to the accumulation of fission products and helium are not expected to be as serious for ceramic fuel elements as for metallic fuels. Previous experience indicates that the open lattice of ceramics will accommodate the extra fission product atoms with little change in dimensions occurring. An exception is the effect mentioned above on physical properties such as thermal conductivity which are particularly structure sensitive.

3.3 Irradiation Programme

As mentioned earlier, it is not possible to cover all variables and for the initial tests the following variables have been considered: -

(i) Composition

(ii) Thermal stress and rate of burn up

(iii) Fissile particle size

(iv) Temperature

These were chosen, bearing in mind that thermal stresses are considered to be the most important factor affecting the irradiation stability of the fuels. As with the beryllium metal samples accelerated damage will be obtained by replacing some thorium with uranium and damage equivalent to 100% burn up of the fissile content of reactor fuel will be obtained in 5 – 6 months. Initially, tests will be carried out in a 4V position but will later be extended to a fast neutron facility to investigate the effects of helium production.

The samples for the initial tests are outlined in Table 2. Samples 19–26 cover the composition range likely to be of interest, i.e., atom ratios of U: Th: Be of 1: 30: 500 to 1: 30: 3000. Samples 21 and 22 have been chosen as reference samples and samples 27–34 have the same composition as 21 and 22. Samples 27–30 are designed to investigate the effect of increased thermal stress. Samples 31 and 32 will be irradiated at a higher temperature where the stresses could be relived by creep. Samples 33 and 34 will have a coarser fissile particle size which as discussed earlier may affect the irradiation behaviour.

4. GRAPHITE MODERATED SYSTEMS

Due to the very large size which is required in order that graphite moderated systems can be self sustaining there is not much interest in a completely graphite moderated system for small reactors. However, it is quite possible to use smaller quantities of graphite in conjunction with beryllium or beryllium oxide.

The main effort in this direction will be concentrated on developing fabrication techniques for producing, by extrusion and baking, graphite cylinders containing up to 50 wt% of heavy metals. In the absence of established fabrication techniques, it would be premature at this stage to detail an irradiation programme. However, as soon as samples are available they will be irradiated in a similar manner to the ceramic specimens.

5. EXPERIMENTAL METHODS

5.1 Irradiation Apparatus

The equipment is based on a principle which has been used successfully by the author in the U.K. and also by various workers in America. Briefly, the fissile sample is separated from a water cooled jacket by an annulus of suitable thermal conductivity. Depending on the heat output of the sample, the type and thickness of the thermal barrier is chosen so that, during reactor operation, the fissile heat is sufficient to raise the temperature of the specimen to the desired level.

The ceramic specimens will be contained in a graphite case which will be supported inside a water cooled stainless steel can by a ceramic support. The annular space between the graphite case and the stainless steel can will be filled with helium and the width of the gap will be chosen in each case so that the sample will be irradiated at the desired temperature.

The beryllium metal specimens will be immersed in liquid sodium inside an inner stainless steel can which will be separated from the water cooled outer can by a helium gas gap of suitable thickness. The immersion of metallic fissile specimens in sodium is standard practice in the U.K. and U.S.A. and has several advantages, namely:-

(i) Accurate temperature measurement can be made by immersing a thermocouple in the sodium, without introducing a thermocouple pocket in the actual sample.

(ii) Density changes of the sample can be accommodated by the sodium without affecting the width of the gas gap and hence the temperature of the sample.

(iii) More highly rated samples can be irradiated, hence shortening the irradiation time.

Sixteen (16) samples will be irradiated at one time in a 4V facility in HIFAR. The samples will be positioned in the hole so that control rod movement should not affect the flux on the samples and hence the operating temperature should be fairly constant. The ceramic samples will be irradiated in duplicate and one of each type will have a thermo-couple pocket in the centre of the specimen. The beryllium metal samples will also be irradiated in duplicate and temperature measurement will be made on all the samples.

Cobalt thermal flux monitors will be incorporated in each specimen.

5.2 Pre-Irradiation Examination

Pre-irradiation examination of all samples will include:-

(i) Accurate dimensional measurements

(ii) Density and porosity determinations

(iii) Visual examination and photography

Physical property measurements (strength, thermal conductivity, etc.,) will, of course, be made on samples prepared under identical conditions.

5.3 Post Irradiation Examination

Post irradiation examination of the samples will cover the following:-

(i) The stainless steel can will be pierced in vacuo and gas samples taken. These samples will be analysed by Analytical Services by mass spectrographic methods.

(ii) The sample will be removed from the can and examined macroscopically.

(iii) Dimensional, density and porosity measurements will then be made.

(iv) Thermal conductivity measurements will be made on selected samples.

(v) Selected samples will be polished and examined under the microscope.

(vi) Selected samples will be subjected to compression or bend tests.

(vii) Calculated burn ups will be checked by analysis of the cobalt monitors and by fission product analysis of some samples using γ spectroscopy.

(viii) An attempt will be made in the case of the ceramic specimens at least to investigate the distribution of solid fission products, particularly caesium and strontium, through the graphite case and stainless steel can.

(ix) Annealing experiments will be undertaken to investigate fission product escape and density changes and possibly changes in other physical properties.

(x) Stored energy measurements will be made on some samples.

(xi) Selected samples will be made available to Chemical Processing Group for processing studies.

Due to probable limitations in space and availability of apparatus, it may be possible only to carry out some of these investigations immediately on completion of the irradiation. Samples will, however, be stored and further tests carried out as apparatus and space becomes available.

6. PROGRAMME ON MODERATING MATERIALS

In addition to the programme on fuel materials, an extensive investigation is planned into the effects of irradiation on the properties of beryllium and beryllium oxide. Samples will be irradiated cold and at controlled elevated temperatures in a high fast flux in a hollow fuel element. Post irradiation measurements will include the following:-

1. Macroscopic and microscopic examination
2. Dimensional and density checks
3. Tensile and bend tests
4. Stored energy
5. Electrical resistivity and thermal conductivity
6. X-ray diffraction
7. Determination of helium and tritium contents

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TABLE 1 – BERYLLIUM METAL DISPERSIONS

Sample No.	Atom Ratio			Weight %			Volume %	Density	Dimensions		Heat Output Watts	Irradiation Temperature °C	Thermal Stress p.s.i.	Metal Atom % Burn up after 0.9x107 Secs
	U	Th	Be	U	Th	Be	U,Th,Be13		Dia.	Height				
1 2	3	28	4000	1.63	15.0	83.3	11.15	2.1	1	2	170	600	2,300	0.025
3 4	3	28	3000	2.05	19.0	79.0	16.3	2.25	1	2	225	600	3,100	0.033
5 6	3	28	2000	2.79	25.7	71.4	24	2.45	1	2	350	600	4,800	0.05
7 8	3	28	1000	4.35	40.1	55.6	46	3.02	0.75	1.5	255	600	4,800	0.1
9 10	6	25	3000	4.11	16.9	79.0	16.3	2.26	1	2	440	600	8,200	0.066
11 12	3	28	3000	2.05	19.0	79.0	16.3	2.26	1	2	225	500	3,100	0.033
13 14	3	28	3000	2.05	19.0	79.0	16.3	2.26	1	2	225	700	3,100	0.033
15 16	3	28	3000	2.05	19.0	79.0	16.3	2.26	1	2	225	600	3,100	0.033
17 18	6	25	3000	4.11	16.9	79.0	16.3	2.26	0.75	1.75	215	600	3,350	0.066

TABLE II – BERYLLIA DISPERSIONS

Sample No.	Atom Ratio			Weight %			Volume %	Density	Dimensions		Heat Output Watts	Irradiation Temperature °C	Thermal Stress	Metal Atom % Burn Up after 1.3×10^7 secs
	U	Th	Be	UO_2	ThO_2	BeO	U, ThO_2		Dia.	Height				
19 20	2	29	3000	0.69	9.2	90.1	3.0	3.08	1	3	140	800	3,700	0.035
21 22	2	29	2000	0.92	13.1	85.9	4.4	3.10	1	3	185	800	4,900	0.048
23 24	2	20	1000	1.61	23.0	75.4	8.4	3.49	1	3	350	800	9,300	0.095
25 26	2	29	500	2.59	37.0	60.4	15.7	4.01	0.75	2	240	800	9,350	0.18
27 28	4	27	2000	1.83	12.2	85.9	4.4	3.18	1	3	365	800	9,700	0.1
29 30	6	25	2000	2.75	11.3	85.9	4.4	3.18	1	2	355	800	14,160	0.15
31 32	2	29	2000	0.92	13.1	85.9	4.4	3.18	1	3	185	800	4,900	0.048
33 34	6	25	2000	2.75	11.3	85.9	4.4	3.18	1	2	355	1000	14,160	0.15

APPENDIX

Swelling of Beryllium Metal Fuels

Foreman has calculated the rate of swelling of gas bubbles in an incompressible medium and obtained the following formula: –

$$x = \left(\frac{3n}{2K(n+1)}\right)^{\frac{1}{n}} \frac{3c}{2n} t^{\frac{n+1}{n}}$$

where x = fractional increase in volume in time t,

n,K = creep constants defined by the quasi viscous creep law for uniaxial stress: –

$E = \frac{Tn}{K}$ where E = rate of elongation

T = stress

C = constant related to the rate of production of gas pressure in the cavities.

In deriving this formula the following assumptions were made: –

(i) Material is incompressible and deforms according to the quasi–viscous creep law.

(ii) For small values of x such that $\frac{1}{x^n} << 1$ the interaction of neighbouring cavities can be neglected.

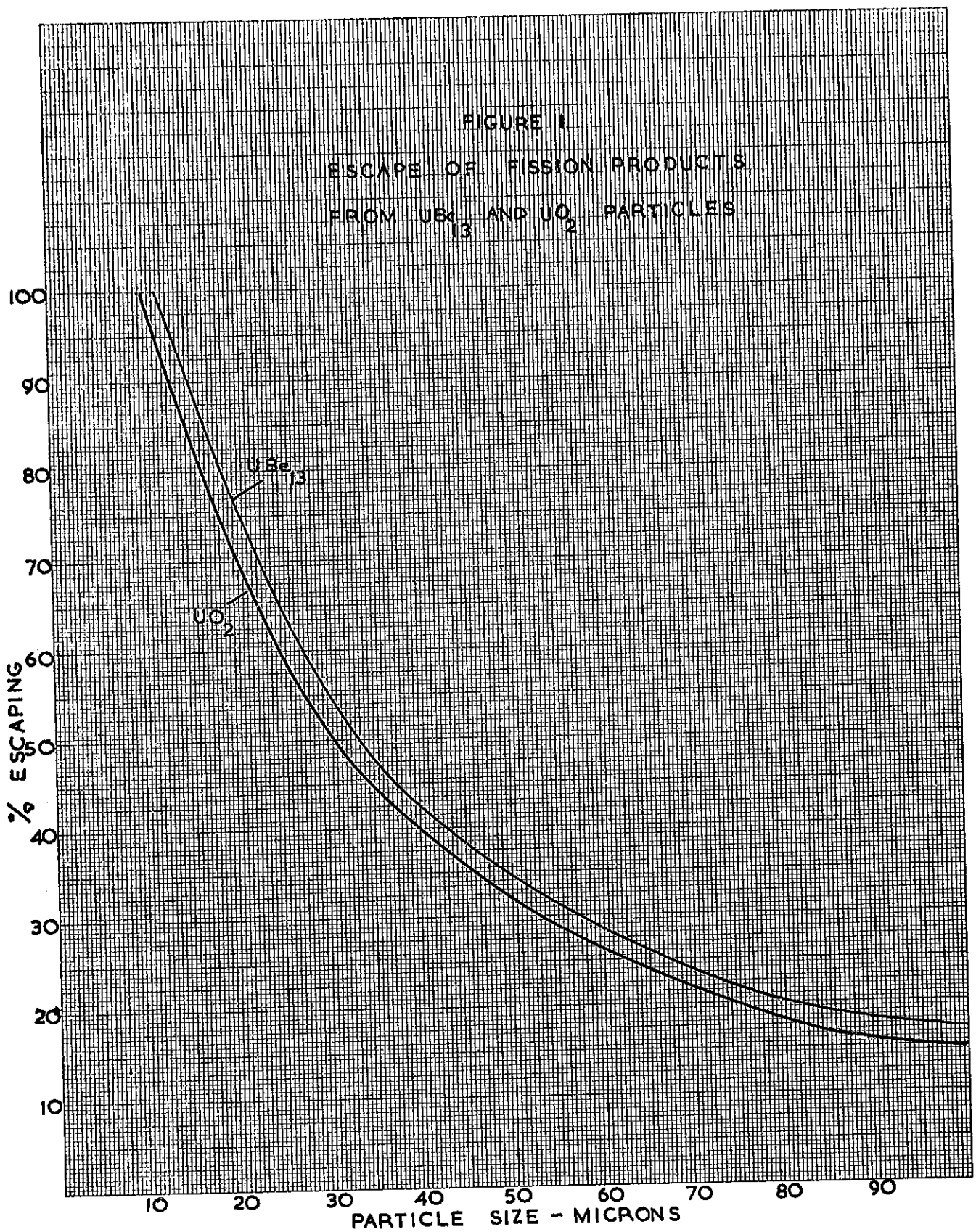
(iii) All the gas produced in the material diffuses to cavities and remains therein.

By plotting creep data for beryllium on a log–log plot values for n and K can easily be obtained. The rate of gas production was calculated assuming the figure for helium production given earlier and substitution was made in the formula to find the swelling after 100% burn up of the original fissile investment in three years. The curve is plotted in figure II.

FIGURE 1

ESCAPE OF FISSION PRODUCTS

FROM UBe_{13} AND UO_2 PARTICLES



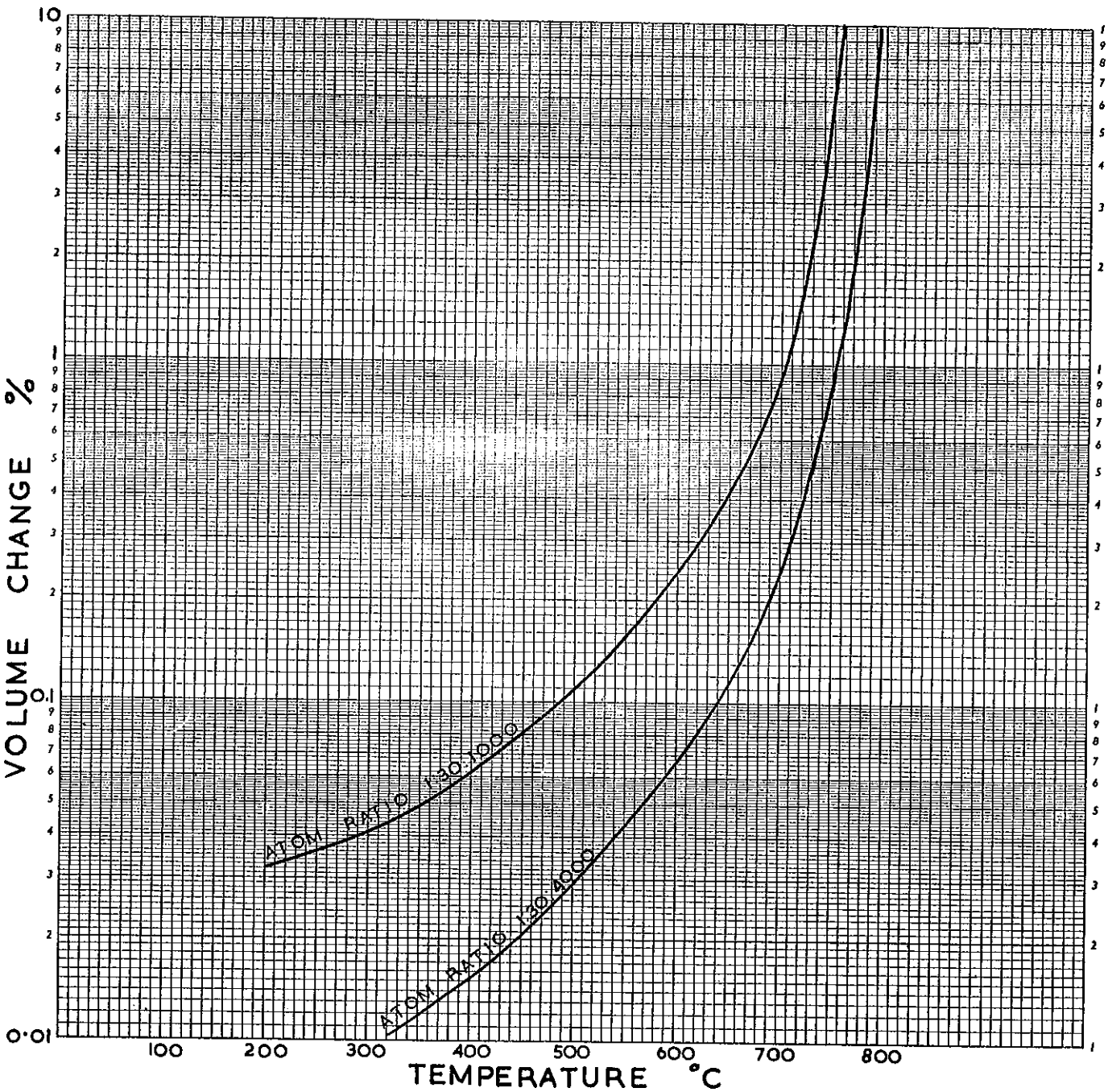


Figure 2. Swelling of H.T.G.C. Fuel element for 100% burn up of U^{233} in three years.