UNCLASSIFIED

AAEC/E. 6.

GROWTH OF BERYLLIUM UNDER IRRADIATION

by

K. F. Alder

SUMMARY

This report gives the results of a calculation estimating the rate of swelling of beryllium metal by the formation of bubbles of helium gas due to irradiation.

1. INTRODUCTION

The (n,α) and (n, 2n) reactions in beryllium both produce helium. This note gives the results of a calculation estimating the rate of swelling of the metal produced by this gas forming bubbles. The calculation makes the following assumptions:-

1. The bubbles are large enough for the operative force resisting their growth to be the creep strength of the material and not its surface tension.

2. The stress produced in the metal is uniformly distributed in the regions between bubbles.

If assumption 1 is unwarranted and the bubbles are in fact very small such that the restraining force due to surface tension is greater than the creep strength of the material, then the answer given by this calculation will be much too large. However, it should be pointed out that the probability of large bubble formation increases markedly with temperature owing to the increase of diffusion rate of the gas, and that a calculation based on similar assumptions has given an answer which agrees with the facts for the swelling of uranium under irradiation at temperatures comparable with those considered here (1).

2. NUCLEAR REACTIONS IN BERYLLIUM

2.1. n, α

Be^9 (n, α) He^6 Threshold 0.71 MeV.

$He^6 \longrightarrow Li^6$ (0.82 secs).

Li^6 (950 b) (n, α) H^3

The cross-section for the primary reaction is less than 2.0% of the total cross-section in the range in which it has been measured, 2 - 4 MeV and 14.1 MeV and does not exceed 50 millibarns.

The "poisoning" effect of Li^6 will be small and will depend on the ratio of thermal to fast flux. Each primary reaction produces 2 atoms of Helium and one atom of Tritium. Taking the worst case (50 mb) and considering the Helium production in 10,000 atoms of beryllium -

$$\sigma c = 50 \text{ m.b.} = 50 \times 10^{-27} \text{ cm}^2$$

No. of He atoms produced per annum = 2 x σc x NPV

$$= 2 \times 50 \times 10^{-27} \times 10^4 \times 10^{14} \times (3.15 \times 10^7) \text{ (secs/year)}$$

= <u>3.15 He atoms per year.</u>

Volume of He produced per gm. of Be

$$= 3.15 \times \frac{N}{10^4} \times \frac{1}{9} \times \frac{22.4 \times 10^3}{N}$$

$$= 0.78 \text{ ccs/gm Be/year at NTP}$$

and No. of Tritium atoms = 1.57/year/10^4 atoms Be.

The radioactivity of the beryllium due to β decay of the tritium may be calculated thus :-

Half life of Tritium = 12.4 years

$$\text{Decay constant } \lambda = \frac{0.693}{12.4 \times 3.15 \times 10^7}$$

and therefore Specific activity per gm. of Be

$$= \frac{3.15}{2} \times \frac{0.6 \times 10^{24}}{10^4 \times 9} \times \frac{0.693}{12.4 \times 3.15 \times 10^7} \times \frac{1}{3.7 \times 10^{10}}$$

$$= 0.5 \text{ curies/gm.}$$

As tritium is a beta emitter, handling of massive beryllium in this state presents no problems. Powdered metal on the other hand would be a hazard.

2.2 n, 2n

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

Threshold 1.85 MeV.

The cross-section for this reaction is apparently very difficult to measure. The value usually taken is 0.2 barns; on this basis, assuming a fast flux of 10^{14} above the 1.85 MeV threshold (which is probably unreasonably high for moderator), we obtain :-

$$\sigma c = 200 \text{ mb} = 200 \times 10^{-27} \text{ cm}^2$$

No. of He atoms produced per annum per 10^4 atoms of Be

$$= 2 \times 200 \times 10^{-27} \times 10^{-4} \times 10^{14} \times 3.15 \times 10^7$$

$$= 12.6 \text{ atoms/year}$$

Volume of He per gm Be per year

$$= 12.6 \times \frac{N}{10^4} \times \frac{1}{9} \times \frac{22.4 \times 10^3}{N}$$

= 3.4 ccs/gm Be/year at NTP

2.3 Total Helium

Therefore total He production from n, α and n, 2n

= 4.18 ccs/gm Be/year at NTP

An estimate of the amount of helium produced in beryllium used as a reactor moderator may be made using some figures given by Dalton (2) and summarised below :-

Uranium 233 : Beryllium ratio = 1 : 2000

Burnup of U233 atoms = 100%

Helium atoms produced, per U233 fission :-

n, 2n, 0.67

Primary n, α, 0.1

Secondary n, α Li^6, 0.1

Total 0.87

Plus 0.1 atoms Tritium

Therefore total atoms of He produced

= 0.87 x no. of U atoms fissioned

Therefore ratio of He produced to Be present = $\frac{0.87}{2000}$

Therefore Volume of He at NTP per gm of Be

$$= \frac{0.87}{2000} \quad \frac{22.4}{N} \quad \frac{10^3}{9} \quad \frac{N}{} = 1.08 \text{ ccs/gm Be/for 100\% burnup.}$$

Some experiments have been done on Helium production in beryllium irradiated at a fast flux of 2 x 10, for 140 hours (3), the amount of helium produced was 7×10^{-5} ccs. Scaling this up to 1 year at 10^{14} fast flux, we obtain

$$\frac{8750}{140} \times 7 \times 10^{-5} \times \frac{10^{14}}{2410^{11}} = 2.2 \text{ ccs/gmBe/year}$$

It appears that the rate of helium production in beryllium used as a moderator will be in the range 1 – 4 ccs per gram at a flux of 10^{14} and will vary with the neutron spectrum, being greatest near the fuel.

Assuming a figure of 2.0 ccs/gm and considering a moderator operating temperature of 700°C, the volume of gas at this temperature per annum is

$$\frac{973}{273} \times 2.0 = 7.1 \text{ ccs/gm}$$

and expressing this in ccs. per cc of beryllium

$$= 7.1 \times 1.85 = 13.2 \text{ ccs/cc of Be.}$$

3. MECHANICAL EFFECTS

The effect of this helium on the dimensional stability and mechanical properties of beryllium is rather difficult to predict in the absence of diffusion data. It appears that experiments must be done on -

(a) the rate of diffusion of helium in beryllium and its variation with temperature.

(b) the effect of fast neutron flux irradiation on the properties of beryllium, including measurements of helium evolution during irradiation and subsequent dissolution and heat treatment.

However, a rough assessment of growth is possible by considering bubble formation and expansion as follows :-

$$\text{Let } x = \text{proportional growth} = \frac{\Delta V}{Vo}$$

where Vo = initial volume of an element (considered as a cube), containing one bubble volume ΔV

$$\Delta V = \frac{4}{3} \pi r^3 \text{ where V = radius of bubble}$$

Final volume of element considered

$$= Vo + \Delta V = (1 + \frac{1}{x}) \frac{4}{3} \pi r^3$$

Area of cross-section of cube $= \left[(1 + \frac{1}{x}) \frac{4}{3} \pi r^3 \right]^{\frac{2}{3}}$

Max. area of cross-section of bubble $= \pi r^2$

When the pressure in the bubble (p) is in equilibrium with the creep strength of the material (T) we have :-

$$T \left[(1 + \frac{1}{x}) \frac{4}{3} \pi r^3 \right]^{\frac{2}{3}} = p \pi r^2$$

Assuming a helium production of 2.0 cc/gm of Be per annum at NTP and considering a moderator operating temperature of 600 °C, the volume of gas at 1 atmosphere pressure would be

$$\frac{873}{273} \times 2.0 \times 1.85 = 12 \text{ cc per cc of Be.}$$

The pressure in each gas bubble at proportional growth x is given by -

$$12 \times \frac{14.7}{x} \text{ p.s.i.}$$

Thus $\quad T \left[(1 + \frac{1}{x}) \frac{4}{3} \pi r^3 \right]^{\frac{2}{3}} = 12 \times \frac{14.7}{x} \pi r^2$

Making the approximation that creep rate $= \frac{x}{3}$, this equation may be solved for known values of the creep strength by trial substitution.

The creep strength of beryllium is plotted in fig. 1; the published values may be extrapolated down to lower creep rates as shown in fig. 2. The results of calculation from the above equation using values from fig. 2 are plotted in fig. 3.

4. CONCLUSION

It will be seen that this calculation predicts a rise in the swelling due to irradiation from 0.1% per annum to 10% per annum over the temperature range 600-700°C.

5. REFERENCES

1. L.M. Wyatt, A.E.R.E. M/R. 1750 (O.U.O) "The behavior of Fissile Material under Irradiation at Elevated Temperatures."
2. Dalton, G.C.J. Private communication (1956).
3. Richmond, R. A.E.R.E. /NRDC. 79, 1955.

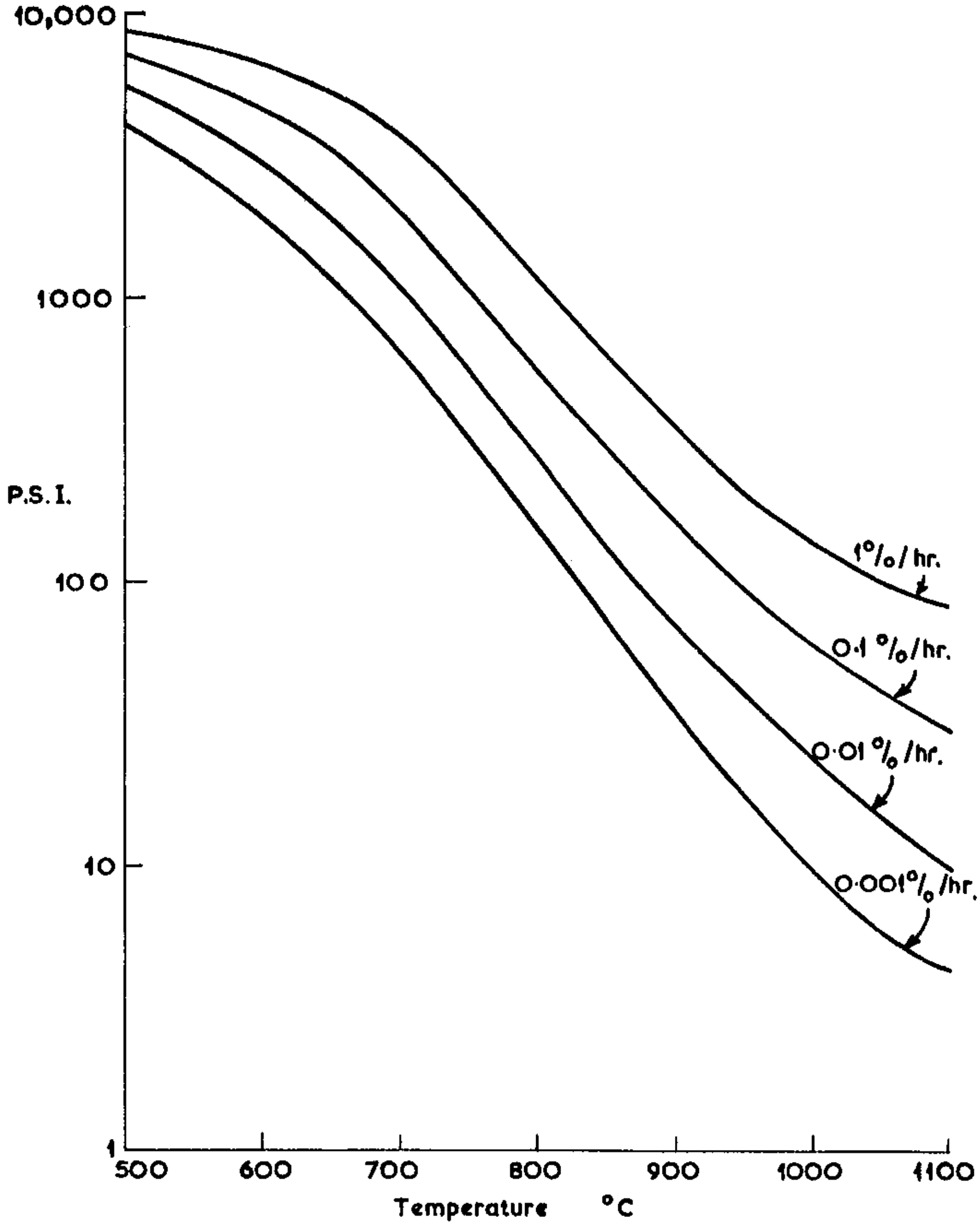

Fig. 4 Beryllium – Stress vs Temp. for arbitrary Creep Rates.

M.I.T. Report
CT 3804
D. Lillie

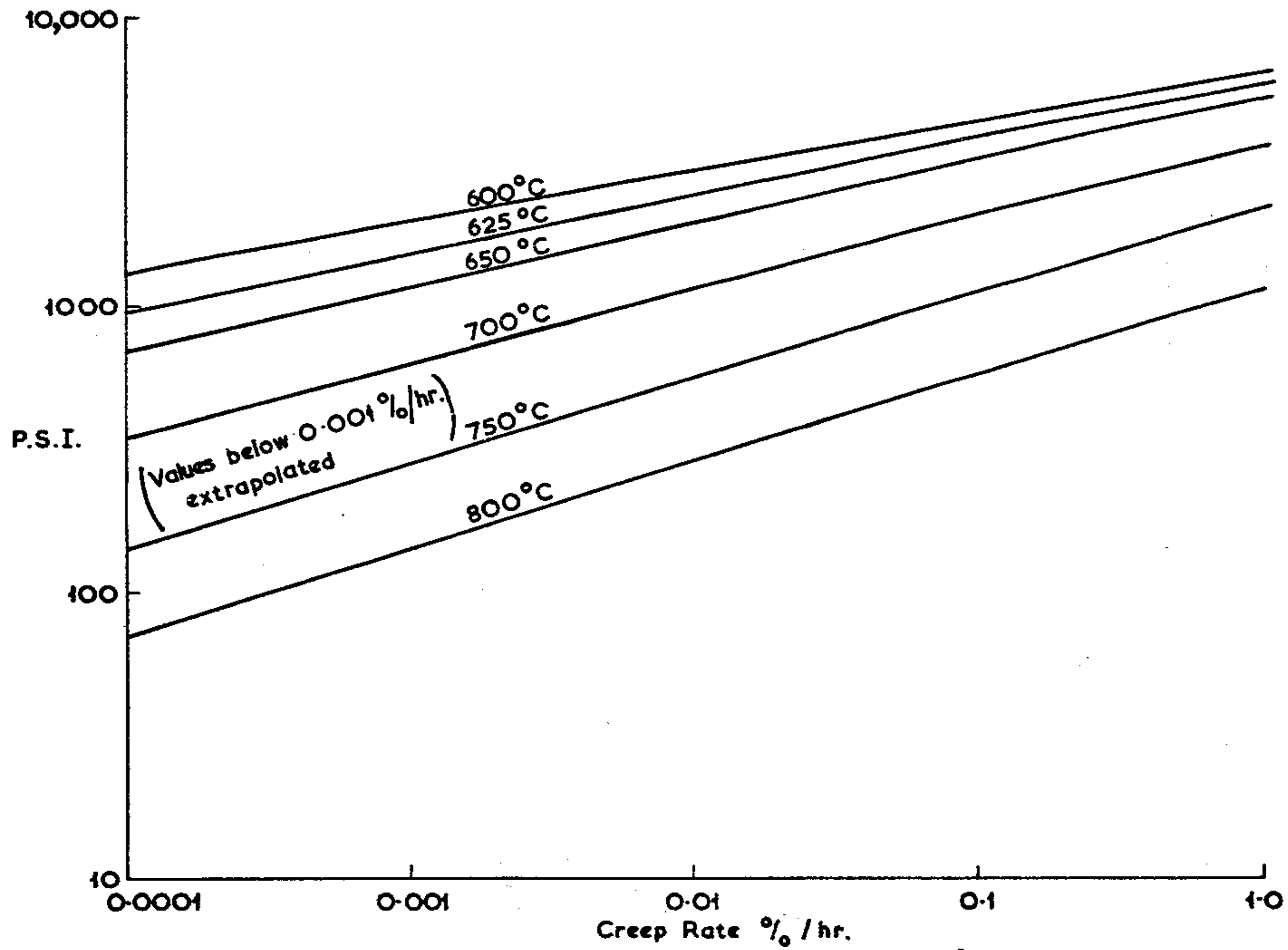

Fig. 2 Creep Rates of Beryllium vs Load (from Fig. 1)

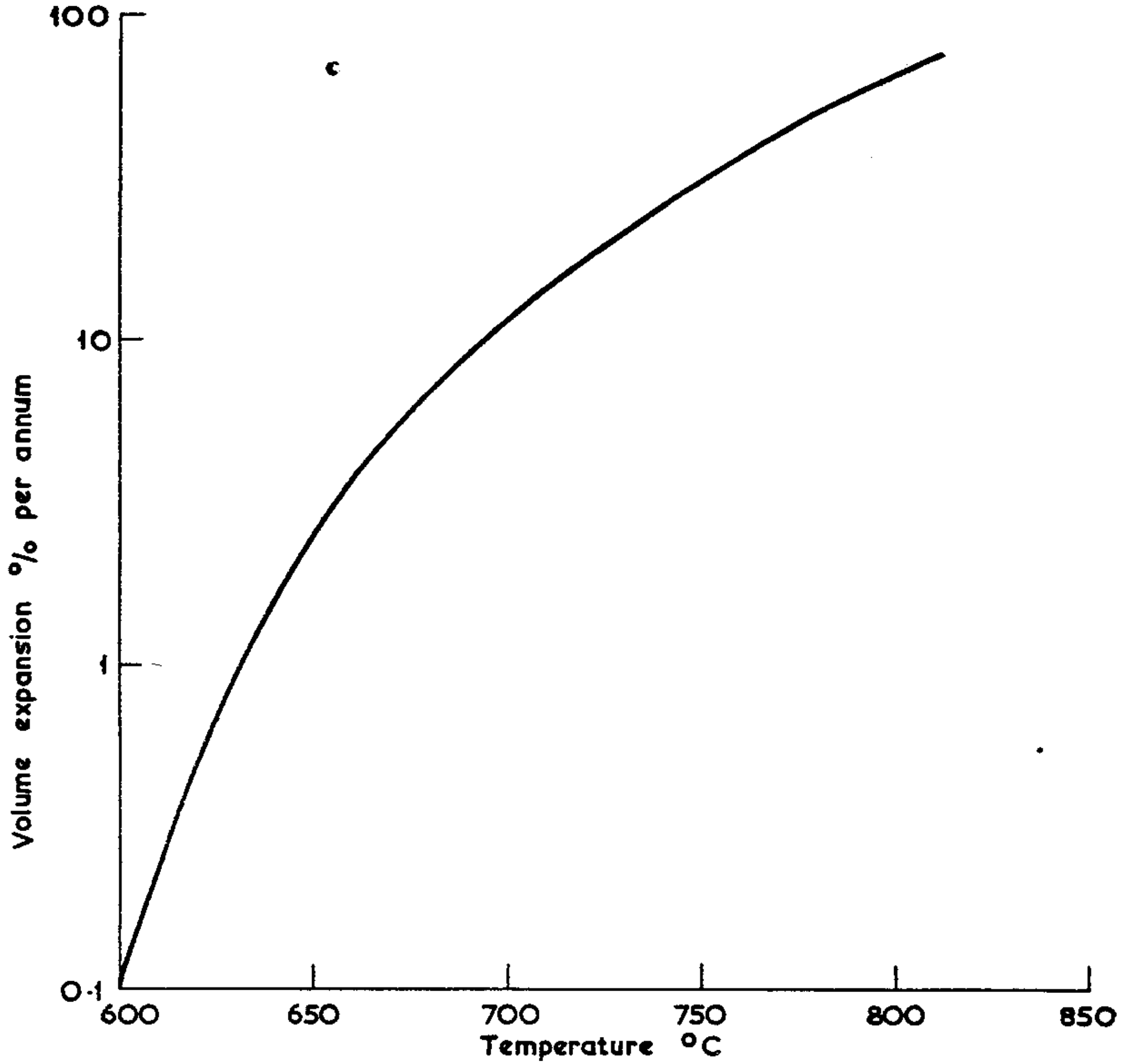

Fig.3 Growth of Beryllium at 10^{14} Flux