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EQUIVALENCE RELATIONS FOR RESONANCE ABSORPTION IN SMALL PARTICLES

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ABSTRACT

The effective resonance integral for fertile material agglomerated into small particles can be evaluated to a good approximation by using a reduced scattering cross section per fertile nucleus. The proportion of the moderator scattering per nucleus obtained from two extreme assumptions has been checked against the exact solution for spherical particles.

CONTENTS

1. INTRODUCTION

The possible use of dispersed fuel in a high temperature gas cooled reactor has aroused interest in the provision of simple techniques for the calculation of the self shielding of small particles of fertile material dispersed homogeneously throughout a moderating material. The particle size is a potential optimizing parameter which could help to improve the reactivity lifetime curve and for this reason will enter into survey calculations where the best combination of simplicity and accuracy in the method of computation is needed.

Lane, Nordheim, and Sampson (1962) have studied the resonance absorption in fertile material agglomerated into small particles. They have taken the particles as the fundamental elements in heterogeneous geometry and have proposed a simple equivalence relation which has been obtained by considering the Dancoff correction for the mutual shadowing of the particles.

A slightly different approach which still considers the individual particles as the fundamental elements, but proceeds more in the spirit of the theory of resonance absorption in heterogeneous systems developed by Gurevich and Pomeranchouk (1955), shows that the simple equivalence relation of Lane, Nordheim, and Sampson leads to an over-estimate of the self shielding. The basic point of departure is that instead of calculating a Dancoff correction we determine the effective cross section of the particles in the same way that Gurevich and Pomeranchouk evaluated the "sticking to the lump" probability.

2. GENERAL THEORY

Consider a homogeneous system of small particles of fertile material each with volume V, dispersed in a moderator with macroscopic scattering cross section Σ_m. If I is the effective resonance integral per fertile nucleus and N is the number of fertile nuclei per cm³ of the particle then NVI is the effective resonance integral per particle. The effective cross section of a particle is $NV\sigma_e$, where σ_e is the effective microscopic cross section of the fertile atoms in the particle. Denoting by V_m the volume of the moderator per particle and by σ_a and σ the absorption and total microscopic cross sections of the fertile nuclei respectively, then on the basis of the Narrow Resonance Approximation, we obtain the effective resonance integral of the particle as:

$$NVI = \Sigma_m V_m \int \frac{\sigma_a \, NV\sigma_e/\sigma}{NV\sigma_e + \Sigma_m V_m} \frac{dE}{E} .$$

Thus the effective resonance integral per fertile nucleus is:

$$I = \sigma_m \int \frac{\sigma_a \, \sigma_e/\sigma}{\sigma_e + \sigma_m} \frac{dE}{E}$$

$$= \sigma_m \int \frac{\sigma_a}{\sigma + \sigma_m \, \sigma/\sigma_e} \frac{dE}{E} , \qquad (1)$$

where σ_m is the scattering cross section of the moderator per fertile atom. In this formulation the mutual screening of the particles has been allowed for automatically and it only remains to determine the effective microscopic cross section of the fertile atoms.

Provided the particles are not too large the flux $\phi(u)$ at any lethargy u is essentially isotropic throughout the moderator so that the number of neutrons passing into a particle with surface area S is $S\phi(u)/4$. The probability of a neutron colliding with a fertile atom while traversing a path of length l through a particle is $1 - \exp(Nl\sigma)$. If f(l) dl is the fraction of paths through the particle of lengths between l and l + dl then the total reaction rate in a particle is:

$$NV \sigma_e \phi(u) = \frac{S}{4} \phi(u) \int (1 - e^{-N\sigma l}) \, f(l) \, dl \quad ,$$

where the integration is over all possible paths. Hence the effective microscopic cross section of the fertile nuclei is given by

$$\sigma_e = \frac{1}{N\bar{l}} \int (1 - e^{-Nl\sigma}) f(l)\, dl \quad , \qquad (2)$$

where $\bar{l}$ is the mean chord length of a fuel particle and equals 4V/S.

If the particles, the resonances, or the atom density of the fertile material in the particles are of such size that $N\bar{l}\sigma$ is small we can expand the exponential term under the integral sign in Equation 2 to obtain:

$$\sigma_e = \sigma - \tfrac{1}{2} N\sigma^2 \,\overline{l^2}/\bar{l} \quad ,$$

where $\overline{l^2}$ is the mean squared chord length, or to the same order of approximation,

$$\frac{\sigma}{\sigma_e} = 1 + \tfrac{1}{2} N\sigma \,\overline{l^2}/\bar{l} \quad .$$

When this expression is substituted into Equation 1 we are led to the result that

$$I = \sigma_p \int \frac{\sigma_a}{\sigma + \sigma_p} \frac{dE}{E} \quad , \qquad (3)$$

where

$$\sigma_p = \frac{\sigma_m}{1 + \tfrac{1}{2} N\sigma_m \,\overline{l^2}/\bar{l}} \quad . \qquad (4)$$

On the other hand if $N\bar{l}\sigma$ is large we must resort to the usual approximations introduced for the treatment of heterogeneous systems. Firstly we assume that Equation 2 is closely approximated by

$$\sigma_e = (1 - e^{-N\bar{l}\sigma})/N\bar{l} \quad ,$$

and then use the rational approximation to obtain:

$$\sigma_e = \frac{\sigma}{1 + N\bar{l}\sigma} \quad .$$

Substituting this value into Equation 1 again leads to Equation 3 as the effective resonance integral, but now:

$$\sigma_p = \frac{\sigma_m}{1 + N\bar{l}\sigma_m} \quad . \qquad (5)$$

The equivalence relations represented by Equations 4 and 5 represent a lower and an upper limit for the estimate of the self shielding effect of the particles. Lane, Nordheim, and Sampson (1962) derived the relation (5) from considerations of the Dancoff correction.

For large low energy resonances, such as the 1.054eV resonance of Pu240, where the self shielding effect of the particles is large it is not valid to use the Narrow Resonance Approximation to calculate the resonance absorption. It seems reasonable in these circumstances to follow the procedure initiated by Goldstein and Cohen (1962) and developed further by McKay (1964). The result obtained by McKay can be expressed as:

$$I = \mu \sigma_m \int \frac{\sigma_a}{\sigma + \lambda\sigma_m} \frac{dE}{E} \quad ,$$

where λ and μ are functions of, among other variables, the scattering properties of the moderating nuclei and depend to some extent on σ_m. Following through the previous analysis for particle self shielding we find that:

$$I = \mu \sigma_p \int \frac{\sigma_a}{\sigma + \lambda\sigma_p} \frac{dE}{E} , \qquad (3')$$

where in place of Equations 4 and 5 we obtain:

$$\sigma_p = \frac{\sigma_m}{1 + \frac{1}{2} N \lambda \sigma_m \overline{l^2}/\overline{l}} , \qquad (4')$$

and
$$\sigma_p = \frac{\sigma_m}{1 + N\overline{l}\lambda\sigma_m} . \qquad (5')$$

This is no longer a strict equivalence relation since the λ and μ in Equation 3' are dependent on σ_m and not σ_p. However it does lead to a simple evaluation of the effective resonance integral.

3. VALIDITY OF APPROXIMATIONS FOR SPHERICAL PARTICLES

Case, de Hoffman, and Placzek (1953) give the distribution of chord lengths in a sphere of diameter D as

$$f(l)\,dl = 2l\,dl/D . \qquad (6)$$

Substituting this expression into Equation 2 it is possible to perform the integration and obtain:

$$\sigma_e = \left[3\sigma/(N\sigma D)^3\right]\left[\tfrac{1}{2}(N\sigma D)^2 - 1 + (1 + N\sigma D)\, e^{-N\sigma D}\right] . \qquad (7)$$

Equation 1 with σ_e given by Equation 7 has been solved numerically for a selection of resonances and a variety of values of D and σ_m. The Doppler broadened contour function:

$$\psi(x,t) = \int_0^\infty e^{-p-p^2 t} \cos px \, dp ,$$

which enters into the calculation of the resonance cross sections, was evaluated for small x by the method used by McKay and Pollard (1963). This uses the differential equation:

$$4t^2 \frac{d^2\psi}{dx^2} + 4tx \frac{d\psi}{dx} + (2t + 1 + x^2)\psi = 1 ,$$

to reduce the Taylor series expansion of $\psi(x,t)$ to an expression containing only the function and its first derivative. The evaluation then proceeds step by step from $x = 0$, where $\psi(0,t)$ is known to be $\frac{1}{2}\sqrt{(\pi/t)}\exp(1/4t)\,\mathrm{erfc}(1/2\sqrt{t})$. For large x the function can be evaluated using one or two terms of the asymptotic expansion:

$$\psi(x,t) \simeq \sum_{n=0}^{\infty} \frac{t^n}{n!} \frac{d^{2n}}{dx^{2n}} \frac{1}{1+x^2} .$$

The mean chord length and mean square lengths of a sphere are easily obtained from the distribution (6) as $\overline{l} = 2D/3$ and $\overline{l^2} = \frac{1}{2}D^2$. Thus for spherical particles the equivalence relations (4) and (5) may be expressed as:

$$\sigma_p = \frac{\sigma_m}{1 + aND\sigma_m} , \qquad (8)$$

where $\frac{3}{8} \leqslant a \leqslant \frac{2}{3}$. The best value of a to obtain agreement with the exact evaluation for spherical particles lies somewhere between these two extremes. In order to make a comparison the effective resonance integrals for equivalent homogeneous systems taking $a = \frac{3}{8}, \frac{1}{2}, \frac{2}{3}$ have been calculated for the same selection of resonances and the same parameters as used in the exact solution.

4. NUMERICAL RESULTS

Figures 1 to 4 give curves for the exact and approximate effective resonance integrals for the 21.84 eV and 170.8 eV resonances of Th232 and the 6.88 eV and 190 eV resonances of U238, all at 300 °K, for σ_m = 1000 b, and 3000 b, and for particles of up to 1000 μ diameter. Figure 5 is a repetition of the calculation for the lowest resonance of Th232 for a temperature of 900 °K. Figures 6 and 7 give the same comparison for the 1.05 eV resonance of Pu240 for N = 3.37×10^{-4} with $\sigma_m = 1 \times 10^5$ and 2×10^5 and N = 4.33×10^{-3} with $\sigma_m = 1 \times 10^4$ and 1×10^5, in each case at a temperature of 300 °K. The values of N chosen for Pu240 correspond to plutonium containing 17% Pu240 in particles containing 1 plutonium atom to 10 thorium atoms, and in particles of pure plutonium. The large low energy resonances were chosen since they should show the greatest tendency towards the large value of a . In all these calculations the Narrow Resonance Approximation was used since the only object of the present study was to discriminate between the extreme values of a in the equivalence relation.

Figures 1, 2, 5, and 6 show that, for particles of mixed plutonium and thorium in the ratio of one to ten, the effective resonance integral is well represented by the small resonance approximation for particles up to 200 μ. The magnitude of the effective resonance integral for Pu240 is not correct as it was calculated using the Narrow Resonance Approximation instead of introducing the parameter λ in Equation 3'. However it can be seen from Equations 4' or 5' that since λ is fractional its neglect is equivalent to considering larger particles. This reinforces the deduction that, for our small concentration of Pu240, the small resonance approximation is valid.

Figures 3 and 7 show the resonance integrals for the 6.68 eV resonance of U238 and the 1.054 eV resonance of Pu240 in particles consisting entirely of natural uranium, or plutonium with 17% Pu240. Here the exact curve lies almost midway between the two extremes indicating that a value of a = $\frac{1}{2}$ should be used in the equivalence relation.

Figure 1 shows that, for the 21.84 eV resonance of Th232 at 300 °K, the large a approximation over-estimates by 57% the self shielding in a 200 μ diameter particle in a system with σ_m = 3000 b. Alternatively we can compensate an error in a by assuming that the results are for a different diameter. With this interpretation, Figure 1 shows that calculations, for supposedly 200 μ diameter particles using the large a approximation, will be correct for roughly 350 μ particles.

The resonance integral for the resolved resonances of Th232 with σ_m = 1011 b and at temperature T = 300 °K and 1500 °K have been obtained for comparison with the curves given by Lane, Nordheim, and Sampson (1962). Figure 8 gives the result of this calculation in the form I_D/I as a function of the particle diameter.

5. CONCLUDING REMARKS

When fuel particles are dispersed throughout a moderator it is to be expected that they will be randomly distributed both in size and shape about a mean. The distribution of size will have little effect on the calculation since I_D/I is approximately linear for small variations of particle size so that its mean value over the distribution will correspond to the value for the mean particle size. The variation in shape about some mean approximating a sphere implies that a large number of particles will be roughly spheroidal thus giving a greater value for $\bar{l^2}/\bar{l}$ than for a sphere with the same mean chord length. This would tend to increase the value of a in the equivalence relation but introduces the difficulty of interpreting the quoted size of the fuel particles. There seems to be no alternative but to disregard the shape of the particles at this stage.

The equivalence relation set up by the equation $\sigma_p = \sigma_m/(1 + aND\sigma_m)$ has been incorporated into the MULGA programme (Clancy et al., 1963) used to obtain multigroup averaged cross sections. In the studies at the A.A.E.C. Research Establishment it is proposed to investigate the possibility of dispersing particles of mixed plutonium and thorium oxides throughout a beryllia moderator. Since $N\sigma_m$ is the scattering cross section of the moderator per unit volume of the particle it will be the same for each nuclear species in the particle, so that apart from variations in a which are insignificant for the concentrations we are considering, the equivalence relation gives the same σ_p for all resonances whether they occur in Pu240 or Th232. It also follows that even though N varies with burn-up, σ_m varies like 1/N so that $N\sigma_m$ always remains equal to its initial value. In view of the random distribution of particle shapes and the obvious advantage of using a single formula for survey calculations, only one value of a is allowed in MULGA for all resonances and the input requires a single specification of $aND\sigma_m$.

The infinitely dilute resonance integral for fertile material lumped in a particle does not approach the homogeneous infinitely dilute value. As $\sigma_m \to \infty$, we have $\sigma_p \to 1/aND$ which shows that the maximum value of the effective resonance integral for dispersed fertile material is given by the homogeneous resonance integral with the scattering per fertile atom equal to 1/aND.

There is the possibility of self shielding of the thermal cross sections of the fissile nuclei, particularly the 0.3 eV resonance of Pu239. For 200 μ particles containing plutonium (83% Pu239) and thorium in the ratio of one to ten, the 2000 b peak of the Pu239 resonance would be reduced by about 3 per cent, so that the self shielding in the thermal groups would be less than this. We have not considered the change to be sufficiently significant to warrant further work at this stage.

6. ACKNOWLEDGMENT

The numerical results given above have been obtained from programmes written for the IBM1620 by Miss E. Kletzmayr.

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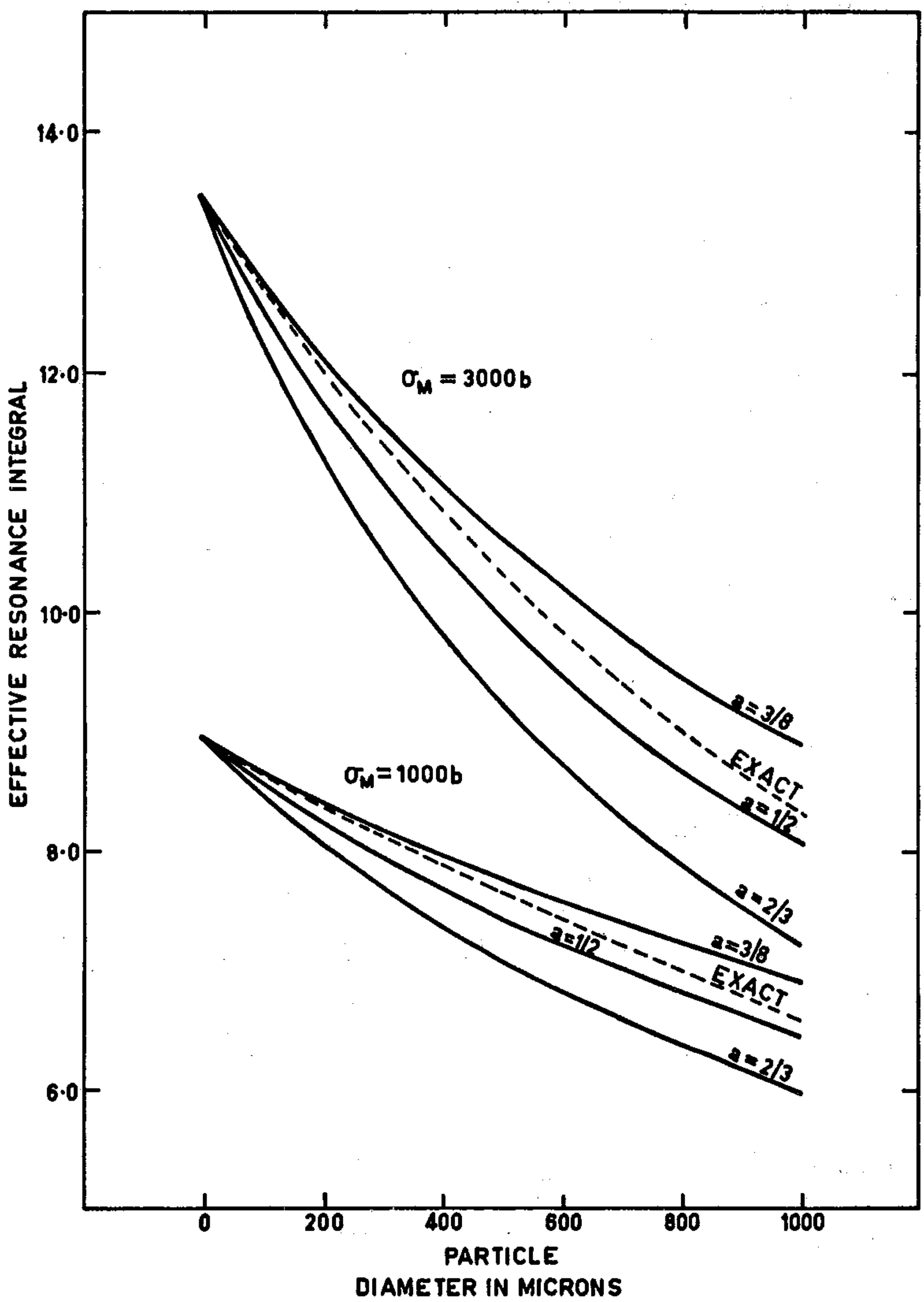

Figure 1. Th^{232} – 21.84 eV RESONANCE AT T = 300°K

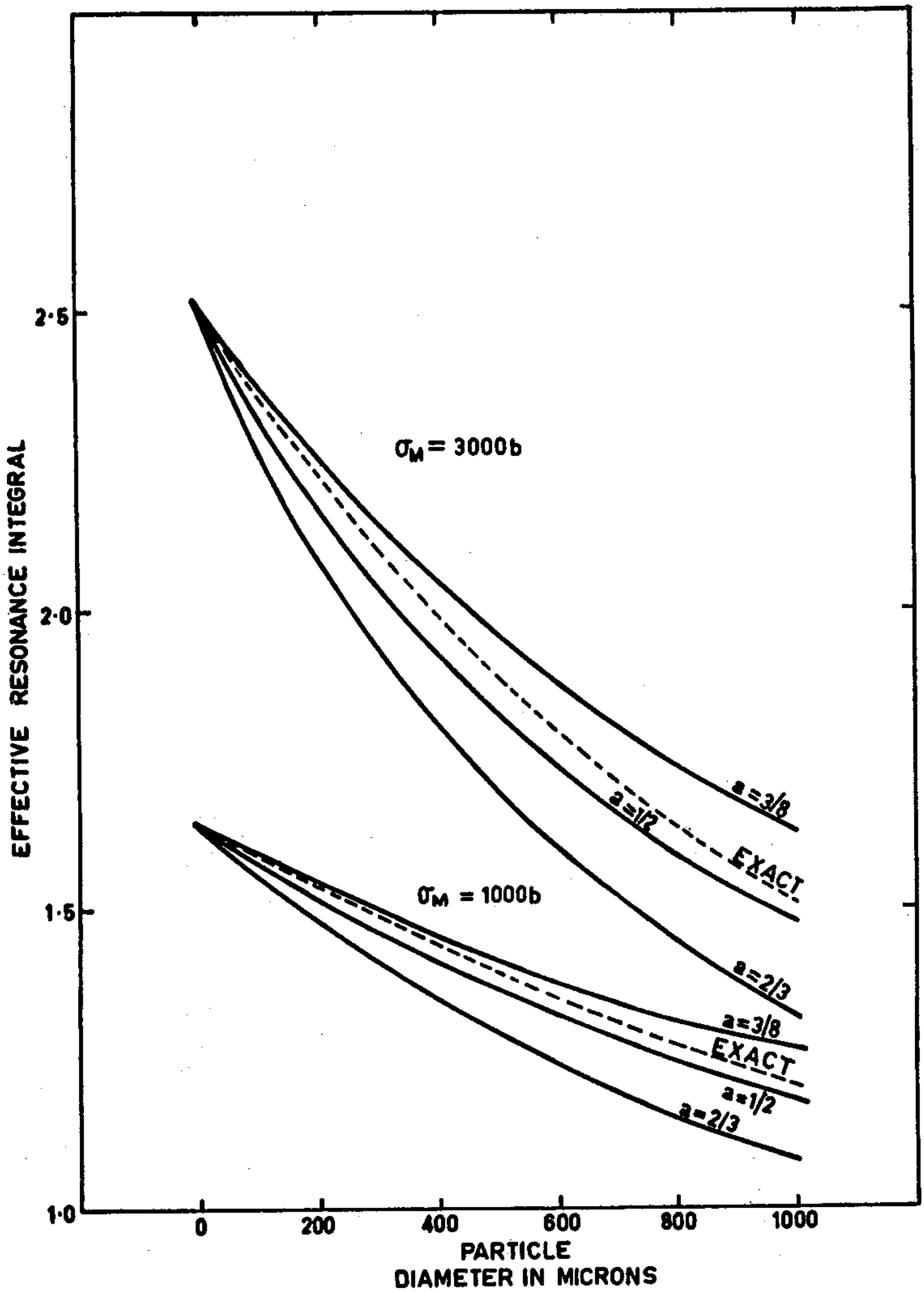

Figure 2 . Th^{232} —170.8 eV RESONANCE AT T = 300° K

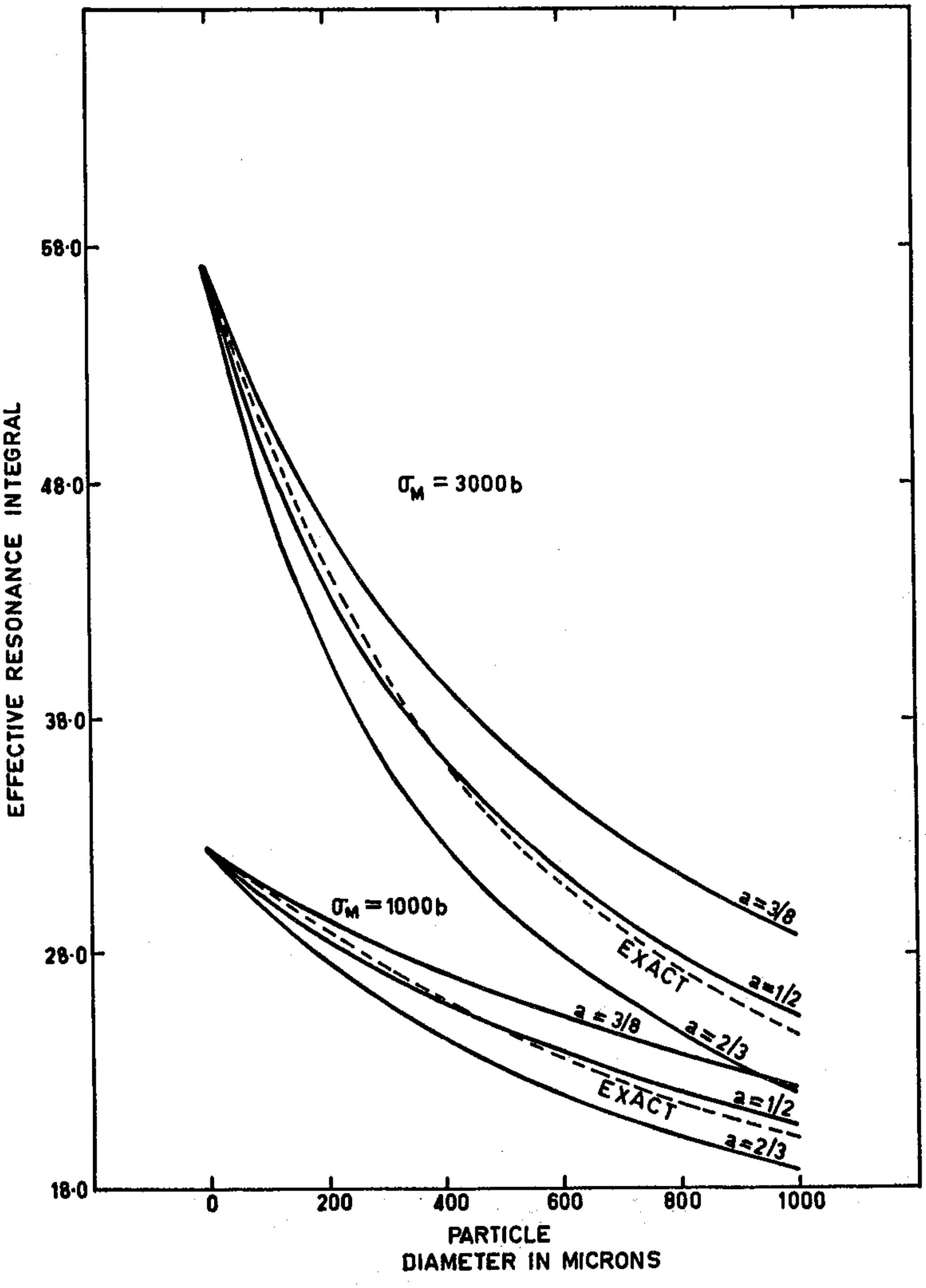

Figure 3. U^{238} — 6.68 eV RESONANCE AT T = 300° K

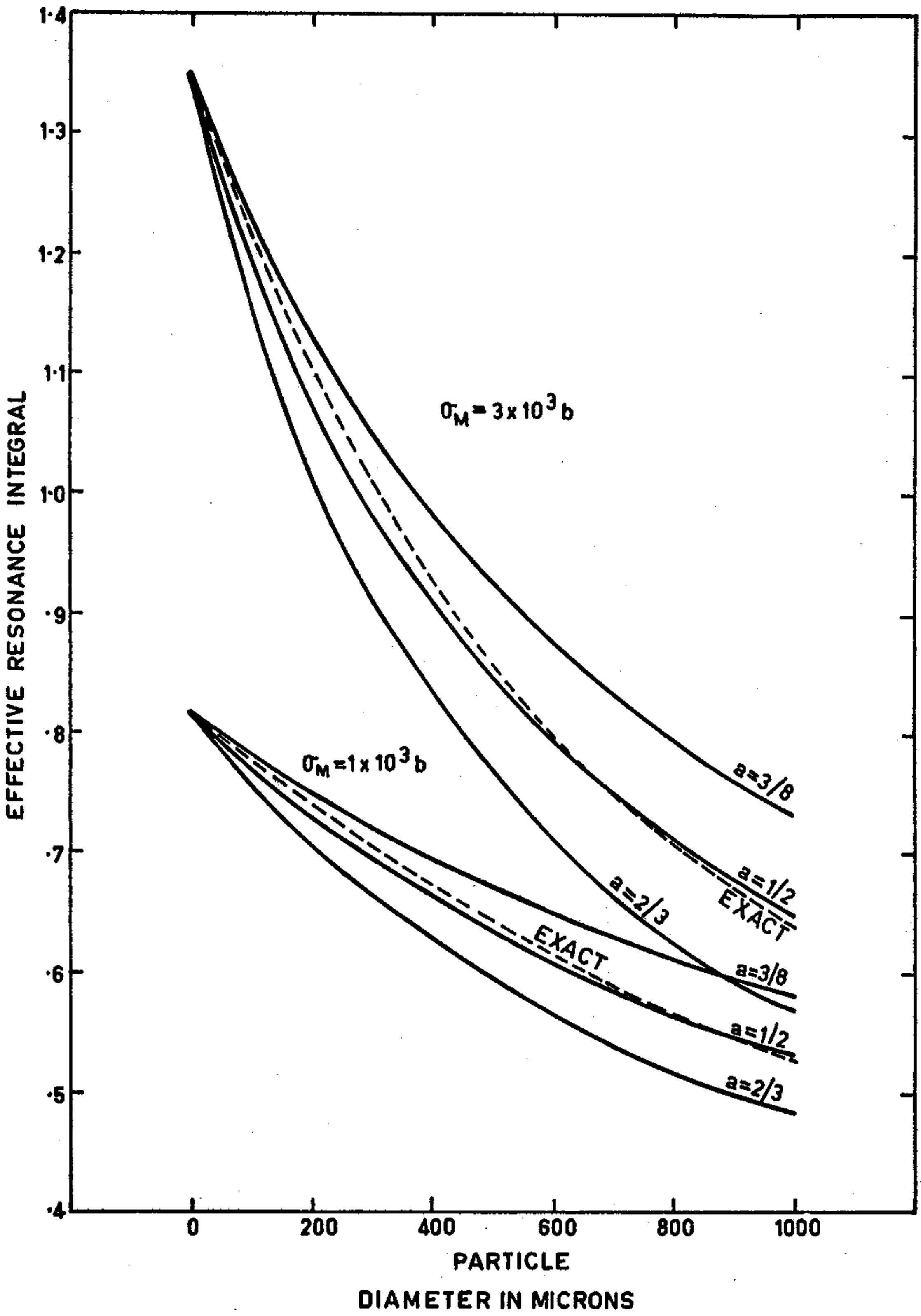

Figure 4. U^{238} — 190 eV RESONANCE AT T = 300°K

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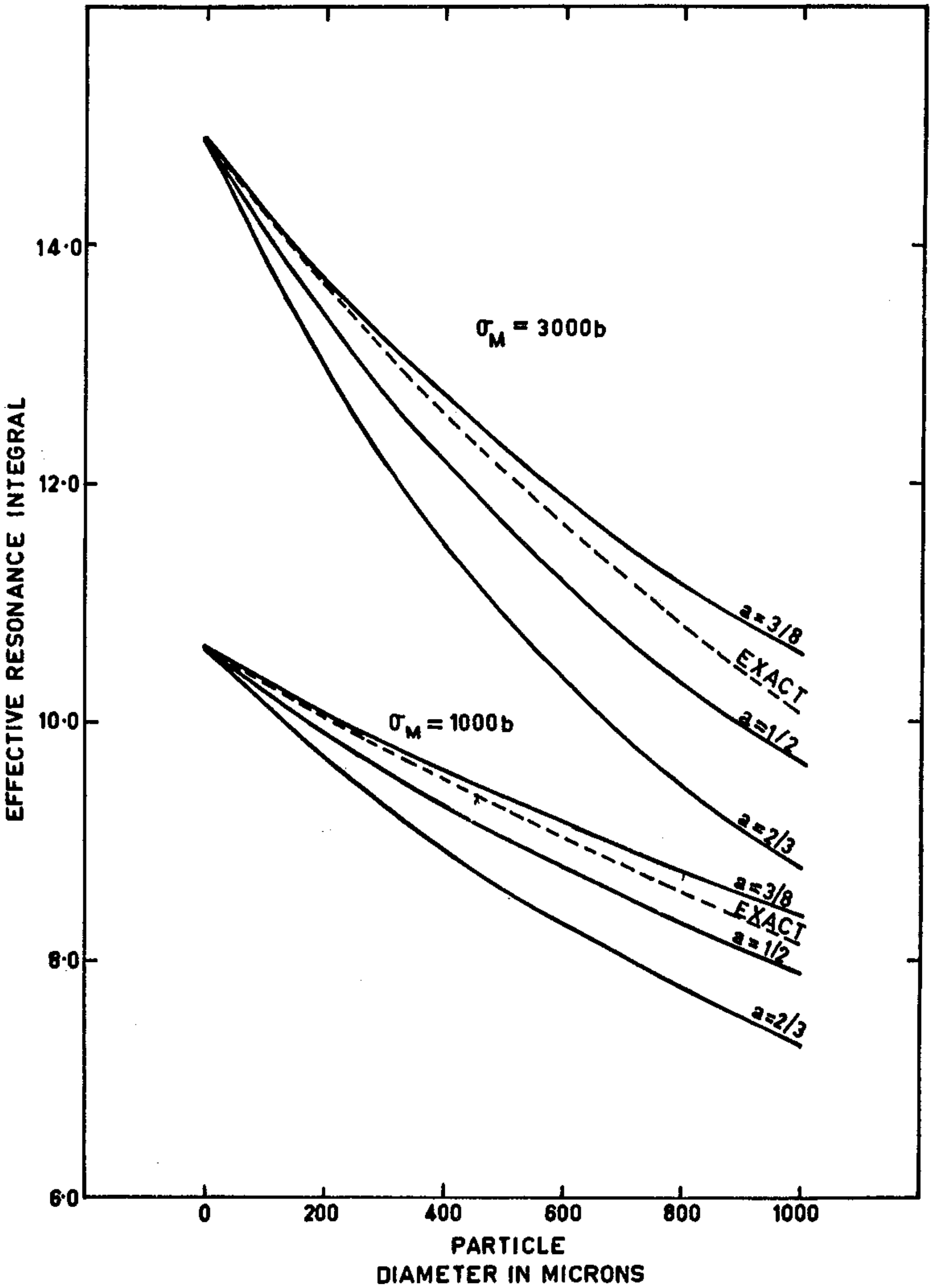

Figure 5. Th^{232} — 21.84 eV RESONANCE AT T = 900°K

P611

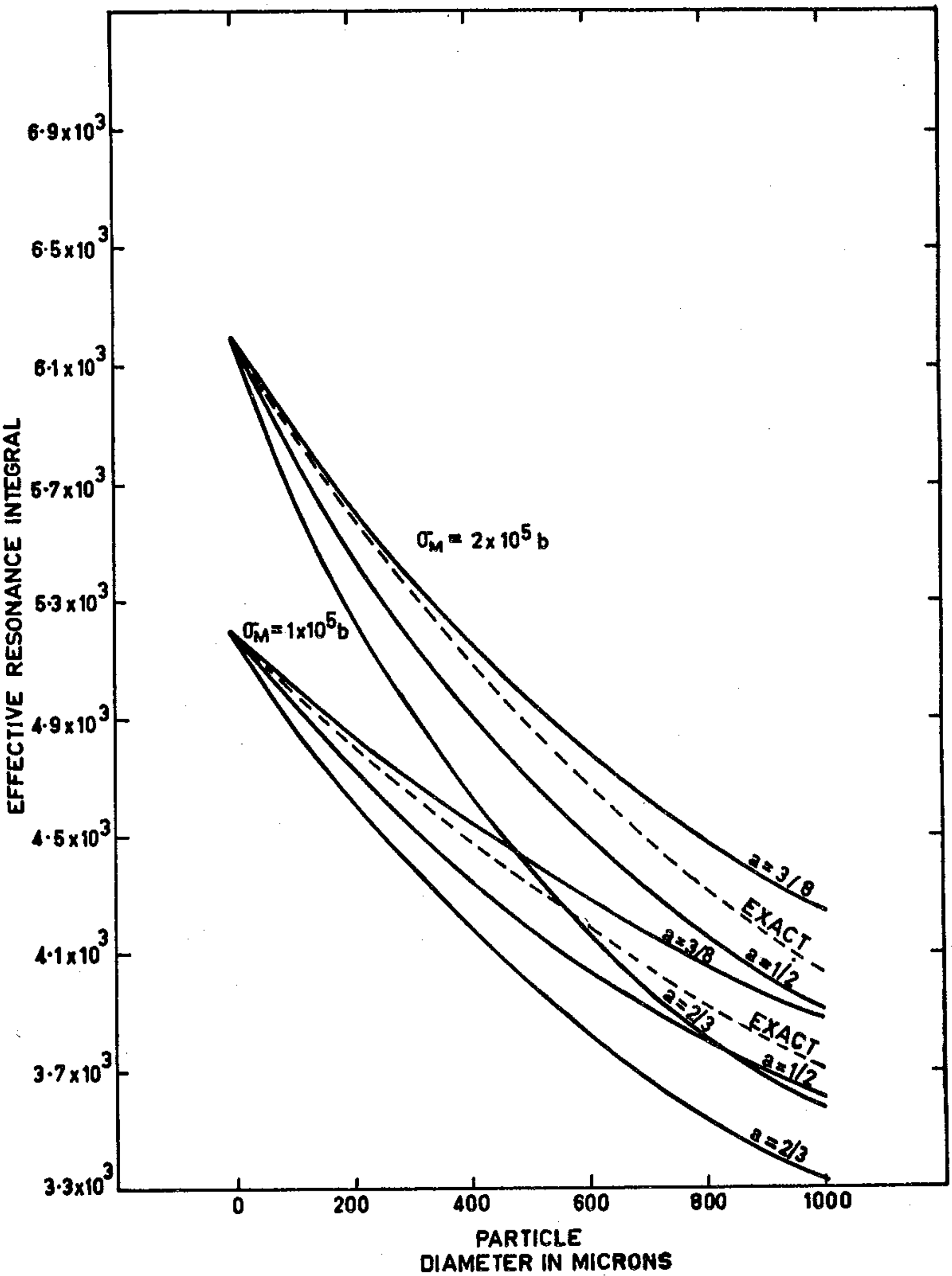

Figure 6. Pu^{240} — 1.054 eV RESONANCE AT T=300°K FOR PLUTONIUM/THORIUM PARTICLES (N=3·37 x 10^{-4}).

P611

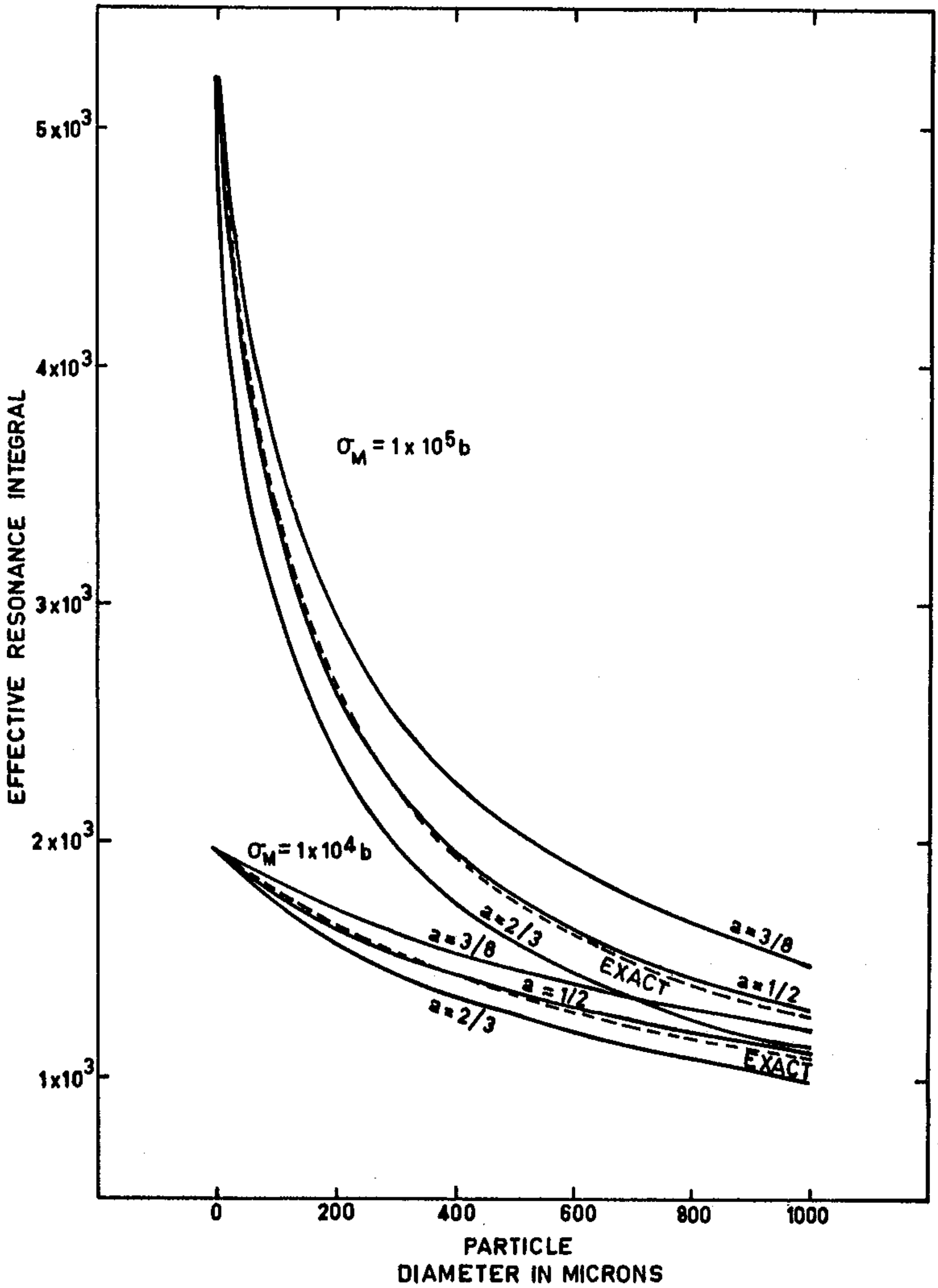

Figure 7. Pu^{240} — 1.054 eV RESONANCE AT T = 300°K FOR PLUTONIUM PARTICLES CONTAINING 17% Pu^{240} ($N = 4.33 \times 10^{-3}$)

P611

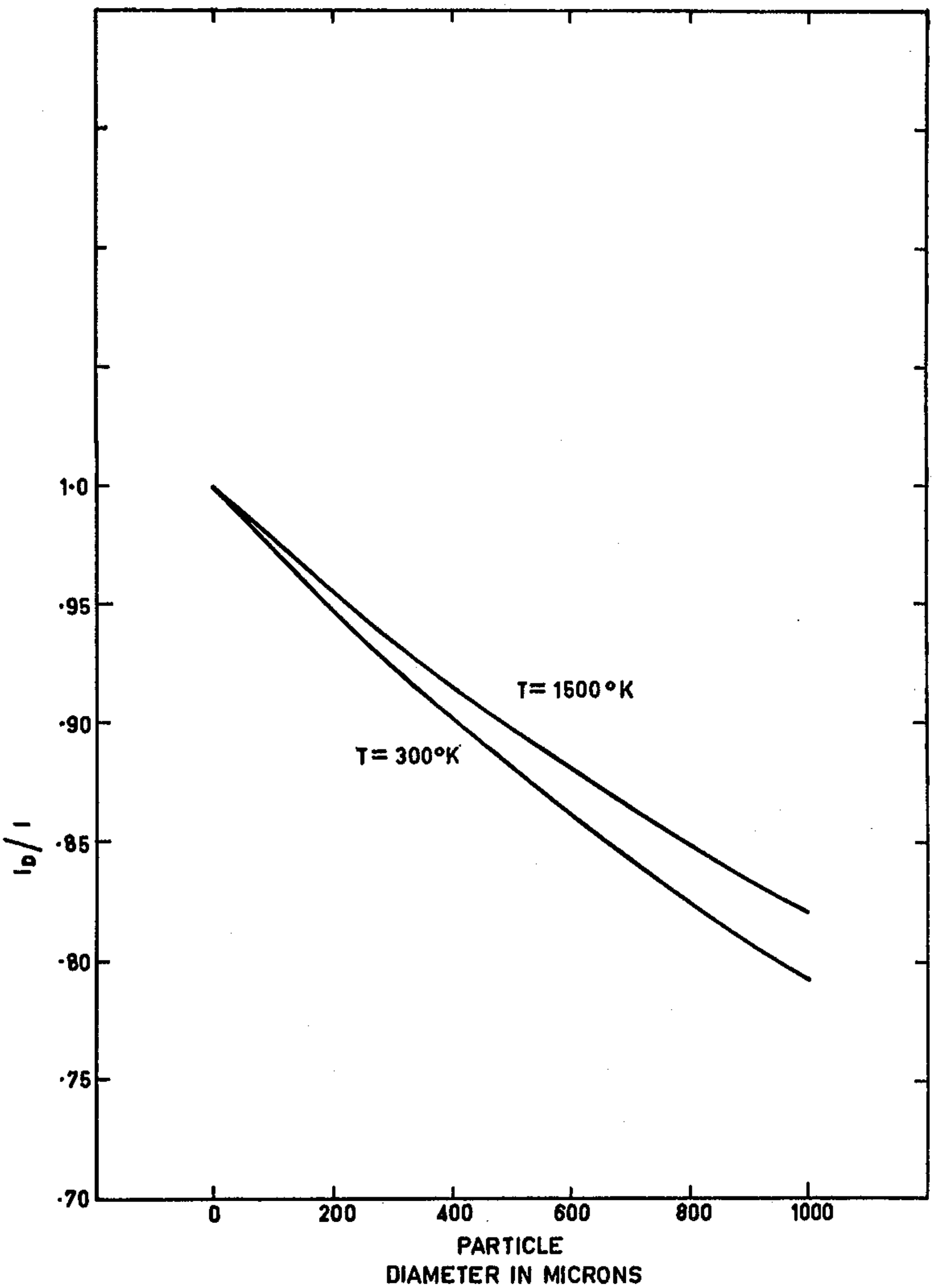

Figure 8. I_D/I FOR Th^{232} AT T= 300°K AND T = 1500°K WITH σ_M = 1011b AND a = 3/8

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