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ON THE EFFECTS OF NUCLEAR RADIATIONS ON SOLIDS

Sidney Siegel

AEC RESEARCH AND DEVELOPMENT REPORT

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PHYSICS DIVISION

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Sidney Siegel

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ON THE EFFECTS OF NUCLEAR RADIATIONS ON SOLIDS

Sidney Siegel

I Introduction

This paper is concerned with the effects of nuclear radiations on solids; these phenomena depend on the type of radiation and on the nature of the solid. In a nuclear reactor there are several types of radiations present, all having their ultimate origin from nuclear fission.

These radiations are:

1. γ rays, electromagnetic waves of energy of the order of several Mev.
2. Fast fission neutrons, the fission spectrum having a maximum near 1 Mev.
3. Slow neutrons, of thermal energy, resulting from the moderation of fission neutrons.
4. Fission fragments, heavy particles of mass near $A \approx 100$, of energy of the order 80 Mev each. These begin as highly charged ions, $Z^* \approx 25$. These fragments are not stable elements and soon decay radioactively, giving rise to other γ rays and to
5. Beta particles.

These radiations interact with the materials of the reactor. The energy of the incident radiations is transferred to the solids with which they interact, and is ultimately degraded to heat or potential energy. The effects produced by this process depends on the nature of the solid and on the conditions, such as temperature, stress, electric field, etc., existing in the solid during the absorption of radiation. The effects produced in most of the solid types, pure metals, alloys, semi-conductors, ionic crystals, ceramic mixtures, are studied. Some of these materials are "inert", some are fissionable.

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II Attenuation of Nuclear Radiations

A. γ Rays

The mechanisms by means of which the energy transfer from the nuclear radiations to the solid occur depend on the type of radiation. γ rays are attenuated through their interaction with the electric charges in matter, electrons in the atomic systems, and the positively charged nuclei. Three processes are important:

(a) Photoelectric effect, predominant for energies $\lesssim$ 50 kev. It is the process by means of which all γ rays are ultimately absorbed. An incident photon of energy E_γ is absorbed with the production of a fast electron of energy $E_\gamma - I$, where I is the ionization potential for that electron.

(b) Compton effect, predominant in the range .05 to 3 Mev. Here again fast electrons are produced by the scattering of the γ photon. The scattered γ's are further absorbed by the photoeffect.

(c) Pair production, increasingly important above 1 Mev. Here again fast electrons are produced.

All three processes result in the production of fast electrons and the ionization and excitation of the atoms or molecules of the solid. The electrons in turn produce further ionization and excitation, losing energy in this way until they ultimately fall into the electronic ground states of the solid.

B. Neutrons

Fast neutrons do not interact appreciably with the electronic structure of the atom, since they bear no charge. They may make elastic, inelastic or capture collisions with the nuclei of the solid. For most elements the elastic collisions are the most important. In such collisions a fast neutron

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gives up a portion of its kinetic energy to the struck atom in the solid, which is knocked-on with this energy, minus the binding energy of the atom to its lattice position. A neutron of fission energy E_o colliding elastically with an atom of atomic weight A imparts to it, on the average, the energy ΔE

$$\Delta E = E_o \frac{2A}{(A+1)^2}$$

$$\approx E_o \frac{2}{A} \text{ for } A \text{ large.}$$

In Table I below ΔE is tabulated for various atomic species; also given is the equivalent energy ϵ that an electron has when it moves with the same velocity as does the heavy particle of mass A and energy ΔE. It will be seen later that the rate of energy loss of such a heavy charged particle is similar to that of an electron having the same <u>charge</u> and <u>velocity</u>.

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Table I, for E_o = 2 Mev

Atom	Energy ΔE (kev) after coll.	ϵ (ev)
H^1	1000	500
Be^9	360	22
C^{12}	280	13
Al^{27}	140	5
Ag^{107}	40	0.2
U^{235}	17	0.04
F.F^{100}	of initial energy 94,000 kev	507
F.F^{140}	" 67,000 "	259

The last two items are for the light and heavy fragments in a typical fission. It is to be noted that except for H^1 and for these fragments, ϵ is only of the order 10 ev or less.

The probability of occurrence of elastic collisions of this type depend on the neutron flux and on the scattering cross-section σ_s. A typical value for σ_s is 5×10^{-24} cm^2; in a neutron flux of 10^{13} neutrons/cm^{2}sec, the probability of an atom being struck elastically by a fast neutron, and knocked-on with an energy ΔE, is therefore

$$5 \times 10^{-11} \text{ per second.}$$

In most substances other than hydrogen the fast neutron retains the major fraction of its incident energy after the collision, and goes on to make other elastic collisions. The recoil ion or atom moves through the

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lattice and loses its initial energy ΔE to it. The fast neutron ultimately transfers all its energy to such recoils, and reaches thermal equilibrium with the lattice. Its capture cross-section σ_a at such a low velocity is generally high, and it may be captured in one of the materials in the reactor, producing a radioisotope which soon decays with the emission of γ rays and β particles, fast electrons. The decaying radioisotope recoils with an energy of only 10 to 20 ev, because it is so heavy compared with the β particle, even though the latter may have an energy of 1 Mev. This recoil energy, as we shall estimate later, is probably insufficient to knock the decaying atom out of the lattice, but it can break molecular bonds and excite lattice vibrations.

If the neutron is captured in a fissionable element, say U^{235}, a fission may occur. Two highly ionized heavy fission fragments are produced, of energy near 100 Mev. These travel through the lattice as heavy charged particles and lose their energy by ionization, excitation, and elastic collisions with the atoms they encounter. Eventually they are stopped and remain in the solid as two foreign atoms.

C. Charged Particles

Evidently the attenuation of any of the radiations present involves the stopping, by the solid, of energetic charged particles -- electrons, recoil ions, or fission fragments -- or neutral recoil atoms produced by lower energy encounters with neutrons or recoil ions. The energy loss of charged particles occurs as a result of inelastic encounters in which they excite and ionize the atoms of the stopping medium. The manner in which an electron and a heavy ion lose energy by this process is nearly the same for each, depending on the

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charge and velocity of the particle. The principal difference in the stopping process for the two types of particles lies in the fact that the electron is too light to transfer appreciable momentum to an atom which it strikes, and so does not lose much energy by elastic collisions. This process is important for heavy particles, however. The energy loss can therefore be considered in two parts, (1) excitation and ionization common to both heavy and light charged particles, and (2) elastic collisions which hold only for heavy ions and atoms.

The rate of energy loss for charged particles is well known:

$$-\frac{dE}{dx} = 4\pi N_o \frac{z^2 e^4}{mv^2} Z \log \frac{mv^2}{\bar{E}} .$$

This is the rate of decrease of energy of a particle of charge z, velocity v, in a medium having N_o atoms per cc, each atom having Z effective electrons of excitation energy $\bar{E}$. The mass of the moving particle does not appear here, only the electronic mass m. For fast electrons $\bar{E}$ will include the energy of all electrons and Z will be the atomic number of the stopping element. For heavy recoil ions, $\epsilon = \frac{1}{2} mv^2$ is only of the order 10 ev, so Z will be 1 and $\bar{E}$ the excitation potential of the valence electrons. For recoils of even lower energy ϵ, it is likely that the recoil will not be charged at all, in which case the expression above is not valid. The equation above is for stopping by electrons bound with energy $\bar{E}$. In a metal, with free electrons in a band of allowed levels filled up to an energy E_p, the expression is nearly the same, with $\bar{E}$ replaced by E_p. Typical values of E_p are given in Table II below:

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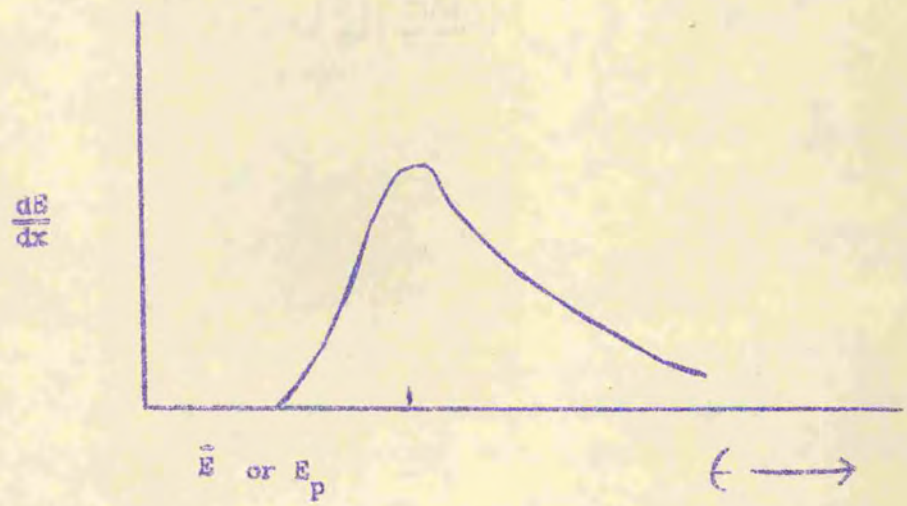
Table II

Metal	E_p (ev)
Li	4.7
Be	14
Al	12
Cu	5.9

Evidently these are of the same order as $\bar{E}$ for valence electrons. The energy loss by fast electrons or ions, for which $\epsilon \gg \bar{E}$ is thus of the form,



becoming zero when $\epsilon \approx \bar{E}$ or E_p because of the presence of the $\log$ term. For electrons of energy less than $\bar{E}$, capture into stable levels

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occurs and the process of stopping is over. For heavy particles, when $\epsilon < \bar{E}$, the ion becomes a neutral atom by capturing an electron, but still has considerable kinetic energy, of the order 100 kev.

D. Energy Loss by Atoms[1]

The neutral atom continues to lose its energy by elastic and inelastic encounters with other atoms of the solid. The energy loss by elastic encounters is of the form

$$-\left.\frac{dE}{dx}\right)_c = 2\pi N_0 \frac{z^4 e^4}{Mv^2} \log E/E^*$$

with $E^* = 35\, Z^{2/3} \frac{m}{M}$ ev ≈ 0.01 ev.

Typical values of E^*, which came from the Fermi-Thomas atom model, are: Be, .005 ev; Al, .004 ev; Ag, .005 ev. Some of these elastic collisions will transfer enough energy to the struck atom to displace it from the lattice. If it is assumed that to displace an atom requires an energy of at least $E_d = 25$ ev (about twice the heat of sublimation for typical materials), the rate of energy loss in such collisions is

$$-\left.\frac{dE}{dx}\right)_{c,d} = 2\pi N_0 \frac{z^4 e^4}{Mv^2} \log E/E_d.$$

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The ratio of energy loss in all elastic collisions to that in collisions in which an atom is displaced is then

$$\frac{-\,dE/dx)_c}{-\,dE/dx)_{c,d}} = \frac{\log E/E^*}{\log E/E_d} \approx 2.$$

Hence, about half the energy dissipated in elastic encounters displaces atoms, about half merely excites lattice vibrations.

Of these collisions which transfer enough energy to displace an atom, i.e., an amount greater than E_d, the average energy transferred is

$$E_{av} = E_d \log E_o/E_d \approx 175 \text{ ev.},$$

for $E_o = 100$ kev.

The energy loss by the moving atom through inelastic encounters with the atoms of the solid occurs by electronic excitation of these atoms. The result differs depending on whether the solid is a metal or an insulator. For a metal the energy loss expression depends on ϵ through expressions of the form

$$\left.\frac{dE}{dx}\right)_e = 12\,\pi\, N_o\, n\, \epsilon\, (A_o) \quad \text{when } v < u_o,$$

where A_o is a constant involving fundamental physical constant and the atomic number $Z^{2/3}$ only, n is the number of free electrons per atom;

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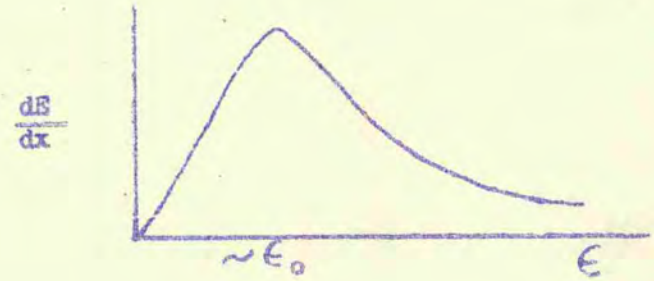
and $-\left.\frac{dE}{dx}\right)_e = \frac{\pi Z^2 e^4 N_0 n}{\epsilon} \log\left(\epsilon/\epsilon_0 + 1.08\right)$ when $v > u_0$,

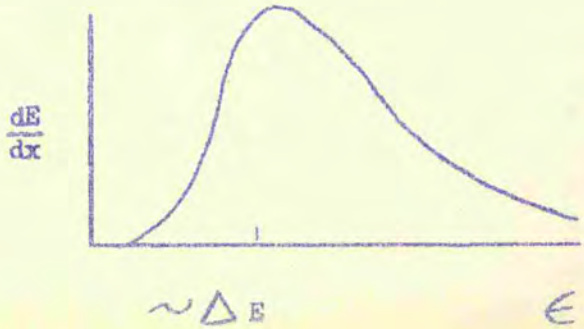
where ϵ_0 and u_0 are the energy and velocity of an electron at the top of the filled band in the metal. Evidently the form of the energy loss curve is



rising linearly at the beginning and then decreasing as $1/\epsilon$, with a maximum near ϵ_0.

For an insulator with a finite energy gap of width ΔE between allowed energy level bands, the curve is of the same form as that given above for energies ϵ substantially greater than ΔE, has a maximum near $\epsilon \approx \Delta E$, and falls exponentially with decreasing ϵ.

This energy loss curve is of the form



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In an insulator the energy loss by excitation falls off very rapidly when $\epsilon < \Delta E/8$, while in a metal the energy loss falls off only linearly when $\epsilon < E_o$. For typical insulators $\Delta E \approx 10$ ev, for typical metals $E_o \approx 5$ ev.

As an idea of order of magnitude, it is instructive to get the ratio of energy dissipated elastically to that dissipated in excitation, for a 140 kev Al recoil atom. This ratio is

$$\frac{-dE/dx)_c}{-dE/dx)_e} = \frac{Z^2}{n} \frac{m}{M} \frac{\log E/E^*}{\log (\epsilon/\epsilon_o + 1.08)} \approx 4\% .$$

Thus about 6000 ev of the original 140 kev are used up in elastic collisions, of which half produce only lattice vibrations, the other half of the collisions transfer enough energy to displace atoms from the lattice.

This picture of the overall energy loss of the recoils may be somewhat simplified if one assumes that in the initial part of the range the energy loss occurs by electronic excitation only, in the latter part, when the recoil energy has fallen below a critical value $E_t = \frac{M}{m} \epsilon_t$, the energy loss is entirely by elastic encounters. For the lighter metals ϵ_t is determined from the condition

$$-\frac{dE}{dx}_c \Big/ -\frac{dE}{dx_e} = 1 = \frac{Z^{10/3}}{3n} \frac{m}{M} \frac{E_r^2}{\epsilon_t^2} \frac{\log E/E^*}{76} ,$$

where E_r is the Rydberg energy, 13.6 ev, while for light insulators ϵ_t is determined by the condition $\epsilon_t = \Delta E/8$. In the table below values

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of ϵ_t and E_t are given for several materials, based on these assumptions.

Table III

Material	ϵ_t	E_t	N_d
Be	0.14 ev	2.3 kev	15
BeO	1.25	20	130
Graphite	0.5	10	70
Al	0.4	18	120

The last column N_d, is the total number of displaced atoms per initial recoil, estimated by assuming that one half of E_t is used up in exciting lattice vibrations, the other half in displacing atoms from the lattice. On the average the energy of a displaced atom is about 150 ev, and each such displaced atom has enough energy to in turn displace a secondary, so $N_d = \frac{E_t}{150}$ yields the value of N_d given in the table.

The critical energy ϵ_t increases as we go toward heavier metals, so a larger fraction of the original recoil energy will appear in elastic encounters. Comparing values of ϵ_t for two materials of nearly the same atomic weight but differing in that one, Be, is a metal, and the other, BeO, an insulator it is evident that ϵ_t differs by a factor of nearly 10. This may indicate that N_d should be significantly larger in an insulator than in a good conductor.

We noted earlier that for $\sigma_s = 5$ barns, $nv = 10^{13}$, the probability that an atom is struck, and displaced, by a fast neutron is 5×10^{-11}.

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In aluminum exposed in such a flux for 100 days the total fraction of displaced atoms will be

$$\left(\frac{\sum N_d}{N_o}\right) = \sigma_s \, (nv) \, N_d \, t$$

$$= 5\%.$$

Not all of these displaced atoms will remain stuck in interstitial positions. Many will return to normal lattice sites through the ordinary mechanisms of diffusion in solids. This point will be examined later.

Of the energy E_t which the recoil atom dissipates in elastic encounters, about 1/2 appears as lattice vibrational energy, i.e., merely raises the temperature of the solid. Depending on the number of atoms among which this energy is initially shared, the temperature may rise locally to high values. A naive consideration of the distribution of the lattice vibration energy indicates that it is shared among the atoms in a tunnel about 500 atoms long, 300 atoms in sectional area, about 10^5 atoms in all. For Al the energy to be shared is about 10^4 ev, i.e., about 0.1 ev per atom. This indicates that these small regions of the material are locally heated to about 1200°K for short instants of time. Since the probability of making a primary Al recoil, in a flux of 10^{13}, is about 5×10^{-11} per second, it appears that perhaps 10^{-5} of the material is locally heated in this way during any second. During 100 days at this flux, every portion of the material will have been heated to this temperature about 50 times, for times of very short duration.

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E. Fission Fragments

The fission fragments start out as heavy, highly ionized particles with energy of the order 80 Mev, $\epsilon \approx 500$ ev. The same type of energy loss expressions as were used previously are valid here, modified to take account of the fact that the moving particle has different mass from the atoms of the stopping material, and has a high effective charge during the initial part of its range.

About 2.4 Mev of the fission fragment's initial energy is dissipated in elastic collisions, and of this about 1/2 goes into displacing atoms, about 1/2 in lattice vibrations. The average knocked-on atom has an energy of about 375 ev if the fissionable atoms are distributed in a heavy metal such as U. Each 375 ev primary recoil can produce about 3 others. Thus the total number of displaced atoms per fission fragment is

$$N_d = 4 \times \frac{2.4 \times 10^6 \text{ ev}}{375 \text{ ev}} \times 1/2 = 13{,}000 \text{ displaced atoms.}$$

In a flux of 10^{13} for 100 days, each atom in a sample of natural U metal will have been displaced

$$\left(\frac{\sum N_d}{N}\right) = 2\, N_d\, \sigma_f\, (nv)t \times .007$$

$$= 9 \text{ times.}$$

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If the fissionable material is dispersed in a lighter element, for example an Al-U alloy, N_d is somewhat less, perhaps 5000 rather than 13,000 as in U metal.

The lattice vibrational energy excited by fission fragments is of the order 1.2 Mev. Their range is 5×10^{-4} cm, about 10^4 atoms long. Assuming 1000 atoms in the cross-sectional area of the "fission track tunnel", the average vibrational energy per atom is about 0.12 ev, about 1500°K. From simple heat diffusion estimates, this fission tract tunnel remains hot for perhaps 10^{-10} second. Since, however, the period of a lattice vibration is 10^{-13} seconds, this is not an inconsiderable time on an atomic scale.

In natural U in a flux of 10^{13}, the probability that an atom will fission is 4×10^{-11} per second. Since each fragment heats 10^7 atoms, during any one second a total fraction of 4×10^{-4} of the material has been heated to 1500°K for short instants. During a period of 100 days, each portion of the metal has been so heated about 3300 times.

Equilibrium Concentration of Displaced Atoms (2)

In the previous sections estimates have been made of N_d, the number of atoms displaced by fast recoils or by fission fragments. Many of these soon return to neighboring lattice vacancies by diffusion. The equilibrium number of displaced atoms is determined by balancing the rate of their production against their rate of disappearance.

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The rate of production per cc of material is

$$R_p = N_o N_d (nv) \sigma_s \quad .$$

The rate of return of displaced atoms is determined by the rate at which lattice vacancies move up to the interstitial atoms. If there are n displaced atoms, and vacancies, per cc, each atom has Z neighbors, and each atom has a probability p per second of hopping to a neighboring vacancy, the probability that a vacancy will be filled is pZn. The probability that it will be filled by a displaced atom and the latter disappear is

$$R_d = \frac{n}{N_o} pZn.$$

In equilibrium

$$\frac{n}{N_o} = (N_d (nv) \sigma_s / pZ)^{\frac{1}{2}} \quad .$$

The rate of jumping p can be estimated from data on diffusion in solids. It is temperature dependent in the manner

$$p = 10^{13} e^{-Q/RT} ,$$

10^{13} being the lattice vibrational frequency, Q the activation energy for the motion of a vacancy. For a value of $Q = 20{,}000$ cal/mole at $T = 300^{\circ}K$

$$p = 10^{13} e^{-33} = 10^{-1.4} = .04 \quad .$$

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Hence, for Al in a flux of 10^{13} at room temperature

$$\frac{n}{N_o} = 1.2 \times 10^{-4} ,$$

i.e., the equilibrium number of displaced atoms is quite small. It is important to note that n/N_o is extremely sensitive to the activation energy Q or to the temperature, through the term $\exp(-Q/RT)$. Hence, in materials of high activation energy the equilibrium number of displaced atoms may be much greater, and conversely operation at high temperatures should markedly reduce the number of stuck displaced atoms. This estimate has neglected the lattice vacancies already existent in the material, so that the rate of disappearance of displaced atoms is even greater than that here estimated, the equilibrium number smaller.

III Effect of Pile Radiations on Materials

In the last section it was estimated that in simple pure metals like Al there are only a few permanently displaced atoms at room temperature, $n/N_o \approx 10^{-4}$. However, in semi-conductors and ionic crystals many of the electrical and optical properties are extremely sensitive to impurity and vacancy concentrations even smaller than this. Furthermore, even in metals there are many non-equilibrium situations where an atom once displaced has little likelihood of returning to an equivalent site. In cases where internal or external stresses are present, where non-equilibrium phases are present, the effects of the local temperature spikes may be important. In fissionable materials, the rate of production of displaced atoms is higher,

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and two foreign atoms per fission remain stuck in the material. It is apparent, therefore, that many properties of various types of solids may change in significant ways. In this section the experimental evidence for these changes will be reviewed.

A. Inert Metals

(1) Aluminum. The aluminum water tubing in the Hanford piles is subject to the most intense neutron radiation available. Sections of these tubes have been removed and conventional physical properties examined; no significant changes have been observed. More sensitive experiments[3] on single crystals of aluminum have been made, after exposures in hollow uranium slugs in the Oak Ridge pile of 2×10^{18} neutrons/cm^2. The damping capacity of single crystals was measured before and after exposure. Normally the damping in single metal crystals is extremely sensitive to small amounts of cold work or impurity. No significant changes were observed, on samples measured within 30 minutes after removal from the pile. No changes in density, measured with a sensitivity $\frac{\Delta\rho}{\rho}$ of 5×10^{-5}, were observed.

(2) Be Metal.[4] Samples of Be metal prepared by extrusion were irradiated for a total nvt of 5×10^{19} neutrons/cm^2 after the electrical resistance, density, elastic modulus and damping, and hardness had been measured. No significant changes in any of these properties were observed. The sensitivity on density is 5×10^{-5}, on electrical resistance 10^{-3}.

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(3) Copper-Gold Alloy.[5] The alloy studied has the composition Cu_3Au, and can exist as an ordered, or a disordered, solid solution. The crystal structure is face centered cubic; in the ordered state Cu atoms are located at face centers, gold atoms at cube corners of the unit cell; in the disordered structure each lattice site is occupied by either Cu or Au, with a 75% probability that it is Cu, 25% that it is Au. The ordered state is obtained by slow cooling from 400°C, the disordered state by quenching from 500°C. Numerous properties depend on the degree of order, in particular, the electrical resistivity, which has the value 4.6×10^{-6} w cm in the ordered, 11.5×10^{-6} w cm in the disordered state at room temperature.

In this material atomic displacements followed by subsequent return of the displaced atom to another, but not equivalent, lattice site can be detected because of the change in order, even if at the time of measurement all atoms are again on lattice sites. Also, atomic rearrangement among the sites of the f.c.c. structure during the postulated high temperature spikes near a recoil can also occur with accompanying changes in order.

Samples of this alloy were prepared in the ordered and disordered states and exposed for various durations in a reactor. The data for the electrical resistance of the two types of samples before and after exposure are tabulated below:

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Initial Res.		Neutron Exposure	Final Res.	
Order	Disorder	Neutrons/cm^2	Order	Disorder
2.10	5.14	2.5×10^{19}	2.63	5.16
2.12	5.14	4.2	2.88	5.15
2.08	5.07	6.7	3.47	5.05

All resistances are in 10^{-4} ohm.

It is evident that the degree of order in the initially well ordered sample is decreasing, but no change occurs in the disordered sample. The formulae of previous sections indicate that only about 1 atom in 100 has been displaced, too small to account for the degree of disorder observed to occur in the initially ordered samples. However, the lattice vibrational energy appears to be sufficient to have produced the amount of disordering observed.

(4) Copper-Be Alloy.[5] This is a typical dispersion hardening alloy. At 800°C the alloy exists as a solid solution. By quenching from this temperature the solution is retained at room temperature. Further heat treatments, at temperatures from 250 to 550°C for various times cause the supersaturated solution to decompose, with a Cu-Be compound precipitating out of solution first, followed by a growth in size of the precipitate particles with further heat treatment. As the precipitation process progresses, numerous properties of the alloy change. In the table below numbers proportional to the electrical resistance of samples as quenched, and after heat treatments at temperatures shown, are given before and after a neutron exposure of 10^{20} neutrons/cm^2.

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Electrical Resistance

	Before Exposure	After Exposure
As quenched	2345	2734
100 min at 250°C	2383	2518
100 min at 310	2000	2115
4 hrs at 400	1303	1431
18 hrs at 550	1653	1922

The electrical resistance increases in all cases. This change could be interpreted as being due to interstitially stuck displaced atoms, but the results on the disordered Cu_3-Au alloy argue against this conclusion. The change in the as quenched sample could be due to radiation-induced precipitation, the changes in the others, or perhaps in all if some precipitate is already present in the as-quenched sample, could be due to radiation induced resolution.

(5) Stainless, Monel, Kovar.[6] Exposure of these alloys has indicated changes in Rockwell B hardness number of about 15 in some cases. Monel in an initially cold worked state did not harden, initially soft annealed material showed an increase of $\Delta R_B = 20$.

Kovar is an "irreversible" Fe-Ni-Co alloy. Samples were prepared in the f.c.c. form by slow cooling from 800°C, in the b.c.c. form by quenching in liquid air. Exposures of 10^{18} neutrons/cm^2 in the hollow U metal slugs at Oak Ridge produced changes in electrical resistance of several percent, indicative of radiation-induced transformation from the f.c.c. to the b.c.c. phase.

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(6) Stress-Relaxation.[7] Metals in general exhibit the phenomena of creep and stress relaxation, particularly at elevated temperatures. If a material is placed under constant load, its dimensions change (creep); if it is stressed to a fixed deformation, the stress decreases (stress-relaxation).

Samples of stainless steel, 24ST aluminum alloy, and Be metal have been exposed to 5×10^{19} neutrons/cm^2 while being constrained in an elastically bent form, with a maximum strain of 5×10^{-4}. After exposure the constraint was removed and from the shape taken on by the sample the degree to which the original elastic stress relaxed during radiation was determined.

The fraction of residual stress in the irradiated sample after exposure and the residual stress in an identical control sample not irradiated, are tabulated below for Al alloy 24ST, stainless steel, and Be metal:

Metal	Control Sample	Exposed Sample
Al-24ST	97%	93%
Stainless	93	82
Be	78	74

The changes for the Al24ST and Be are probably not greater than the precision of the measurement, but the change in the stainless steel specimen is significant. These measurements indicate that the rate of stress-relaxation, and probably creep, at 50°C, is significantly increased by the presence of radiation.

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B. Semi-Conductors

(1) Ge, Si, Se, Cu_2O, etc.[8] These materials are used as diode rectifiers in high frequency communication systems. Their properties are markedly dependent on small amounts of impurity, and on heat treatment. Samples of these materials have been exposed in various positions in the Oak Ridge pile, and extensive data have been accumulated. Changes in electrical properties by orders of magnitude are observed.

The changes that have been observed in Ge rectifiers are shown in Fig. 1. The conductivity in Ge can be either by positive holes, P type, or by electrons, N type, depending on the impurity causing the conductivity. Materials of these two types have been exposed, the resistance being measured while the samples are in the pile. Material originally N type behaves as shown in Fig. 2, the resistance reaching a high maximum value, then decreasing. Originally P type material behaves as in Fig. 3, showing a merely monotonic decrease.

This behavior can be understood and semi-quantitatively described in terms of a model in which neutrons produce lattice vacancies and displaced atoms of Ge which remain interstitially stuck in the lattice and that probably the former act as electron acceptors, producing P type conductivity.

(2) Graphite.[9] This material is used in very large amounts in reactors as the moderating material, and has been studied more extensively than any other substance. The crystal structure, dimensions, strength, heat and electrical conductivity, elastic modulus, energy content all change in very significant ways. The results to date have been recently summarized by Fowler. Some of his data are given in the curves of Figs. 4 to 8. Most of the curves are

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self-explanatory. The unit of exposure given is 1 MWD/CT, which is equivalent to an integrated neutron flux of 6×10^{17} neutrons/cm^2.

The change in lattice constant observed can be described in terms of displaced atoms stuck in the region between the graphite crystal structure planes, probably as C_2 molecules. The changes observed, and the degree of broadening of the X-ray diffraction lines indicate about 6% of the atoms displaced in the longer exposures.

The changes in properties that are produced by exposure at temperatures of the order 50 to 100°C can all be partially annealed out by heating the graphite to elevated temperatures. During such heating the energy stored in the lattice as additional potential energy of the displaced atoms is liberated, and this has been measured by two methods, one a dynamical one, one a measurement of the excess heat of combustion.

In the dynamical method the sample is heated at a constant rate to about 450°C and the energy released measured, the data being shown in Fig. 5. Evidently the stored energy is released at a maximum rate near 200°C, but a considerable fraction is released only at high temperatures. Furthermore, material of longer exposure has most of the energy stored in such a manner that it can be released only at higher and higher temperatures.

The total stored energy is plotted in Fig. 6. Capsule graphite has been exposed near 50°C, test hole graphite near 125°C. The effect of temperature of exposure on the stored energy is apparent. The magnitude of the stored energy is evidently quite large, reaching values of 400 cal/gm, sufficient to increase the temperature by 2000 C if it were adiabatically released.

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The change in thermal conductivity is shown in Fig. 7. The effects of subsequent annealing in reducing this change, and the fact that longer exposure makes the change in property more difficult to anneal out is shown in Fig. 8.

C. Ionic Crystals

These materials are insulators, and differ in an important way from the metals and semi-conductors, the effect of γ rays being of considerable importance here. Electrons may be excited from the filled bands into normally empty bands, leading to photoconductivity. These excited electrons may become trapped in lattice vacancies, giving rise to new absorption bands.

(1) MgO.[10] The effect of pile radiation on the absorption spectrum of single crystals of MgO has been studied. New absorption bands in the ultra-violet and visible are produced. The rates at which these bands disappear with thermal annealing after exposure, and the similarity between the pile irradiated and the X-ray irradiated samples indicate that the effects are primarily due to electronic excitation rather than to displaced atoms.

(2) BeO.[11] This material is proposed for use in the high temperature power pile. Samples have been exposed up to 2.2×10^{20} neutrons per cm^2. A fractional change in length of 2×10^{-4} was observed, a change in thermal resistivity by a factor of 1.6. It is interesting to note that in similar exposures of Be metal, no fractional change in density greater than 5×10^{-5}, in resistivity greater than 2×10^{-3} was observed. This may bear out the ideas of the theory that the insulators are much more sensitive to neutron

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bombardment than are metals of similar elements. However, the detailed nature of the polycrystalline BeO mixture may play an important role here, rather than true volume effects, and experiments on single crystals are needed.

(3) NaCl. Experiments to measure the electrical conductivity of single crystals of NaCl in the temperature range 300° to 750°C, while being irradiated in the Oak Ridge pile, have been carried out by D. K. Stevens. No significant changes have been observed. Since the conductivity ordinarily is almost entirely by diffusion of Na^+ ions under the action of the electric field, this experiment was designed to study the effect of neutron bombardment on rate of diffusion, a topic of fundamental interest in the whole subject. No change was observed at temperatures above 500°C. Below this temperature small effects were noted but the interpretation of these is not clear, since the increase in current observed may be due to photocurrents.

D. Fissionable Materials

(1) U Metal. In the Hanford and Oak Ridge piles the fissionable material is natural uranium metal, in the form of γ extruded rods which have been subjected during fabrication to various thermal cycles and mechanical cold work. This material is quite coarse grained, grain size of the order 5 mm, and the mechanical and thermal properties are anisotropic. After an exposure in which about 10% of the U^{235}, i.e., about 1 atom in 1500 of the U metal, has fissioned, severe deformations in the shape of the slugs are observed. An originally cylindrical rod becomes wavy, with raised and depressed

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areas 1 cm^2 in area, several mm in height or depth. The rods become warped and out of round by several mm over a length of 20 cm.

No changes in the fine detail of the surface have been observed. No changes in density or in thermal conductivity, both of which have been measured, have been found. The surface hardness after exposure becomes quite variable, from 40 to 100 Rockwell G, although originally it is quite uniform, 75 ± 3. No correlation between the hardness and the geometrical contour has been found. Casual observations at Hanford indicate that the exposed material is considerably more brittle than originally. The changes in form may be due to the temperature spikes during irradiation, or perhaps to flow under thermal stresses resulting from the non-uniform heat development during exposure. The changes in hardness could be plausibly explained merely in terms of the foreign atoms produced by the fission process.

(2) Aluminum-Uranium 235 Alloys.[12] This material is an alloy of Al and U with a eutectic between pure Al and UAl_5 compound near 15% U. Samples containing 5.7, 15 and 17% U, enriched to 95% U^{235}, have been exposed for various periods, at a temperature between 50 to 100°C. Properties such as dimension, electrical and thermal conductivity, and hardness have been measured after bombardment and compared to original values, or to properties of similar samples.

The changes in electrical conductivity, plotted as ratio of final to initial resistance for each alloy, are shown in Fig. 9. The axis of abscissae is the fraction of total atoms of alloy that have fissioned. Evidently the change in resistance, for a given fraction of fissioned atoms, is

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less in the lower concentration alloy. This implies that there is an "aglomeration" effect, that the principal damage occurs in the UAl_5 particles rather than in the Al matrix. This conclusion is based on the fact that the UAl_5 particles are larger than a fission track range, so that the Al matrix is not much affected by fission recoils.

The changes in thermal resistivity, plotted in the same way, are shown in Fig. 10. Here the data all lie on one curve, probably because of the much lower precision of these measurements.

The changes in hardness of these alloys on exposure are shown in Fig. 11. Severe cold rolling of the original unexposed material produces a maximum hardness of about 80. The sample of greatest fission fraction showed some brittleness, and broke after a small amount of plastic bending.

It should be noted from these results that no changes seriously affecting its utility in a reactor have occurred in this alloy after 1 atom in 150 of the total alloy has fissioned; this is a depletion of 30% of the original U^{235} present. It is ten times the fraction fissioned that produces in U metal severe deformations in shape.

(3) <u>BeO-UO_2 Mixtures.</u>[11,13] This material is a hot pressed mixture of the two oxides, proposed for use in the fuel elements of the high temperature power pile. Exposure up to 2×10^{20} neutrons/cm^2, at temperatures of the order $700^{\circ}C$, produces changes in dimensions, elastic modulus,

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and thermal conductivity. The length change observed was + 0.8‰, the modulus and strength change - 25‰, the thermal conductivity decreased by a factor of 6.

Much more complete experiments have been made by L. P. Hunter at Oak Ridge, in which the thermal conductivity is measured at elevated temperatures while the samples are under exposure in the pile. The data for the change in thermal conductivity are given in Fig. 12, for the Argonne and the Oak Ridge experiments. In the former, the conductivity is measured near room temperature, after exposure near 700°C. In the latter, the experiment is done at temperature. The exposure units here are in kwh/cc. In these samples 1 kwh/cc is equivalent to a fraction of total atoms fissioned $\simeq 3 \times 10^{-6}$. It is important to notice that the thermal conductivity, for example, has changed by a factor of 4 for a fissioned fraction $= 3 \times 10^{-5}$ while considerably smaller changes are observed in Al-U alloys for a fissioned fraction as high as 7×10^{-3}, 200 times as great a degree of depletion of U^{235}. This may indicate again the relative merits of metals vs insulating materials.

A comparison may be made of the effects observed in these mixtures, subject to fission recoils, and the BeO subject only to neutron recoils. The pure BeO reached a value of $K/K_0 = 1.6$ in a time about 100 times that

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required for the $BeO-UO_2$ mixture to reach $K/K_o = 1.6$. During these periods there have been 3×10^{-6} fission fragments per atom, in the mixture, and 5×10^{-4} neutron recoils/atom in the pure BeO, indicating a ratio of effectiveness of about 60.

(4) Graphite Impregnated with U Compounds. Hunter has performed experiments on the thermal conductivity of graphite impregnated with U_3O_8 and UC, exposed and measured while at elevated temperatures. The data are shown in Fig. 13, for both graphite and for $BeO-UO_2$. These experiments also permit a comparison to be made of the relative effectiveness of annealing during bombardment, vs annealing to subsequent to bombardment, using Hunter's data and the data of Fowler on graphite annealed after bombardment. This comparison is shown in the curves of Fig. 14; evidently simultaneous bombardment and annealing prevents the formation of displaced atom pairs to some extent and so reduces the damage. The curves are plotted to arbitrary exposure scales, forced to have the same slope near the origin.

These data also permit, when the exposure scales are compared as to absolute magnitude, the estimation of the relative effectiveness of fission fragments and neutron recoils. In this case the relative effectiveness appears to be about 100.

IV Summary

Numerous materials have been exposed to pile radiations and changes in properties sought for. In all cases except simple pure metals like Al and Be changes, varying in magnitude over wide ranges, have been found. The

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rate of change of properties in materials containing fissionable nuclei, and subject to fission fragment bombardment, is markedly higher than that due to neutron recoils. Some experiments suggest, as does the theory, that metals are less damaged than similar insulating materials.

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Dr 5737

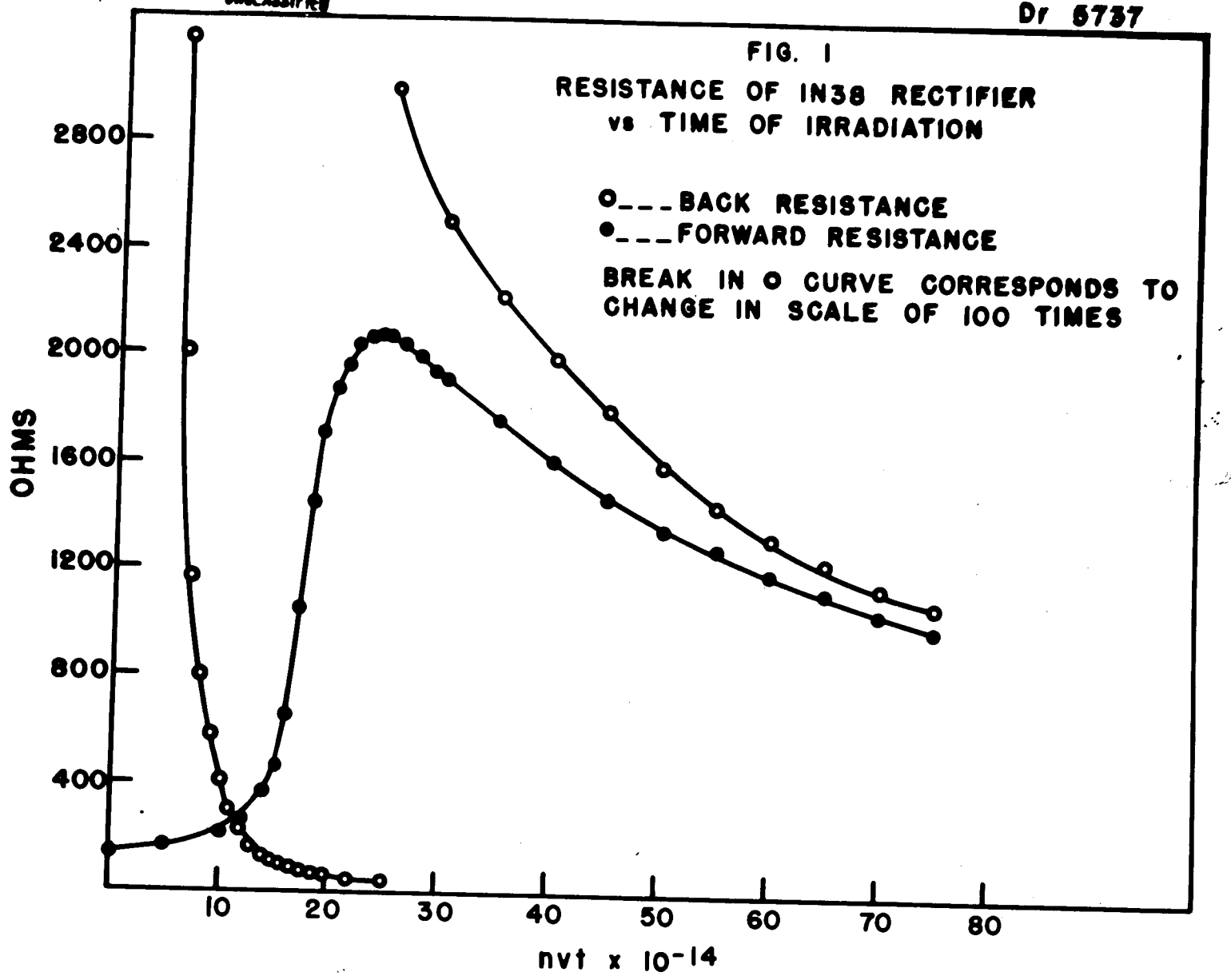

FIG. 1
RESISTANCE OF IN38 RECTIFIER vs TIME OF IRRADIATION

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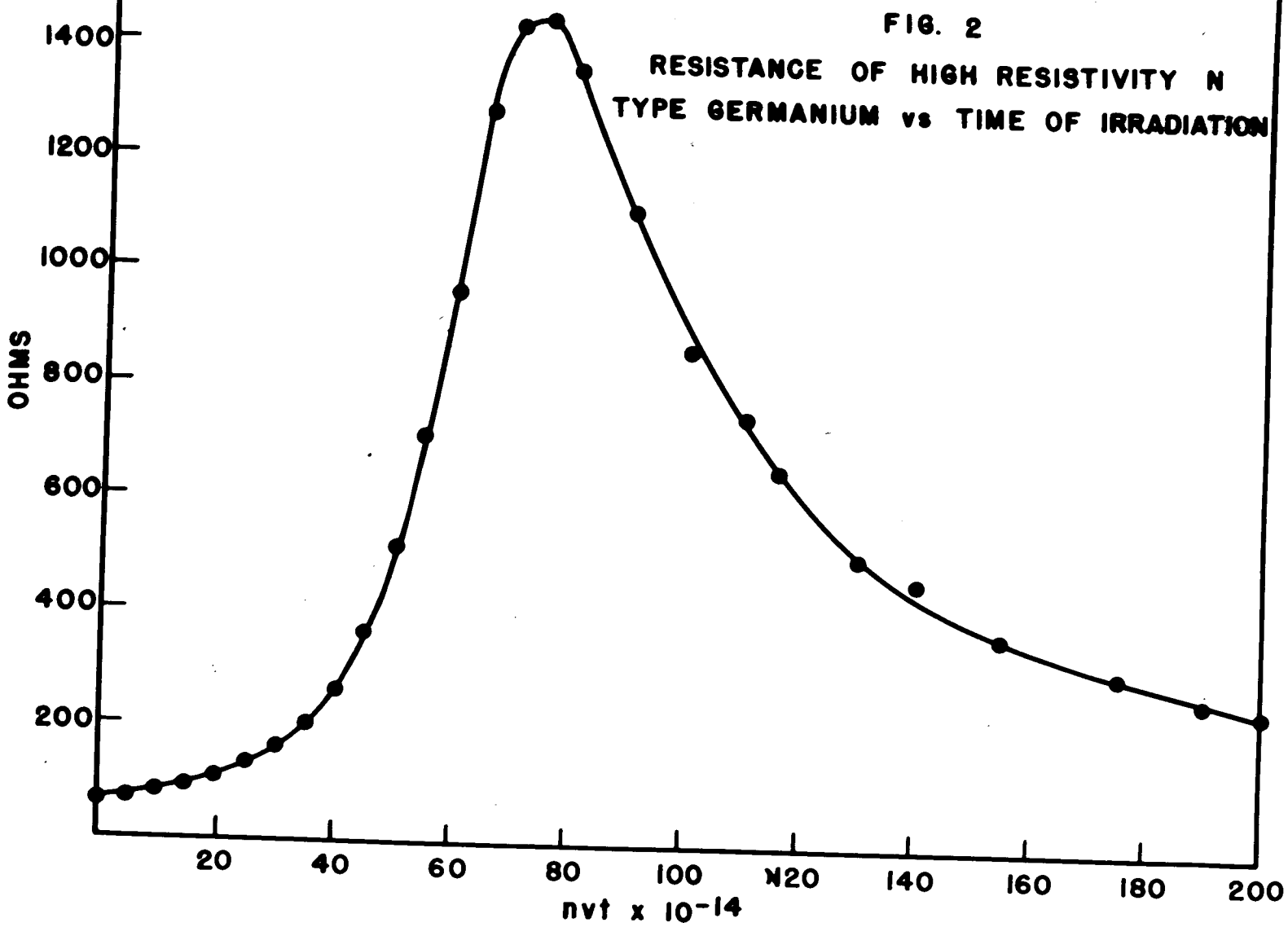

FIG. 2

RESISTANCE OF HIGH RESISTIVITY N TYPE GERMANIUM vs TIME OF IRRADIATION

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FIG. 3

RESISTANCE OF HIGH RESISTIVITY P TYPE GERMANIUM vs TIME OF IRRADIATION

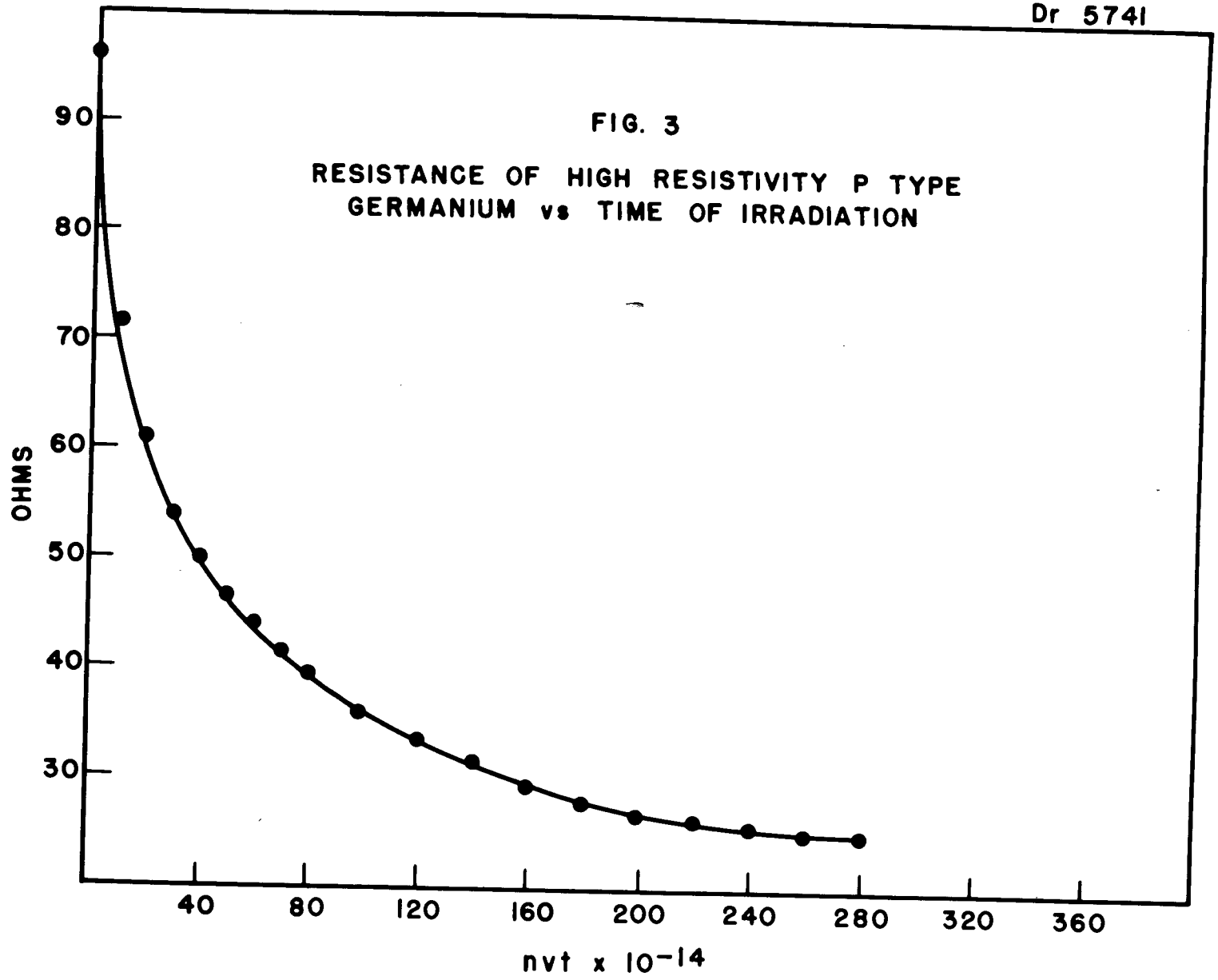

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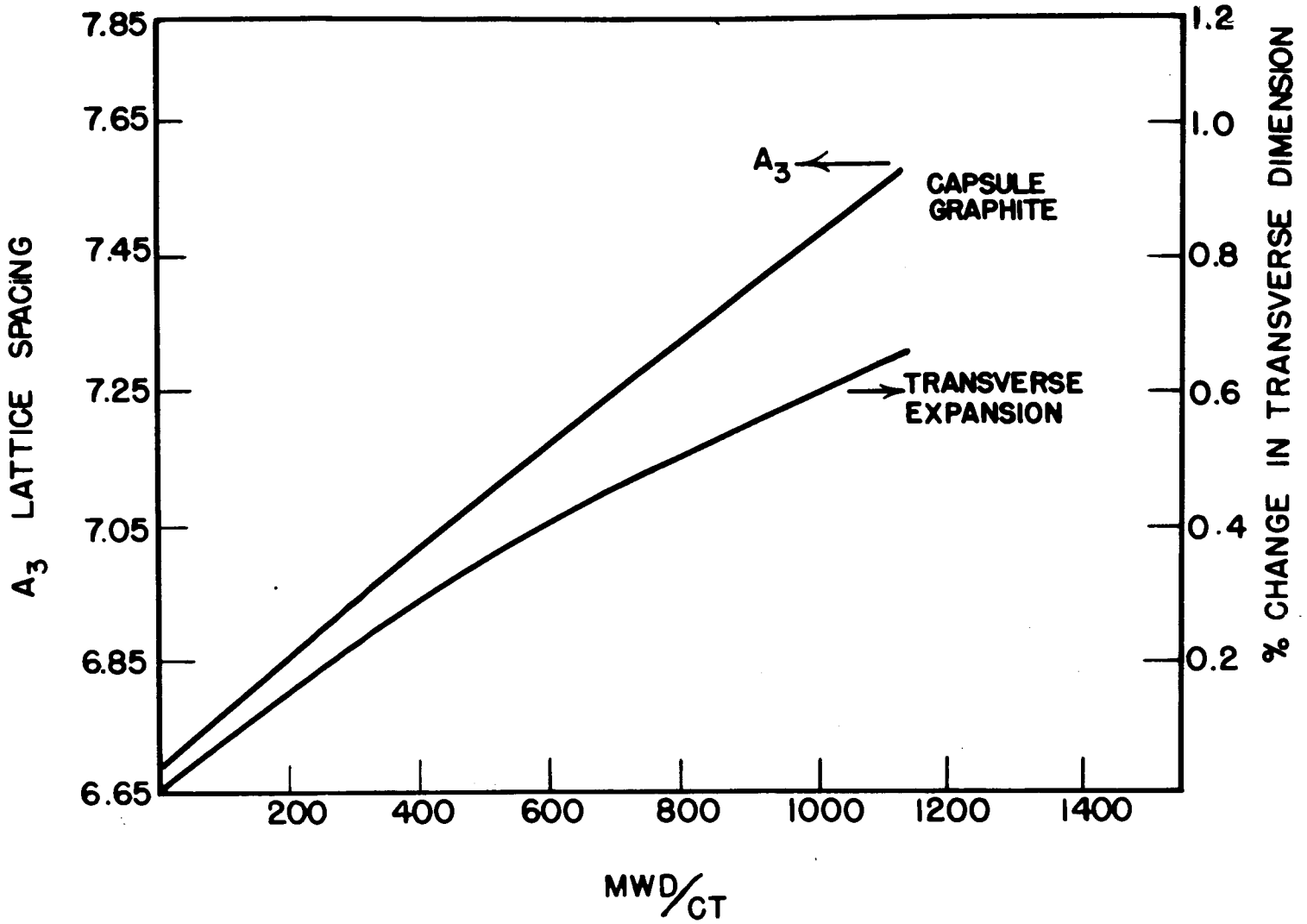
FIG. 4
EXPANSION OF GRAPHITE



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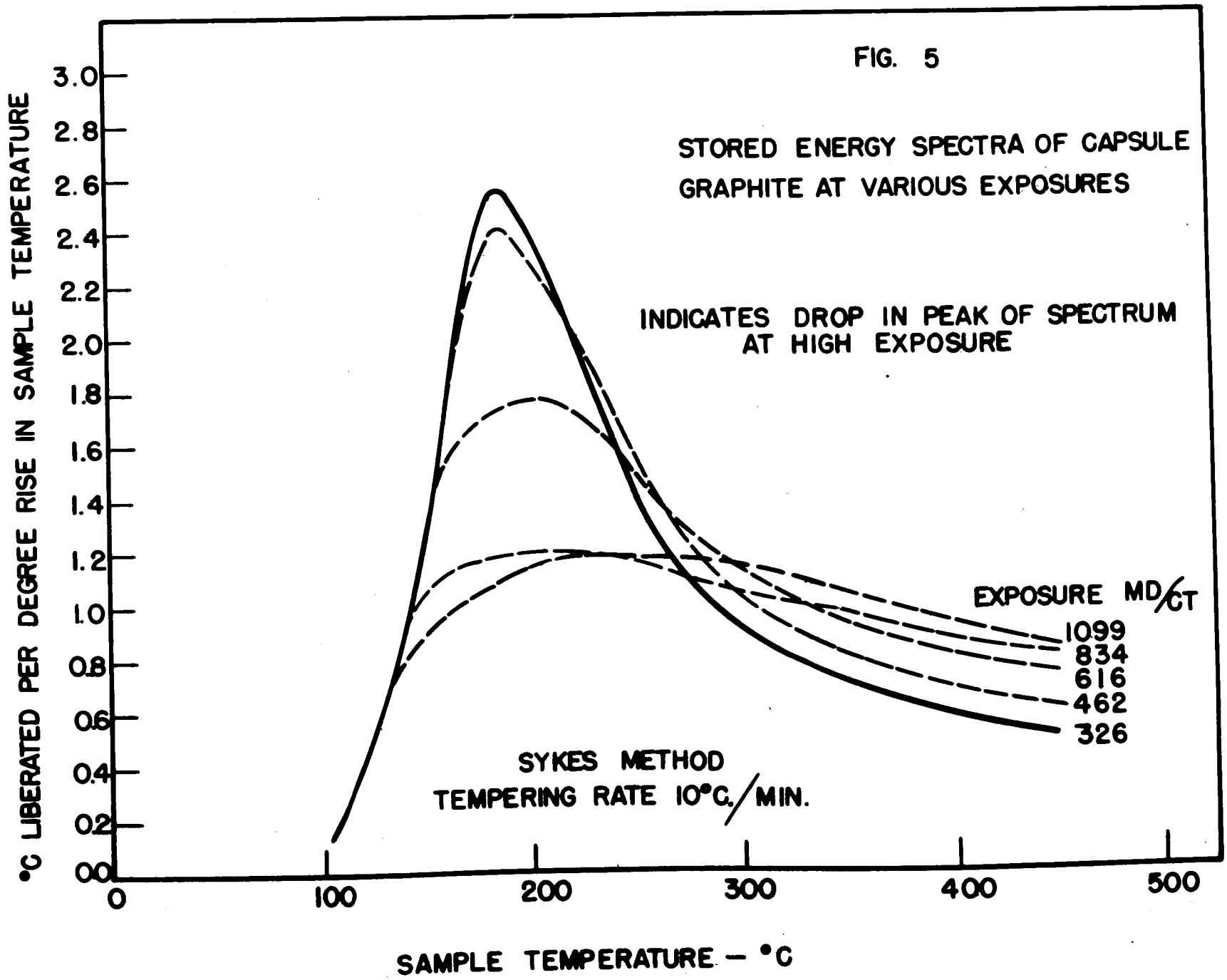
FIG. 5
STORED ENERGY SPECTRA OF CAPSULE GRAPHITE AT VARIOUS EXPOSURES
INDICATES DROP IN PEAK OF SPECTRUM AT HIGH EXPOSURE
EXPOSURE MD/CT
1099
834
616
462
326
SYKES METHOD
TEMPERING RATE 10°C./MIN.
°C LIBERATED PER DEGREE RISE IN SAMPLE TEMPERATURE
3.0
2.8
2.6
2.4
2.2
2.0
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
0
100
200
300
400
500
SAMPLE TEMPERATURE — °C

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FIG. 6 DR. 5734

INCREASE IN TOTAL STORED ENERGY AS FUNCTION OF EXPOSURE
(NBS DATA)

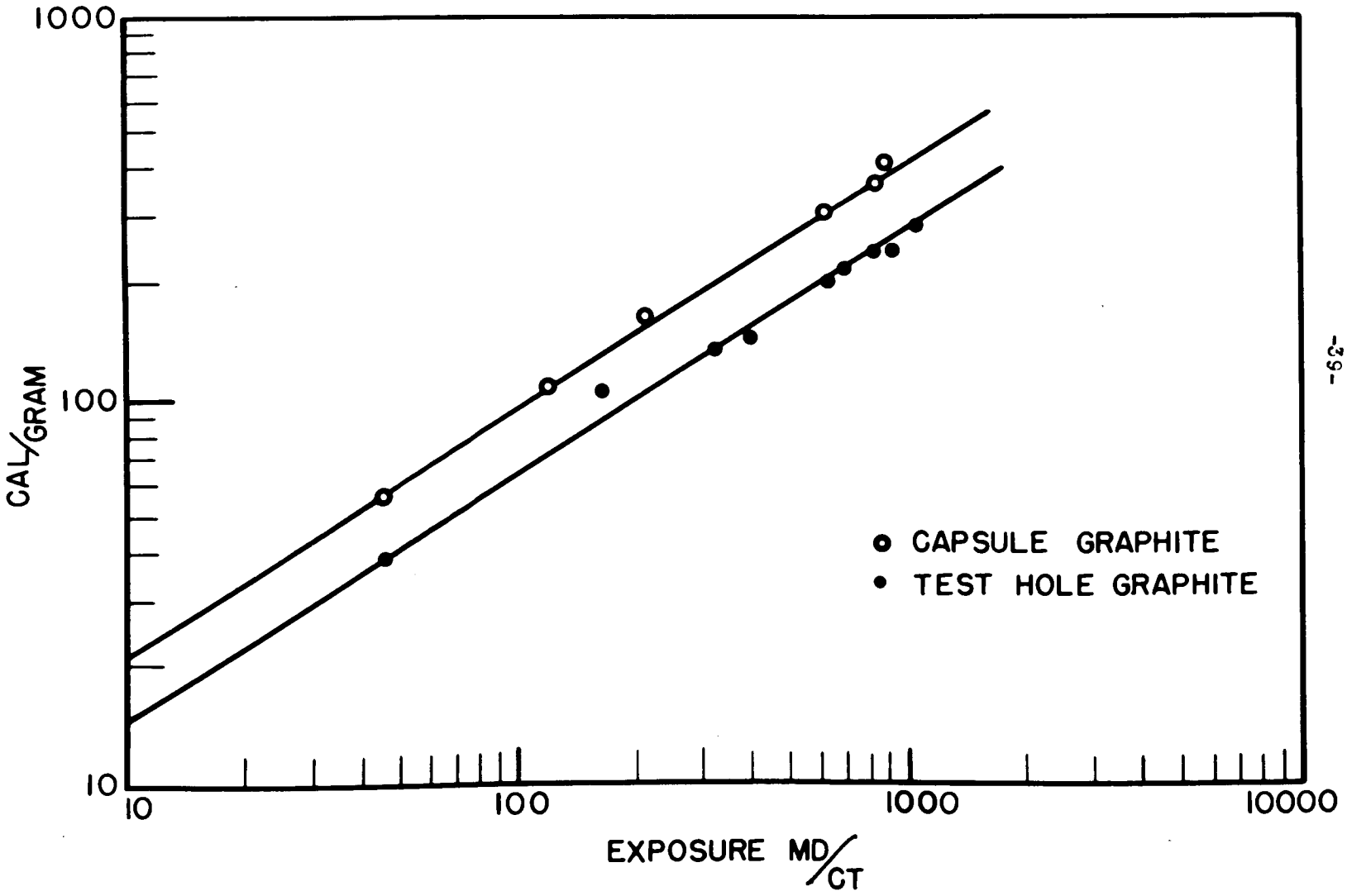

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FIG. 7

FACTOR OF INCREASE OF THERMAL RESISTANCE AS FUNCTION OF EXPOSURE

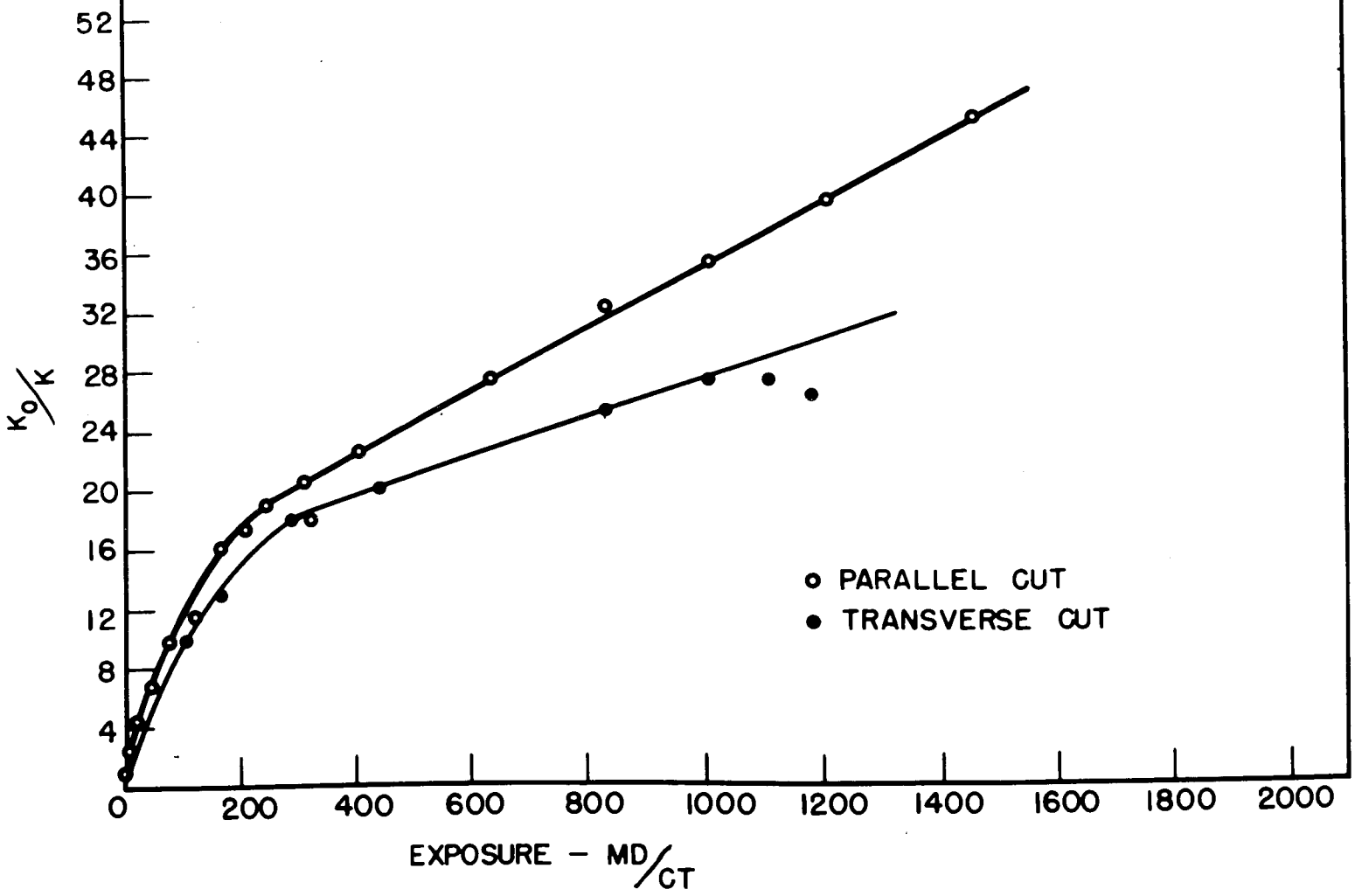

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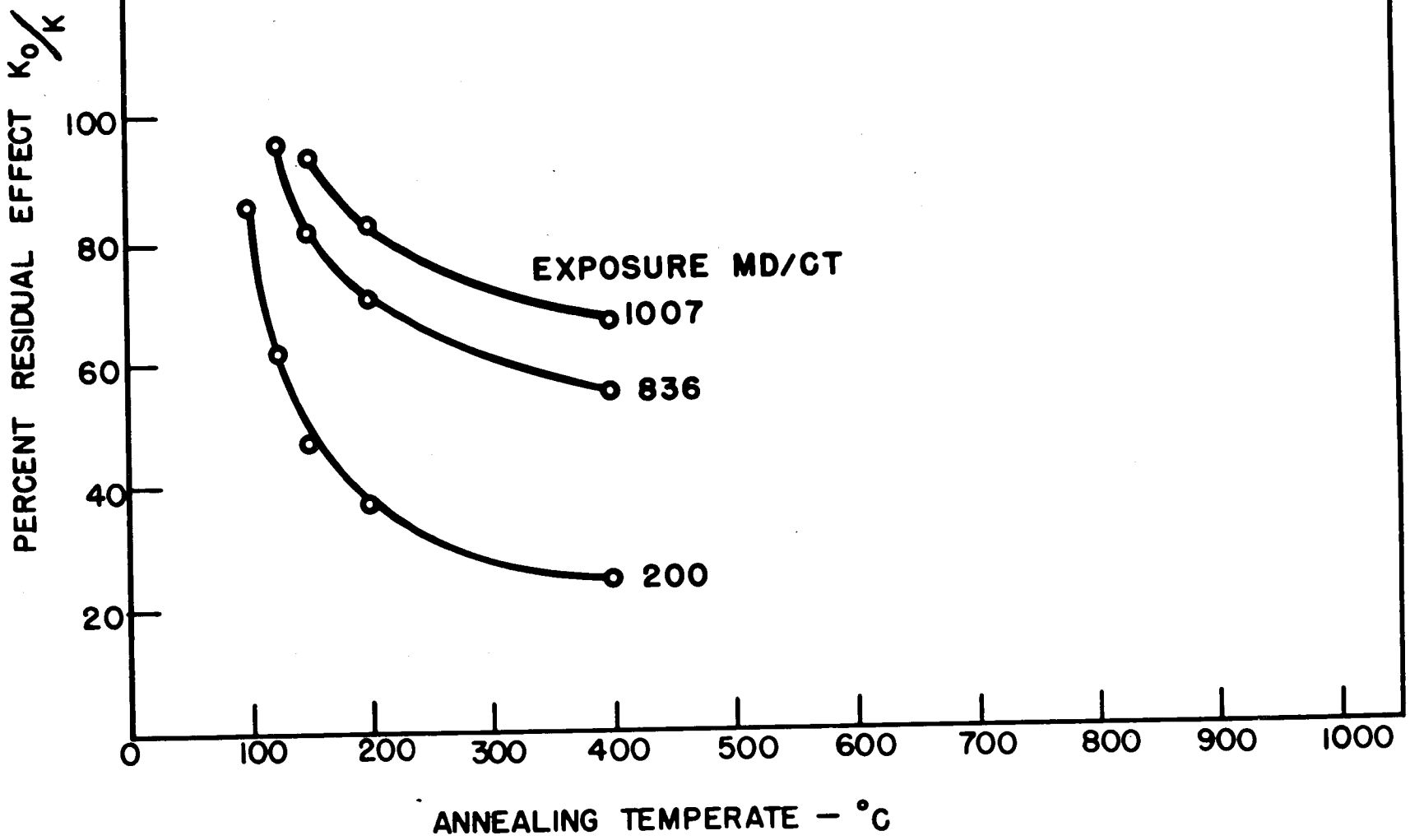
FIG. 8

CHANGE IN THERMAL RESISTANCE
EFFECT WITH THERMAL ANNEALING



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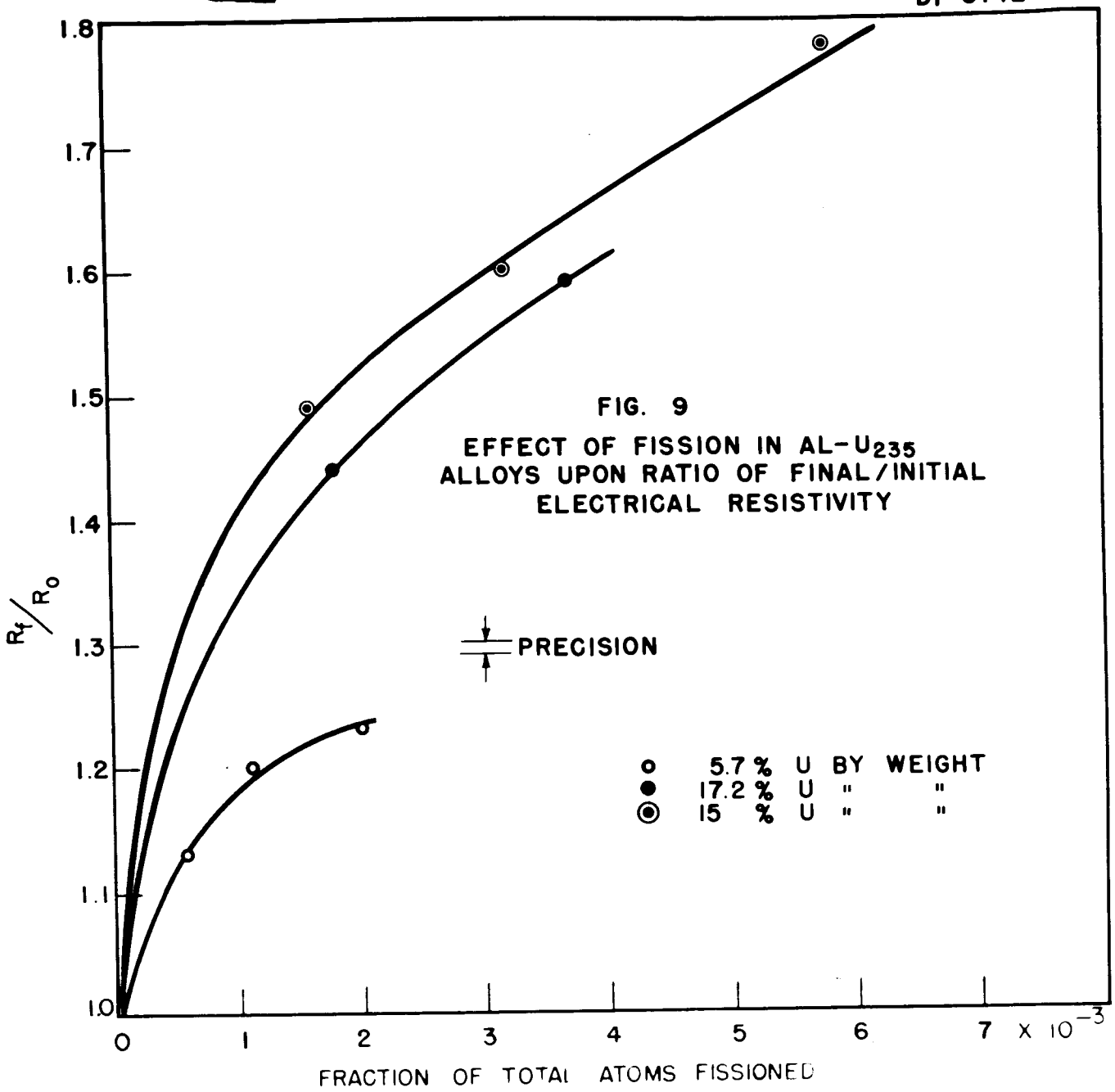
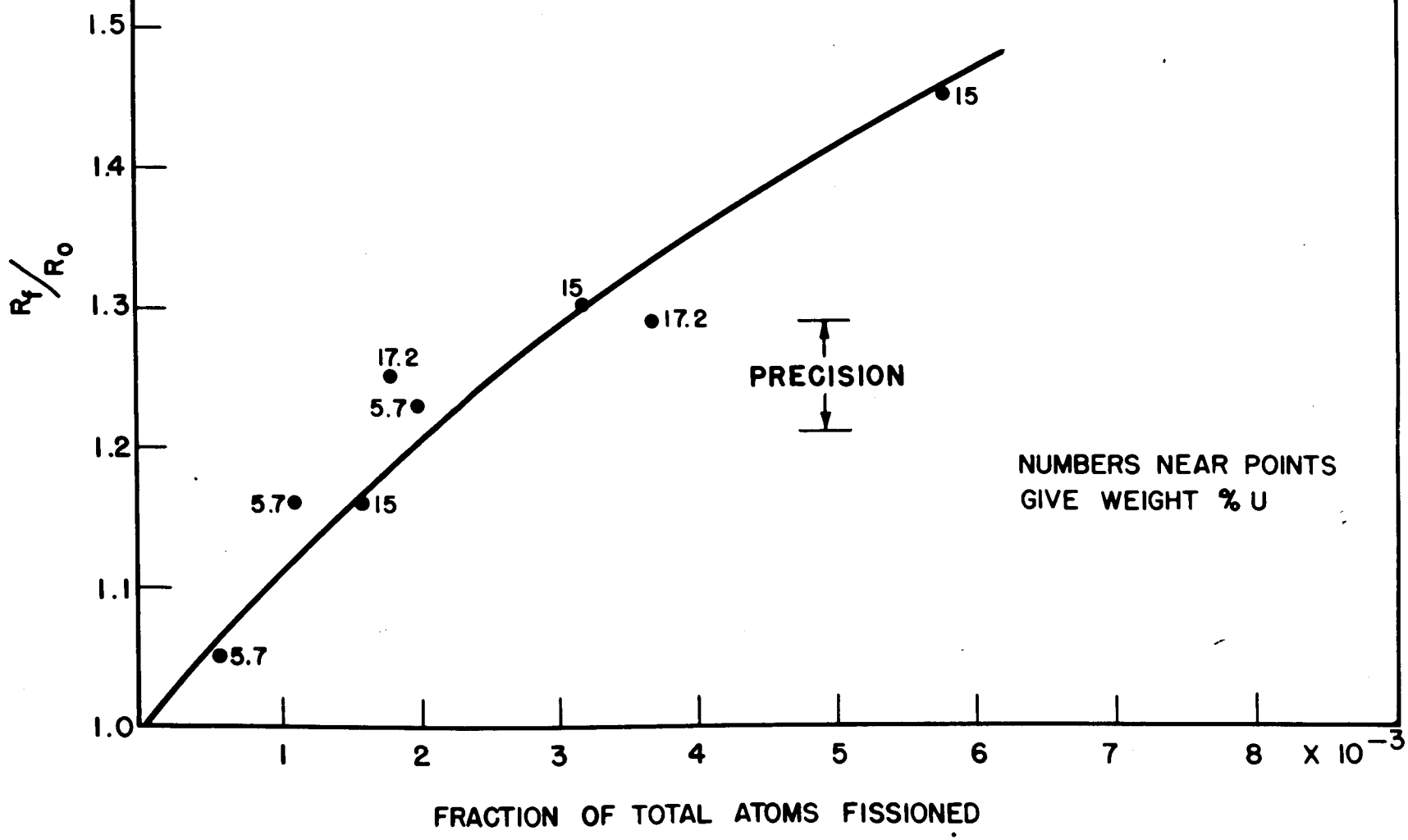


FIG. 9

EFFECT OF FISSION IN AL-U_{235} ALLOYS UPON RATIO OF FINAL/INITIAL ELECTRICAL RESISTIVITY

FIG. 10
EFFECT OF FISSION IN AL-U_{235}
ALLOYS UPON RATIO OF FINAL/INITIAL
THERMAL RESISTIVITY



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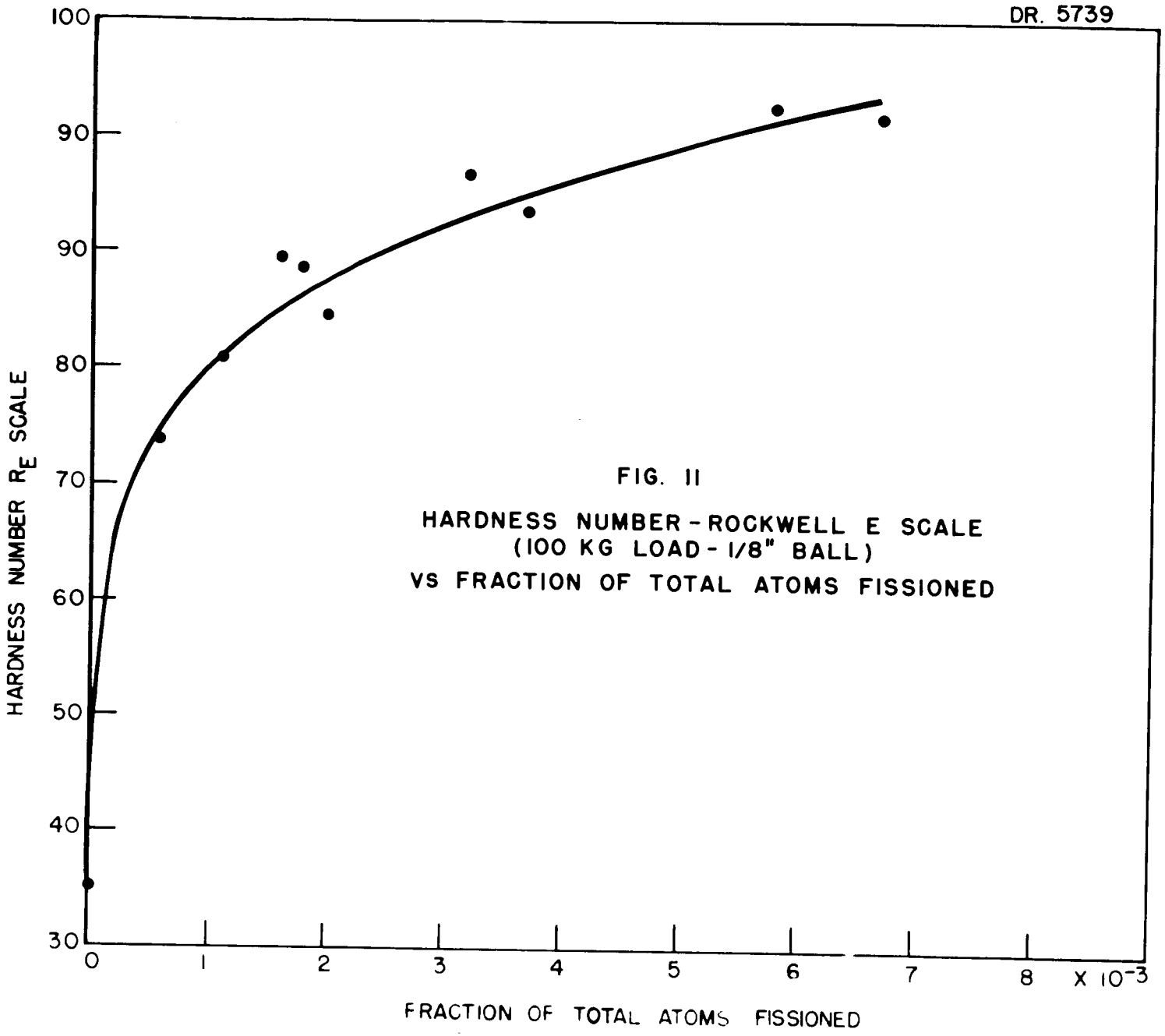
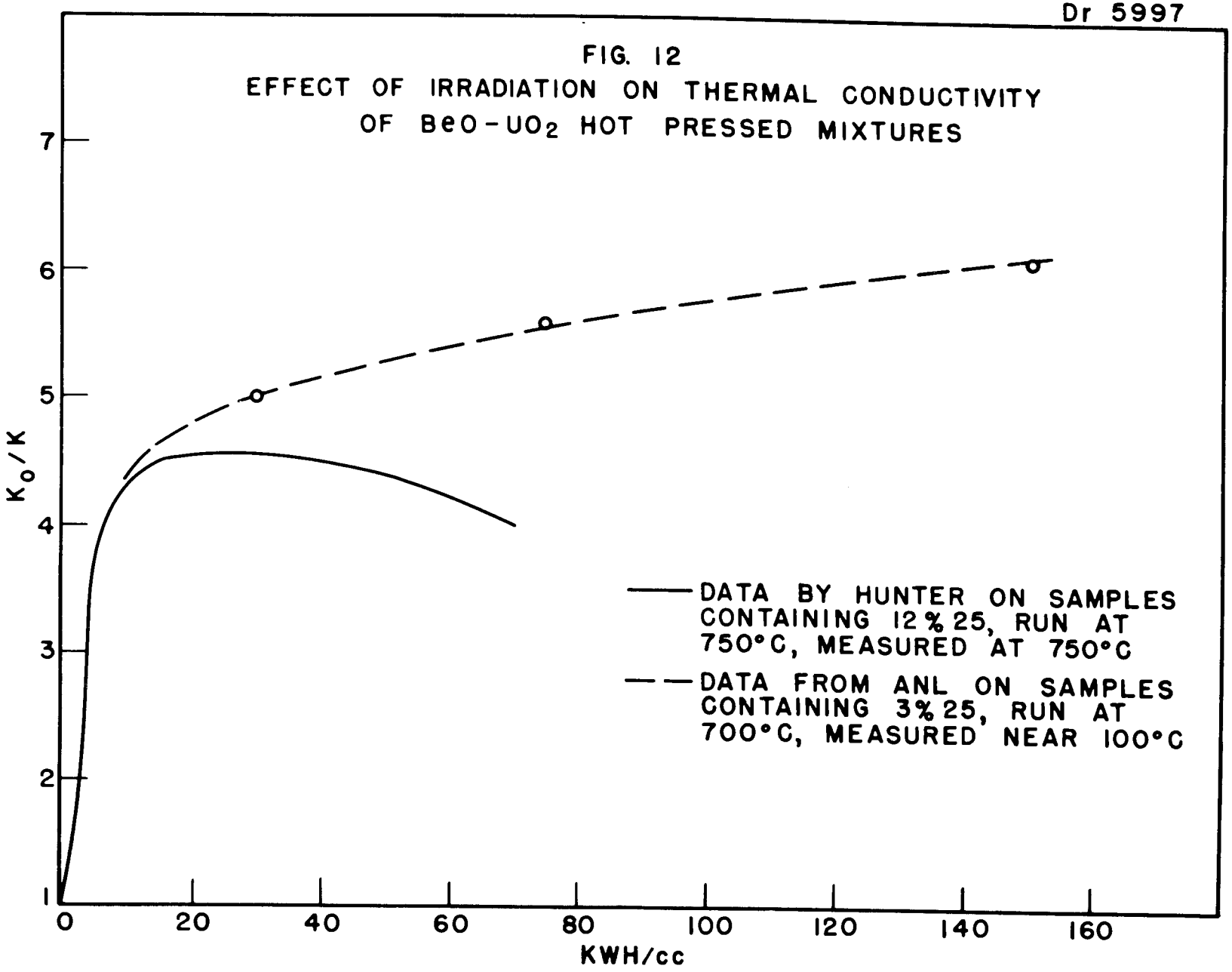


FIG. 11

HARDNESS NUMBER - ROCKWELL E SCALE
(100 KG LOAD - 1/8" BALL)
VS FRACTION OF TOTAL ATOMS FISSIONED

Dr 5997

FIG. 12
EFFECT OF IRRADIATION ON THERMAL CONDUCTIVITY OF BeO-UO_2 HOT PRESSED MIXTURES



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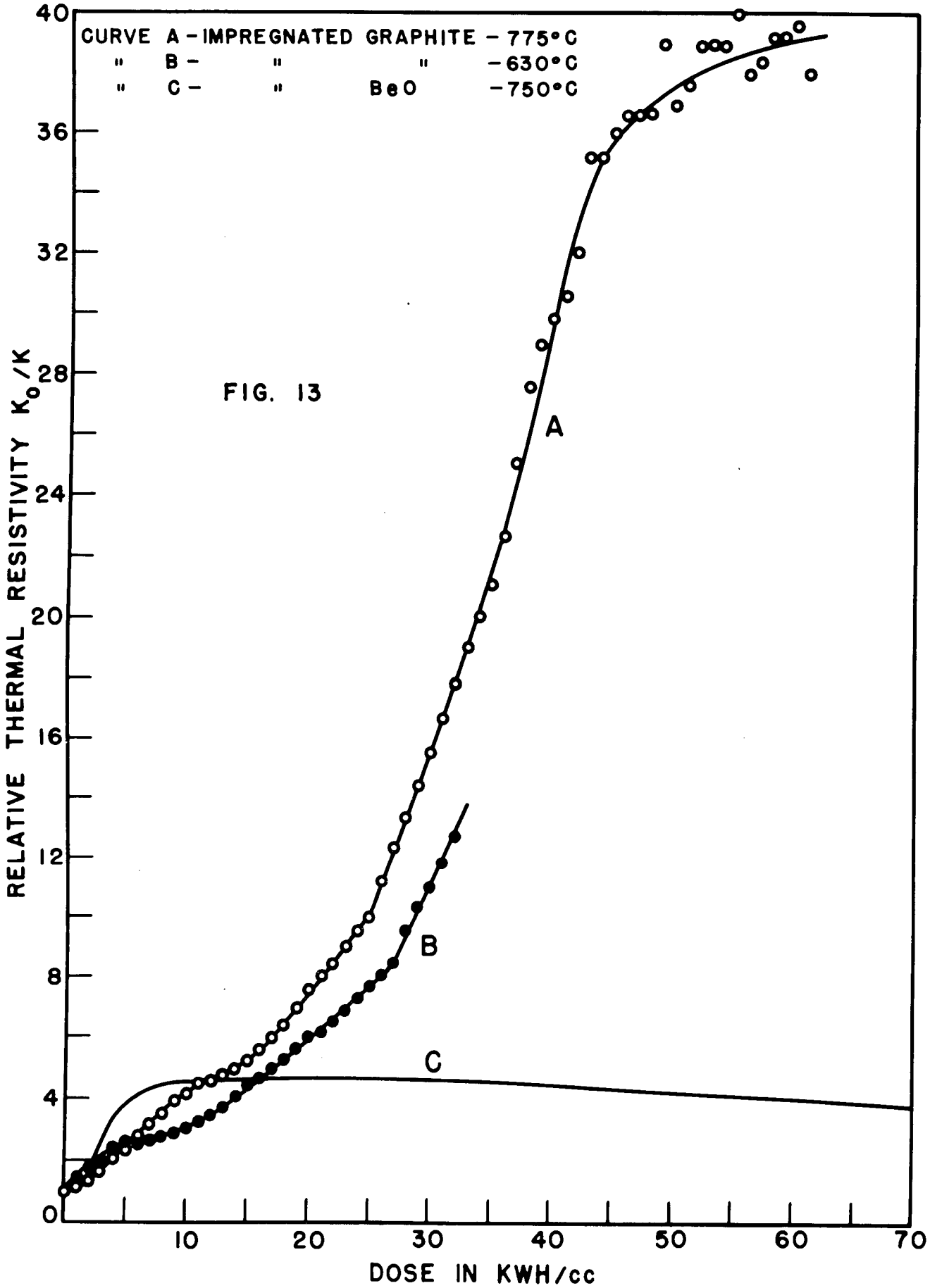

FIG. 13

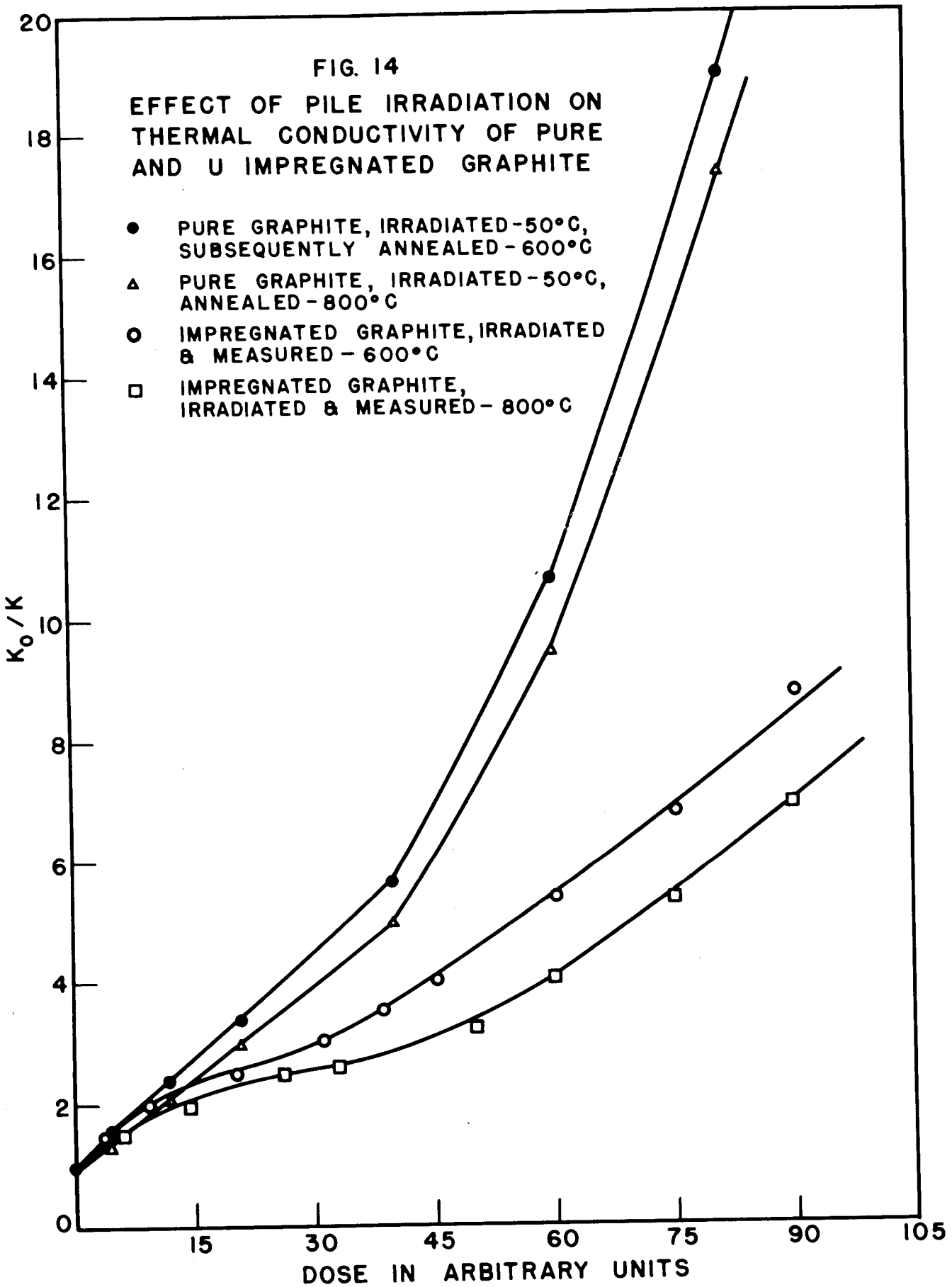
FIG. 14
EFFECT OF PILE IRRADIATION ON THERMAL CONDUCTIVITY OF PURE AND U IMPREGNATED GRAPHITE
PURE GRAPHITE, IRRADIATED-50°C, SUBSEQUENTLY ANNEALED-600°C
PURE GRAPHITE, IRRADIATED-50°C, ANNEALED-800°C
IMPREGNATED GRAPHITE, IRRADIATED & MEASURED-600°C
IMPREGNATED GRAPHITE, IRRADIATED & MEASURED-800°C
K_o/K
0
2
4
6
8
10
12
14
16
18
20
15
30
45
60
75
90
105
DOSE IN ARBITRARY UNITS