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PHYSICS DIVISION

QUARTERLY PROGRESS REPORT

FOR PERIOD ENDING MARCH 15, 1950

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PHYSICS DIVISION

A. H. Snell, Director
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QUARTERLY PROGRESS REPORT
for Period Ending March 15, 1950

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SUMMARY

The classified section of this Quarterly Report for December 25 - March 25, 1950, is, as usual, presented separately from the unclassified section (Unclassified Section, ORNL 694). After a short note on the present state of the important question of Pile Controls, the report devotes itself entirely to aspects of the Radiation Damage work.

The Pile Control instrumentation is now under active test in the chain-reacting MTR Mock-up. So far, it is very successful. Operating tests of the servomechanism will start shortly; reports on its performance will be awaited with interest.

Many of the lattice parameters for the orthorhombic UAl_4 crystal have now been evaluated by X-ray and neutron diffraction.

An improvement in technique has been worked out in relation to the use of bonded wire strain gauges for the study of dimensional stability of metals under irradiation (the slug blistering problem). The SR-4 gauges are better than those previously used, and irradiation tests on bonding cements have revealed some that are satisfactory so far as electrical resistance is concerned.

Dimensional and resistance changes induced in enriched and non-enriched U-Al alloy samples by Hanford irradiation show a variety of effects, some of which deserve closer examination. Resistance and volume effects do not parallel one another as a function of irradiation time. The Al-clad U-Al sandwiches of the type proposed for the MTR look as if they might show a maximum thickness increase of 0.5 per cent, with little change in other dimensions.

In-pile experiments on the change of thermal conductivity of U^{235}-impregnated graphite with irradiation are being continued. Similar readings are also being taken on a Be-U sample, and a Zr-U specimen is under preparation.

Experience gained at ORNL on the rather difficult in-pile creep experiment is being applied to the design of an improved apparatus for use at Hanford. Preliminary tests indicate that the new apparatus will give much greater precision than did the older device.

[redacted]

PILE CONTROLS

W. H. Jordan

During the past quarter the controls for the MTR Mock-up have been installed and have operated as predicted with a minimum of trouble. The servo developed by Straus and Cole has not yet been installed but should be within the next month. Tests on the simulator have shown that the time required to set back the pile from full power to one percent of power will have to be increased considerably over previous estimates. This may make set-back considerably less useful than anticipated.

A new fission chamber has been designed by Jordan and Gundlach and is now being tested. A power supply for the differential chamber has been designed and built.

The above work will be reported in detail in the Materials Testing Reactor quarterly report.

The differential ion chamber designed by MacNeille and Gundlach has been modified to make manufacturing simpler and give more range in canceling the current due to gamma rays. These modifications have been agreed to by Eastman Kodak Company and will be incorporated in future models.

A colloidal suspension of B^{10} in oil has been made by Acheson Graphite Company. The first samples received had a large amount of iron and other impurities which caused considerable long life activity under neutron bombardment. The process was improved to reduce these impurities and we now have 60 gm in an oil suspension. This is used to coat the plates of the safety chambers, both here and at other AEC laboratories.

PHYSICS OF SOLIDS

THE CRYSTAL STRUCTURE OF UAl_4

B. S. Borie,* M. A. Bredig

An X-ray diffraction study[1] of the crystal structure of the aluminum rich uranium-aluminum compound has shown that the formula is UAl_4. The unit cell is orthorhombic, a = 4.41 Å, b = 6.25 Å, and c = 13.60 Å, and contains four molecules. The space group is either Ima or Imma. The uranium atoms occupy four-fold special positions (e) of space group Imma or (b) of Ima, with $2\pi z/c = 40°$.

The work on this structure determination has been continued. It may be shown that the 16 aluminum atoms must be distributed among special positions 4(b), 4(e), and 8(h) of space group Imma (or among analogous positions of Ima). Other positions consistent with the symmetry of the space group would require the aluminums to be impossibly close to the uranium atoms.

It was assumed that the aluminums were in two sets of four-fold positions, (b) and (e), and a set of eight-fold positions, (h). The parameters were then chosen to agree best with atomic radii and with the intensities as observed with a North American Philips spectrometer. On this basis, the aluminums occupy positions:

$(000;\tfrac{1}{2}\tfrac{1}{2}\tfrac{1}{2})$ +

4: (b) $00\tfrac{1}{2}$; $0\tfrac{1}{2}\tfrac{1}{2}$

(e) $0\tfrac{1}{4}z$; $03/4\bar{z}$

$2\pi z/c = -40°$

8: (h) $0yz$; $0\bar{y}\bar{z}$; $0, \tfrac{1}{2} + y, \bar{z}$; $0, \tfrac{1}{2} - y, z$

$2\pi y/b = -12°$ $2\pi z/c = 113°$

The distribution of the atoms in the unit cell for the proposed structure, as viewed along the a axis, is shown in Fig. 1. Atoms shown in heavy outline

---

* Metallurgy Division

(1) Bredig, M. A., *et al.* "X-Ray Diffraction" *Physics Division Quarterly Progress Report for Period Ending December 15, 1949*, ORNL 576, p. 6 (Feb. 7, 1950).

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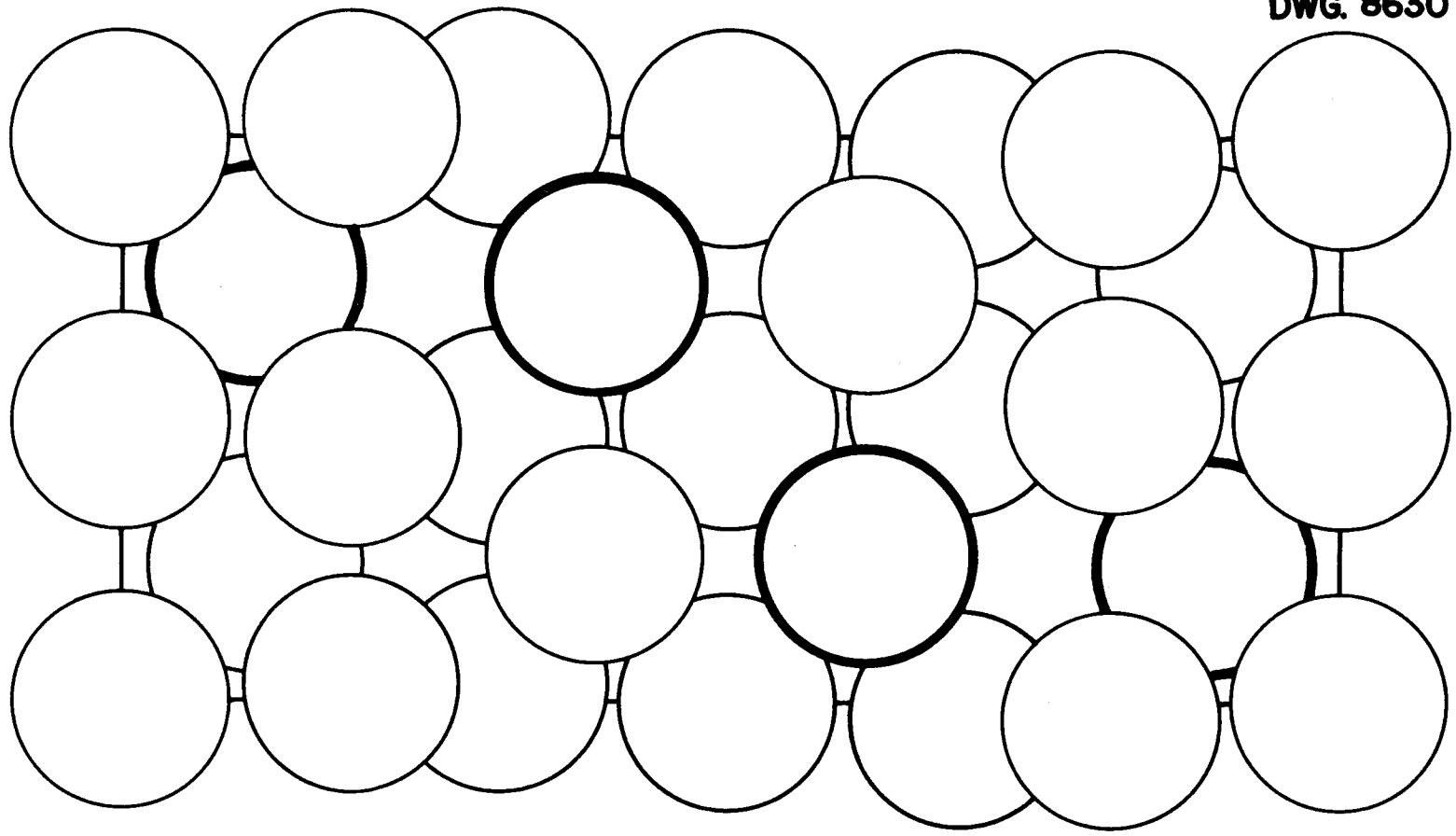

FIG. 1

UNIT CELL OF UAl_4, PROJECTED ALONG a AXIS. URANIUM ATOMS SHOWN IN HEAVY OUTLINE

are uranium. Both the uraniums and aluminums are arranged in two layers parallel to the yz plane at $a = 0$ and $a = \frac{1}{2}$. For this structure, the coordination numbers are, for uranium, 13, for the aluminums in (b), 12, in (e), 13, and those in (h), 11.

A comparison of the observed X-ray diffraction pattern using copper K_α radiation and the pattern calculated on the basis of the proposed structure is shown in Fig. 2. Lines 105, 024, and 006, which were observed as a single line, could probably be made to be coincident by an adjustment of the cell size within the limits of the accuracy of the measurements.

Since the scattering power of uranium for X rays is about seven times that of aluminum, the characteristics of this pattern are not very sensitive to the aluminum positions. However, for neutrons, the scattering power of uranium is only slightly more than twice that of aluminum. With the assistance of C. G. Shull, a neutron diffraction pattern of the compound was obtained and was shown in Fig. 3, compared with the calculated pattern.

The proposed light atom positions must be regarded as approximate ones since indexing for both the X-ray and neutron patterns was possible for only a few relatively low order lines. An effort to obtain sufficiently accurate intensities from the Weissenberg patterns to permit a Fourier synthesis of the electron density, and hence more precise values for the aluminum positions, is planned.

Because the aluminum positions are only approximate, a table of interatomic distances has not been prepared.

Chemical analysis and a direct density determination have been found to differ significantly with the values predicted by the structure. For a perfect UAl_4 structure, the density should be 6.12 g/cc and the compound should contain 68.8% uranium by weight. The corresponding observed values are 5.35 g/cc and 65.6%. This discrepancy may be resolved, to some extent, by assuming that some of the uranium sites are unoccupied or by assuming substitution of aluminum for uranium.

If substitution is assumed, approximately 21% of the uranium sites must be occupied by aluminum atoms to account for the observed density. This corresponds to a composition of 62.4% uranium. If one assumes vacancies, the

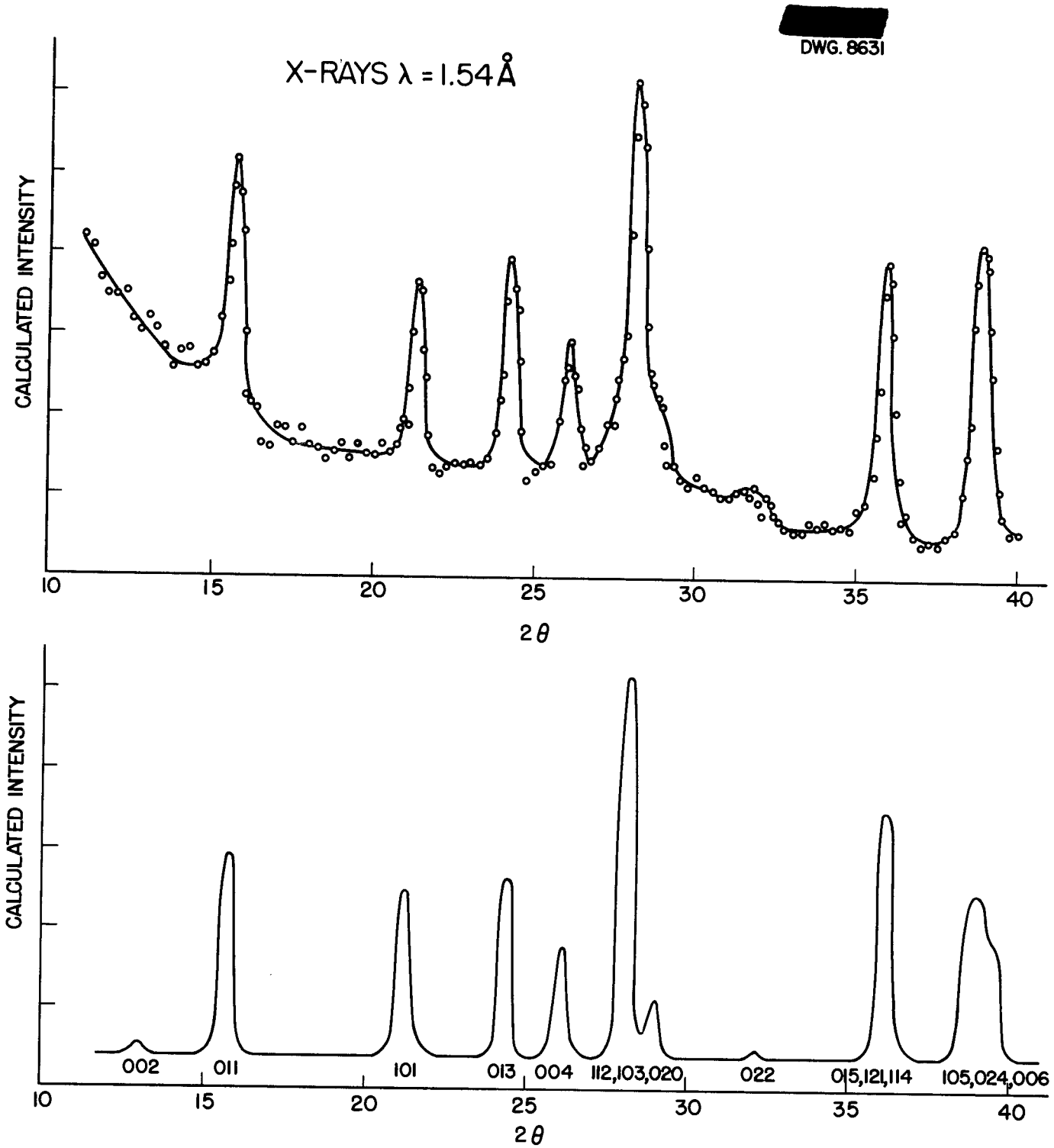

FIG. 2

COMPARISON OF OBSERVED AND CALCULATED X-RAY PATTERNS FOR UAl_4

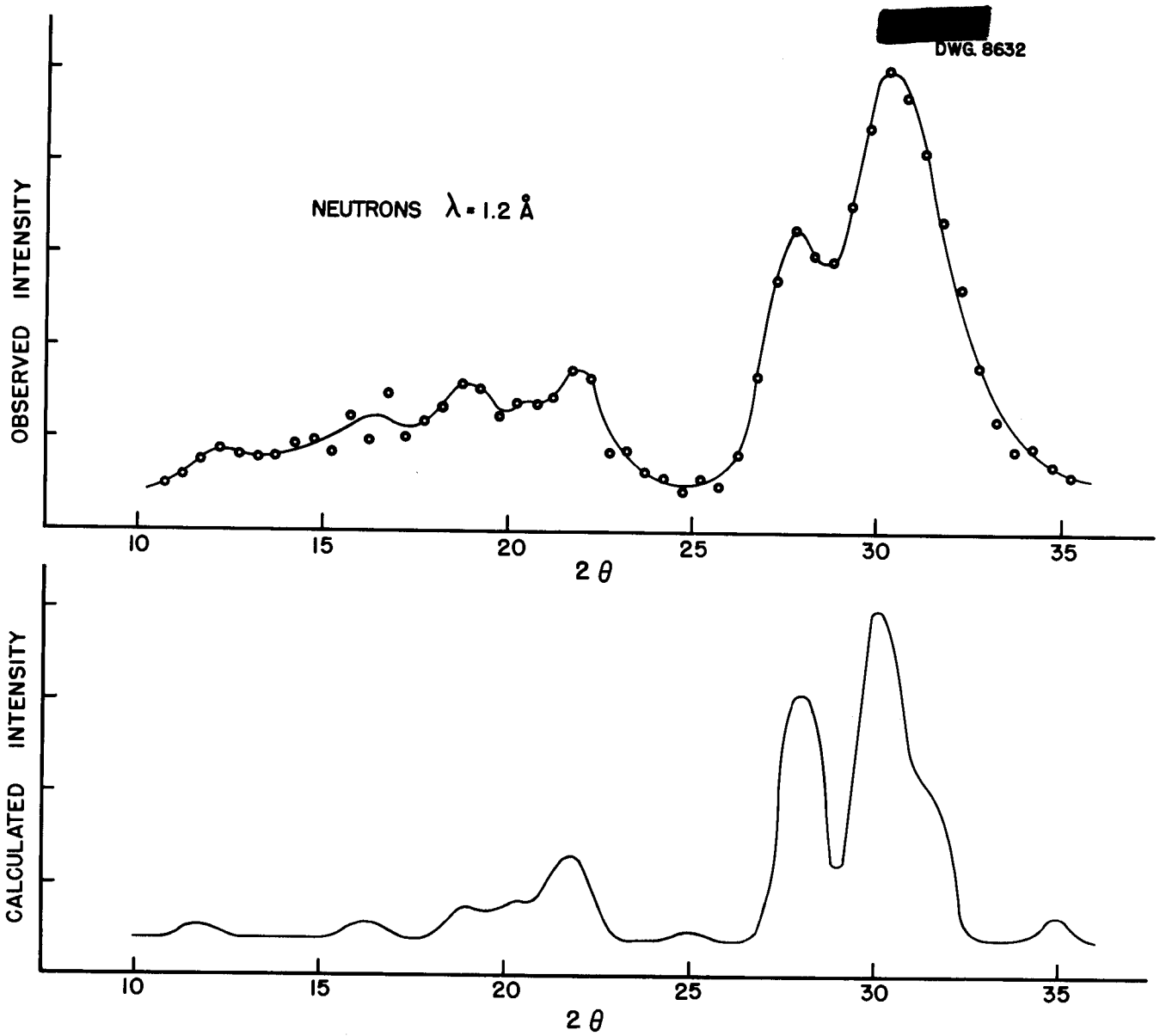

FIG. 3

COMPARISON OF OBSERVED AND CALCULATED NEUTRON PATTERNS FOR UAl_4.

observed density predicts that 18.5% of the sites are unoccupied. For this situation, the composition would be 64.2 percent uranium. Hence, unoccupied uranium sites seems the more plausible explanation. There are some indications that the density and the cell size may be slightly larger than their reported values, and if this is actually the case, the observed and calculated uranium compositions would be in better agreement.

BONDED WIRE STRAIN GAGES

M. R. Goodman

Investigation was made of the stability of SR-4 Dual Lead type gages of 100 ohms resistance. These gages appear to perform much more satisfactory than the previous type of gages used. Their behavior characteristics will be described in a future detailed report.

With satisfactory SR-4 strain gages at hand, the next step toward the ultimate objective of employing such gages to observe the effects of irradiation on uranium and other materials was to irradiate components of the gage itself to see whether or not the components would withstand the irradiations and not deteriorate appreciably. Therefore, irradiation samples were made consisting of gage Bakelite or Duco cement sandwiched between 2S aluminum. Half of the samples were waterproofed by means of application of Cerese wax. Insulation resistance measurements were made by means of a high megohm bridge accurate to within a few percent.

Specimens Nos. 1-8 were irradiated in the pile for a total fast flux of 1.5×10^{17} fast neutrons/cm^2 at a temperature of 50°C. Specimens Nos. 9-16 were exposed to 2.4×10^{18} fast neutrons/cm^2 at a temperature of 25°C. Inspection of all samples after irradiation indicated that the colorless Duco cement had turned bright yellow and had become brittle and partially disintegrated. The Cerese wax turned from yellow to deep red-brown on the specimens irradiated for a greater flux. The bakelite color changed from red-brown to deep black-violet. The original color of the wax and bakelite remained unchanged for the lower flux irradiated specimens.

The results of irradiation appear to indicate that deterioration of strain-gage bonding cements takes place rather slowly for bakelite cement and

TABLE I

Reactor Radiation Damage to Cements Used for Bonding Wire Resistance Strain Gages to Test Specimens

SPECIMEN	RESISTANCE BEFORE PILE EXPOSURE (ohms)	RESISTANCE AFTER EXPOSURE TO 1.5×10^{17} FAST NEUTRONS/CM^2 (ohms)	RESISTANCE AFTER EXPOSURE TO 2.4×10^{18} FAST NEUTRONS/CM^2 (ohms)
#1 Duco cement	4.6×10^{12}	*6.8×10^{11}	
#2 Duco cement	8.2×10^{11}	*3.0×10^{9}	
#3 Duco Cement and Cerese Wax	1.8×10^{13}	3.0×10^{10}	
#4 Duco Cement and Cerese Wax	7.7×10^{11}	4.0×10^{8}	
#5 Bakelite Cement	$> 5.0 \times 10^{14}$	1.1×10^{12}	
#6 Bakelite Cement	$> 5.0 \times 10^{14}$	6.4×10^{12}	
#7 Bakelite Cement and Cerese Wax	$> 5.0 \times 10^{14}$	8.3×10^{13}	
#8 Bakelite Cement and Cerese Wax	$> 5.0 \times 10^{14}$	4.0×10^{14}	
#9 Duco Cement	6.9×10^{12}		Cement disintegrated
#10 Duco Cement	1.6×10^{12}		Cement disintegrated
#11 Duco Cement and Cerese Wax	8.0×10^{11}		1.1×10^{11}
#12 Duco Cement and Cerese Wax	1.3×10^{12}		6.3×10^{10}
#13 Bakelite Cement	$> 5.0 \times 10^{14}$		5.6×10^{10}
#14 Bakelite Cement	$> 5.0 \times 10^{14}$		1.2×10^{11}
#15 Bakelite Cement and Cerese Wax	$> 5.0 \times 10^{14}$		9.4×10^{11}
#16 Bakelite Cement and Cerese Wax	$> 5.0 \times 10^{14}$		8.0×10^{11}

*After measurement the cement sandwich fell apart.

much more rapidly for Duco cement. The waterproof protective coating of Cerese wax does not seem to influence the numerical results. However, it may serve to hold the bond together in the case of the Duco cement.

Lack of uniformity in the making of test sandwiches of the cement material accounts for the lack of greater similarity among specimen resistance and test results. However, since actual test gages are of the order of 100 ohms it is clearly seen that the magnitude of the insulation resistance of the gage bonding and cement materials is more than sufficiently high to prevent spurious gage readings due to insulation breakdown.

Arrangements are being made to insert in the pile satisfactory SR-4 strain gages cemented on metals that will not change in dimensions due to irradiation. Knowing the strain gage and specimen characteristics one should then be able to determine how strain gages react under irradiation and whether or not they will prove practical for research on the dimensional stability of materials under irradiation.

RADIATION STABILITY OF URANIUM-ALUMINUM ALLOYS FOR MTR[2]

L. C. Templeton, S. E. Dismuke, M. A. Bredig

The work in progress at ORNL on a series of aluminum-clad uranium aluminum alloys (Series III) resembling in important aspects actual MTR fuel elements has been mentioned in previous Quarterly Reports of the ORNL Physics Division. Though not complete, it has progressed to a point where a summary of the results obtained thus far and a comparison with previous data appears desirable in view of the urgency attributed to an early answer to the question of radiation stability of this material.

The effects of Hanford pile irradiations on physical dimensions of strips $9 \times 1 \times 0.1$ cm of alloys containing five, 15, and 17.2 percent uranium, 93.3 percent enriched in U^{235}, were reported by S. Siegel and D. S. Billington[3]

(2) Expanded from paper presented at AEC Metallurgy Conference at Knolls Atomic Power Laboratories, March 16, 1950.

(3) Siegel, S. and Billington, D., "Effect of Reactor Radiation on Metals," *Journal of Metallurgy and and Ceramics*, TID 66, 54 (Jan., 1949).

to be less than one percent, the accuracy of measurement then attempted (Series I). However, changes in thermal and electrical resistivity of 50 and 80 percent, respectively, were observed after an irradiation period of one year. Hardness changed from Rockwell E scale 35 to 102.

A second series of specimens (II), containing five, 15, and 30 percent uranium, 93.3 percent enriched in U^{235}, was initiated by S. Siegel and W. E. Johnson and irradiated at Hanford in 1948 and 1949 for periods of three, six and 12 months. The specimens consist of square rods, 0.1 × 0.1 × 3.0 inches, two of which were held tightly between two half cylinders of aluminum, shrink-fitted, for best-possible heat transfer, into a hollow aluminum slug. This assembly was canned at Hanford in the usual way for exposure in the process channels. Most likely because of variations in heat transfer from one slug to another the results of measurements of electrical resistivity, density and length, carried out by W. E. Johnson, L. C. Templeton and S. E. Dismuke[4] scattered rather badly as shown in Figs. 4, 5 and 6. A closer examination of the data reveals that those pairs of specimens that were contained in four of the eleven slugs tested thus far, namely Nos. 1, 3, 7 and 10, show in all three diagrams radiation effects considerably smaller than the remainder of the specimens for which experimental data lie on comparatively smooth curves. It may therefore be assumed that the temperature inside of these four slugs was somewhat higher than in the other seven, and that the self-healing rate during exposure has been greater for those four pairs of specimens. Curves were therefore not drawn through these points, although they were included in the data presented in the diagrams.

The curves for *electrical resistivity* changes (Fig. 4) tend to flatten out somewhat, but with an integrated slow neutron flux of nvt = 10^{20} cm^{-2} in the adjacent ton of metal (six months) seemed far from having reached a maximum, quite in agreement with the earlier report by S. Siegel and D. S. Billington. On the other hand, it is rather evident that the changes in *volume* (Fig. 5) did not continue, in the 15 and 30 percent alloys, beyond the change reached with an integrated flux of 5 × 10^{19} thermal neutrons per square centimeter in the adjacent ton of metal (three months), of the order of somewhat less than three percent. In the five percent alloy, exhibiting a slower rise in the

(4) Johnson, W. E., *et al.*, "Aluminum Uranium Alloy," *Physics Division Progress Report for the Quarter December, January, February, 1948-49*, ORNL 325, 75 (May 6, 1949).

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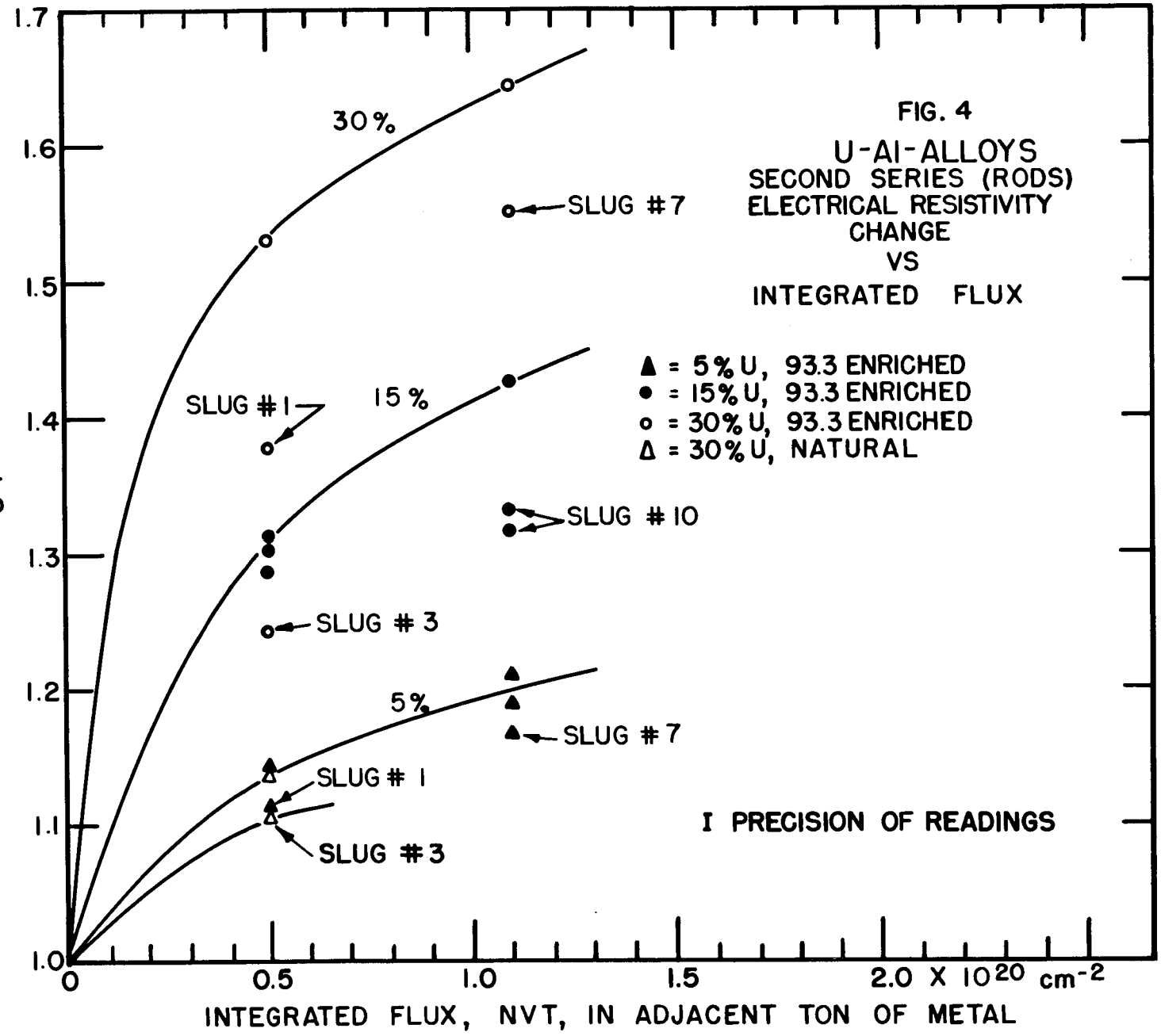

FIG. 4

U-Al-ALLOYS
SECOND SERIES (RODS)
ELECTRICAL RESISTIVITY
CHANGE
VS
INTEGRATED FLUX

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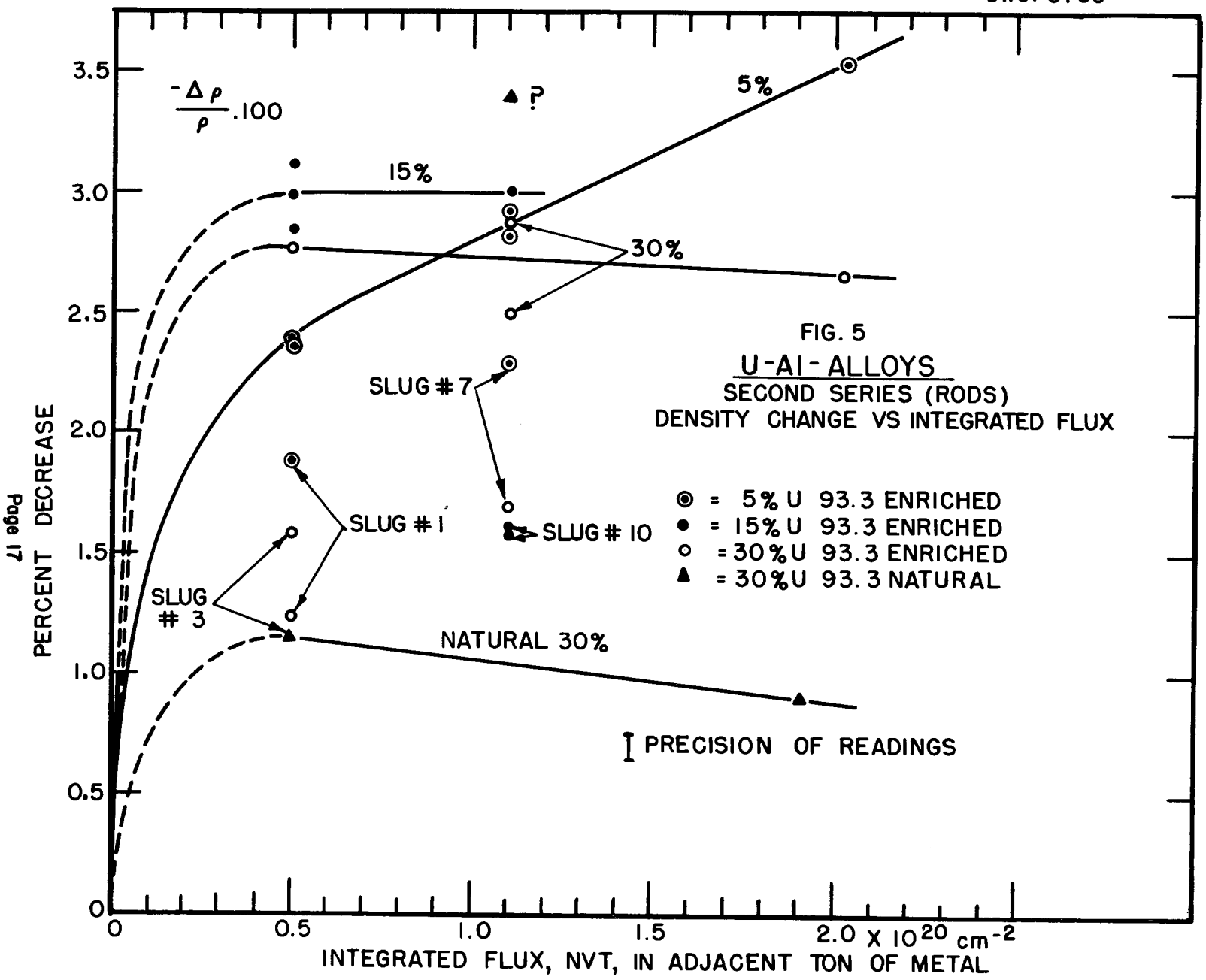

FIG. 5

U-Al-ALLOYS

SECOND SERIES (RODS)

DENSITY CHANGE VS INTEGRATED FLUX

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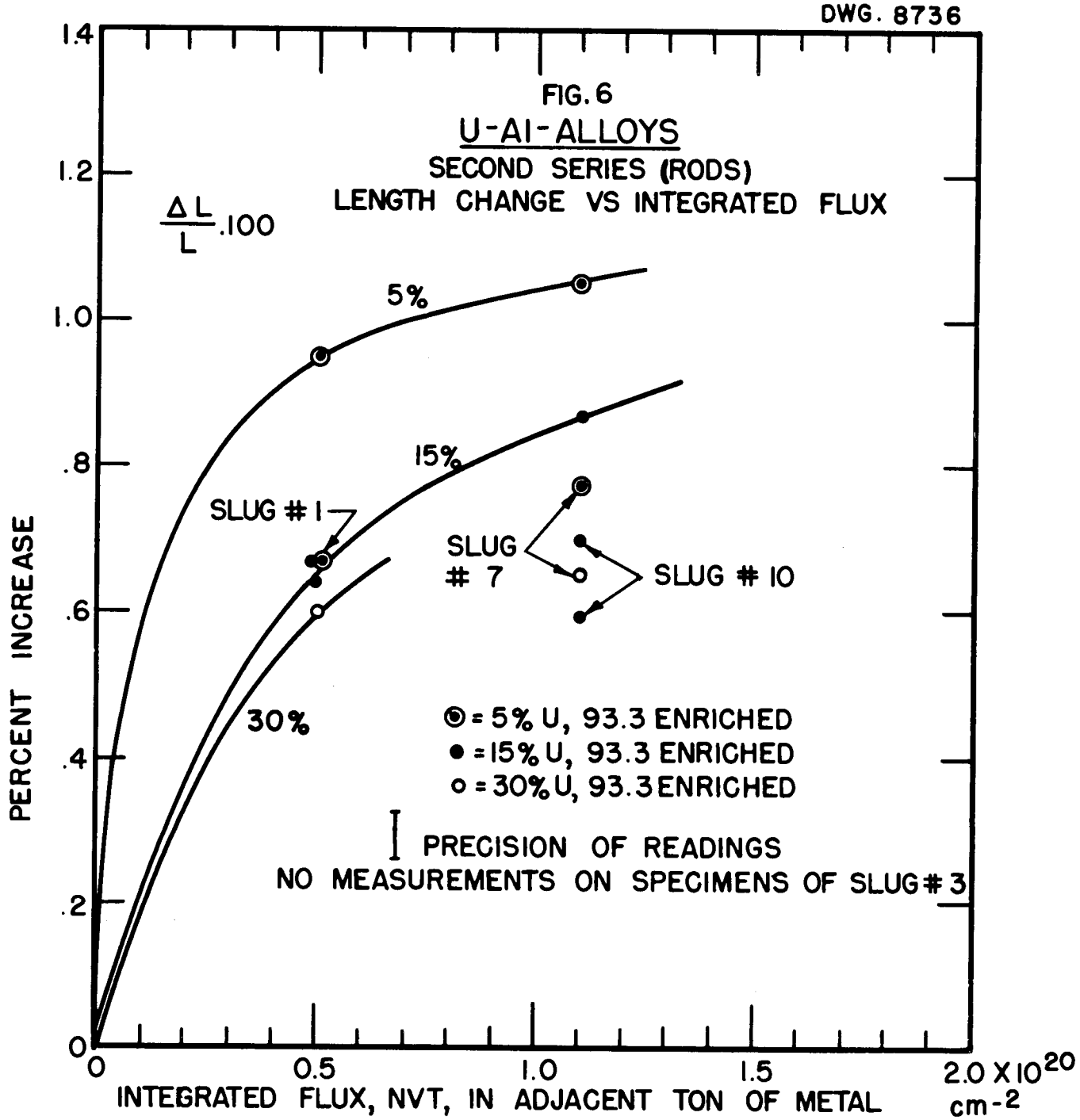

radiation effect, a maximum does not seem to have been reached with even 2×10^{20} neutrons per square centimeters (in the adjacent metal), and a change of 3.54 percent in volume. This observation seems to indicate the need for further examination of alloys of low uranium content. The behavior of the 15 and 30 percent alloys in exposures to less than 5×10^{19} n/cm^2 also deserves further attention.

It may be of interest to note the relative position of the curve for the length change (Fig. 6) in the five percent alloy with respect to those of the 15 and 30 percent alloys. If compared with Fig. 5, representing the density changes, it appears that in the 15 and 30 percent alloys the growth of almost three percent in volume in the first three months occurred to a somewhat greater extent in the directions, or in one direction, perpendicular to the long dimension, probably in the width. Width and depth could not be measured with sufficient accuracy to permit directly checking this observation. When plotted against the fissioned fraction of total atoms in the alloy (U+Al) this behavior becomes even more evident (Figs. 8 and 9). (In these diagrams, as well as in the corresponding one of Fig. 7, the specimens of slugs Nos. 1, 3, 7 and 10, giving the spurious effects, were not included for the sake of clarity.) It seems possible that this effect is connected with the inhomogeneity of the slow neutron flux within the specimens, *i.e.*, its concentration at the surfaces of the specimens.

The final volume change is found to be slightly smaller in the 30 percent alloy than in the 15 (and the five) percent alloys (Fig. 5). Siegel and Billington have reported for the electrical resistivity a damage rate, per fissioned fraction of total atoms, considerably higher in the 15 percent alloy than in the 5 percent alloy, containing aluminum in excess of the eutectic. This was attributed to the influence of a maximum degree of dispersion of fissioning uranium atoms throughout the aluminum matrix in the eutectic composition. In the electric resistivity measurements of the second series reported here (Fig. 7), this effect is much less noticeable. It is significant that the absolute values of resistivity change (*e.g.* $R_f/R_o = 1.20$ for $f = 1 \times 10^{-3}$) check the former data very well in the five percent alloys. The lower damage rate in the 15 percent alloys of the second series as compared with the first one is most probably again connected with the decreased cooling efficiency in the second series (rods, as against thin plates), which is naturally of increasing importance in the specimens containing the higher

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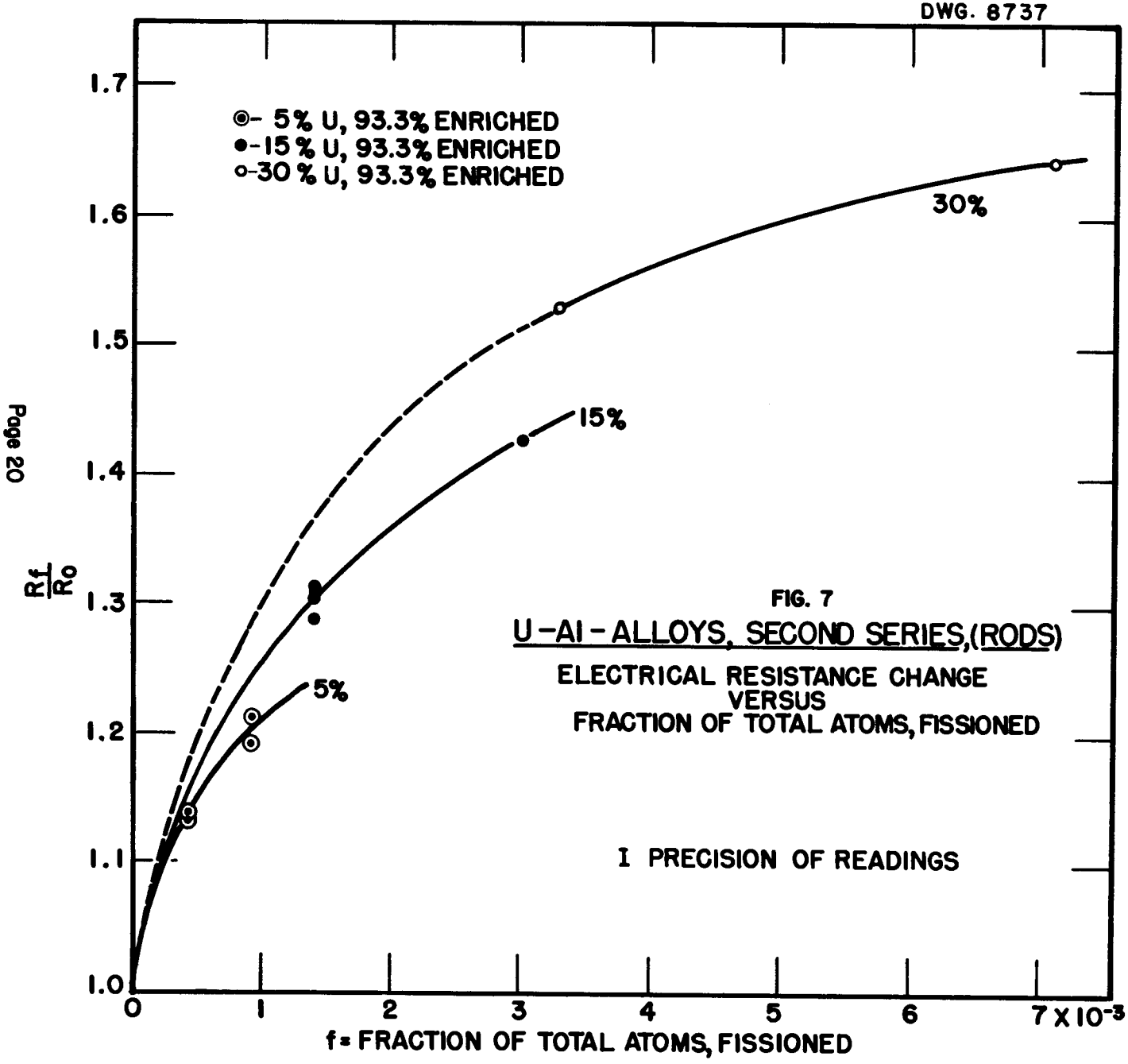

FIG. 7

U-Al-ALLOYS, SECOND SERIES, (RODS)

ELECTRICAL RESISTANCE CHANGE VERSUS FRACTION OF TOTAL ATOMS, FISSIONED

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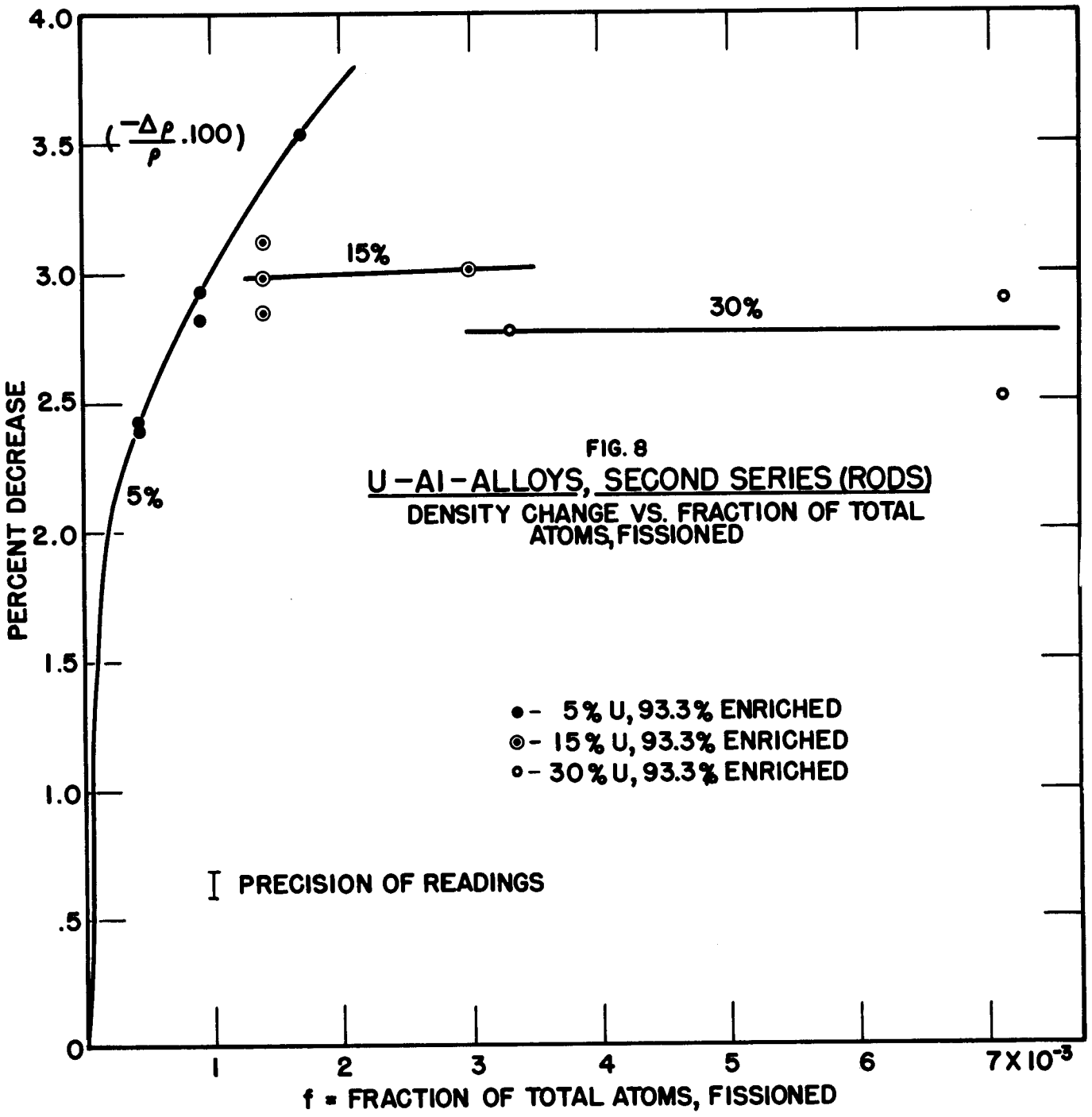

FIG. 8

U-Al-ALLOYS, SECOND SERIES (RODS)

DENSITY CHANGE VS. FRACTION OF TOTAL ATOMS, FISSIONED

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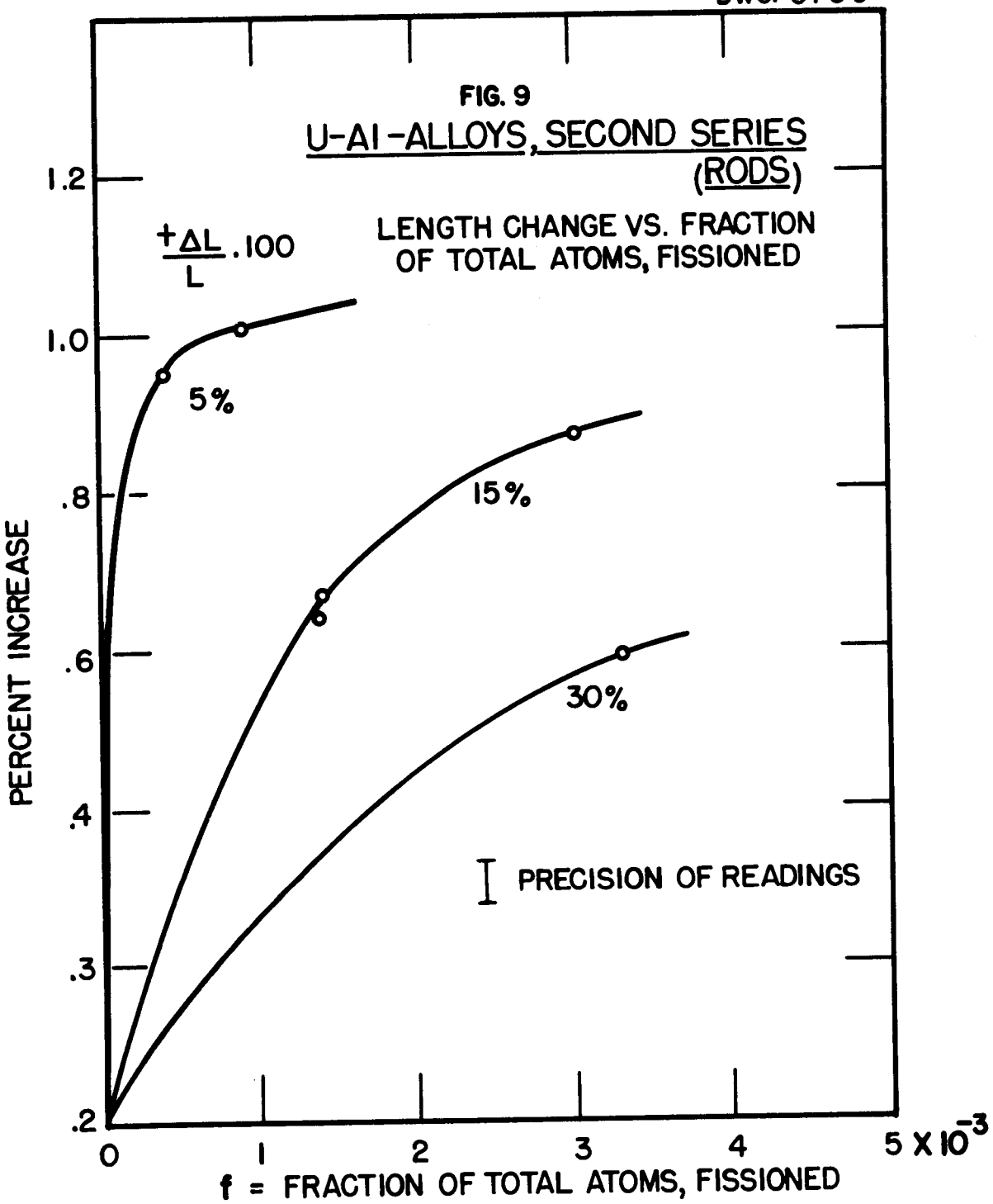

uranium contents. We are presently checking whether microstructure, and particularly an observation reported previously,[5] *i.e.* the occurrence of the phase UAl_3 in considerable amounts in these specimens, in disagreement with the established binary phase diagram, has any significance in this connection. It might indicate the formation of the eutectic to occur under conditions different from those expected from the binary phase diagram. R. Williams of the ORNL Metallurgy Division has recently discovered a strong effect of silicon upon the relative amounts of UAl_3 and UAl_4 present in these alloys.

As soon as practical the data of Figs. 4 to 9 will be supplemented by the results of further measurements especially on the 12 months samples, now in progress. These will also include Tensil tests.

The new, third, series of alloys, initiated by M. M. Mann, and consisting of an alloy core (approx. 3.5 × 0.55 × 0.02 inches) "sandwiched" between a cladding of two layers of 2S and 72S aluminum, of overall dimensions 4.0 × 0.84 × 0.064 inches, was prepared by C. D. Smith, F. Drosten and F. Kerze, ORNL Metallurgy Division.[6] It is expected to be essentially free from the disturbing heating effects encountered in the second series, because of more effective cooling. Figures 10 and 11 show the changes in density as measured thus far on the specimens in the "as received" condition, after periods of 43, 95 and 157 days of exposure in the Hanford process channels. Again considerable scattering of the experimental points may be noted. This scattering is almost fully explained by the observation that specimens showing various amounts of a white corrosion product (*E* for extensive, *M* for moderate, and *S* for slight scale formation, in Fig. 10 are those that have abnormally low densities, probably because of air trapped between the deposit and the metal surface. By measuring the density after removal of the corrosion scale from one specimen a value was obtained that fits rather smoothly into the curve drawn through the experimental points for the corrosion-free specimens. It is significant that corrosion was observed only in those specimens which in a brazing cycle had been treated with a fluoride flux. Since there seems to be no connection between the length of the irradiation period and the amount of corrosion, it may be assumed that this corrosion took place, not in the pile, but during storage in stagnant water at both Hanford and Oak Ridge.

(5) Bredig, M. A. and Sherrill, F. A., "X-Ray Diffraction Examination," *Physics Division Quarterly Report Ending June 25, 1949*, ORNL 365, 12 (July 27, 1949).

(6) *Reactor Technology Progress Report for Quarter Ending May 31, 1949*, ORNL 267 (July 18, 1949). Kerze, F., *Technical Division, Engineering Materials Section 4 Progress Report for Period February 15,- April 1, 1949*, ORNL CF 49-5-40 (Apr. 27, 1949).

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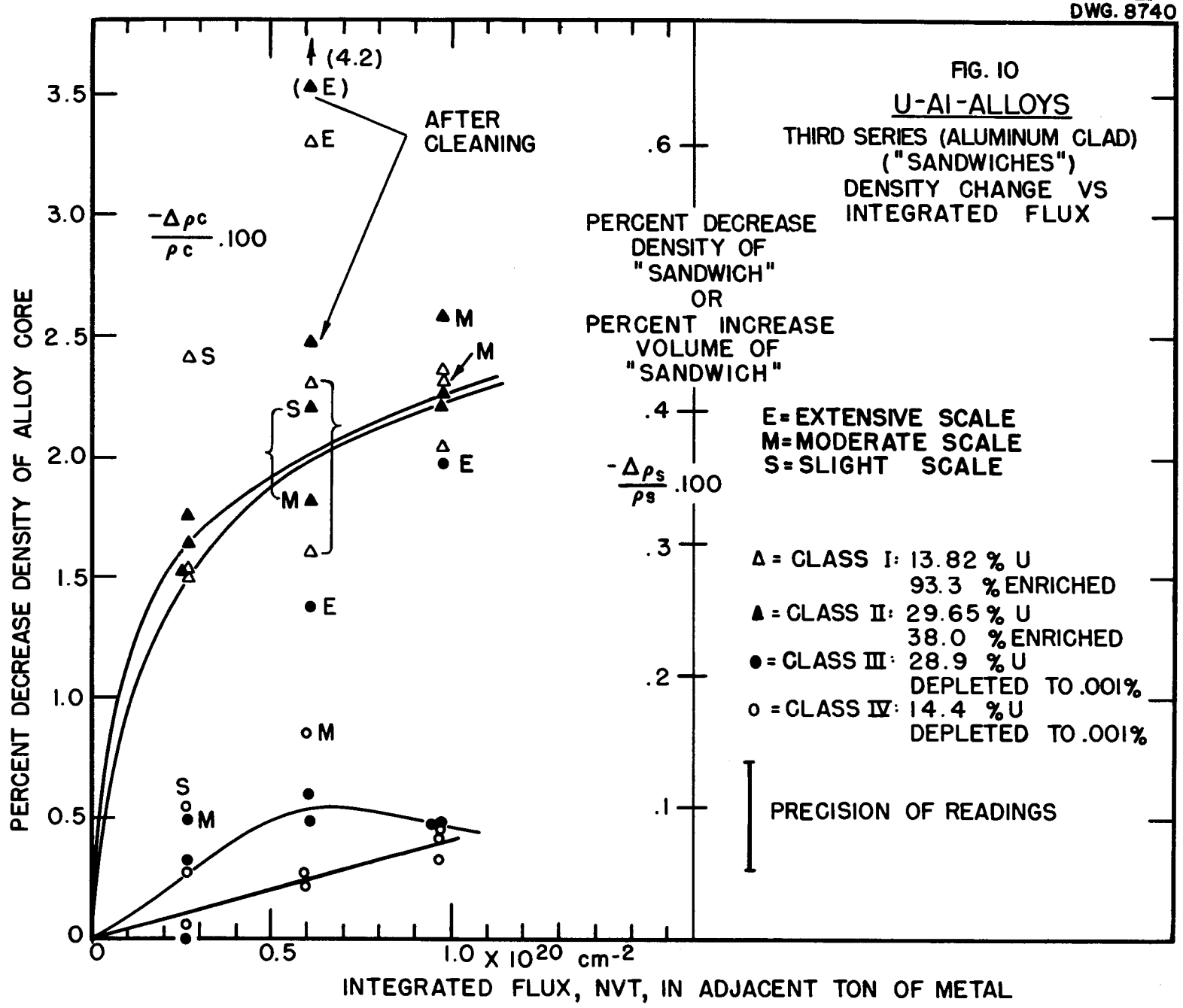

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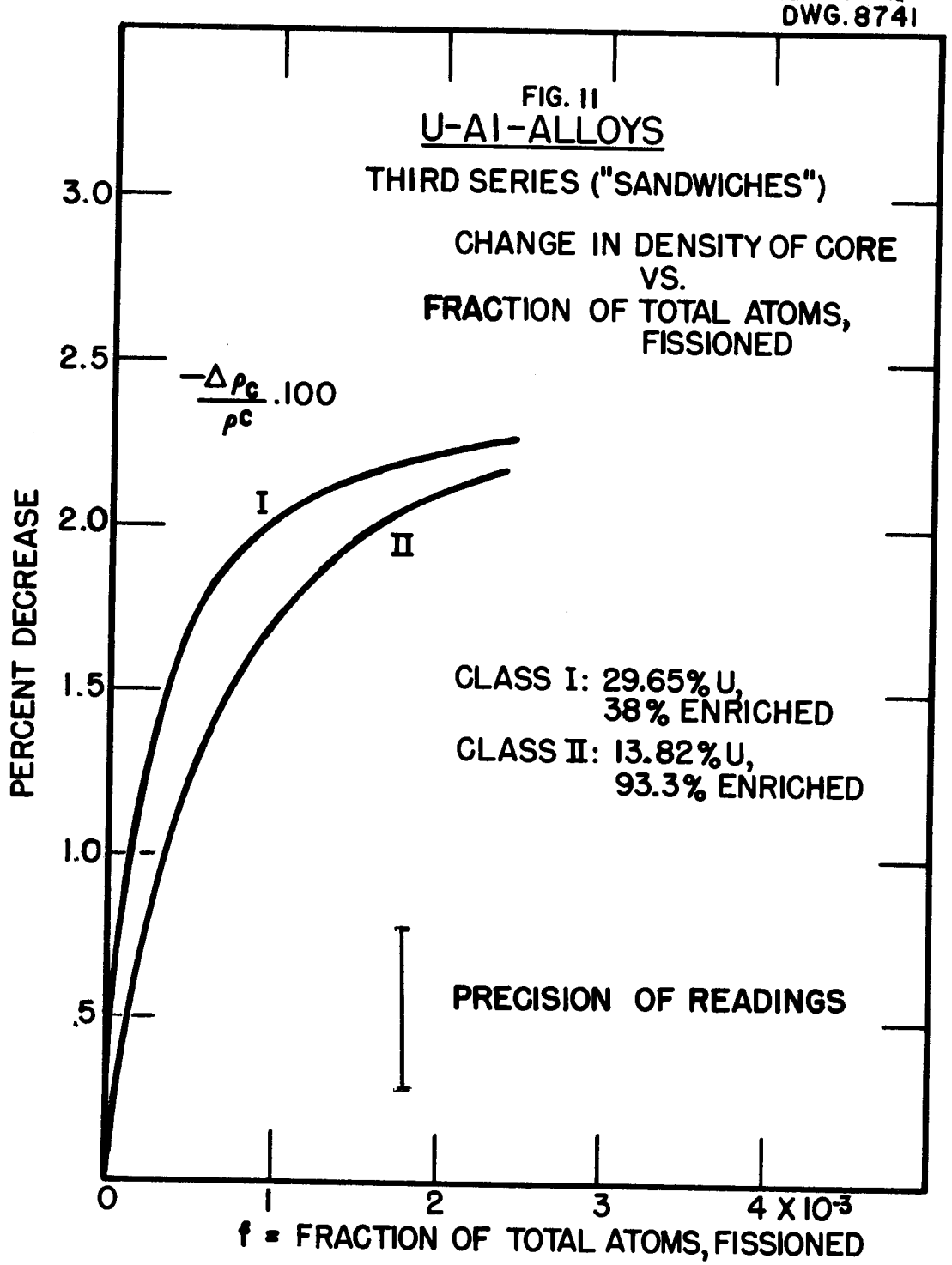

Weight changes were of the order of ±0.1 percent (10 milligrams) or less, except in one case, where it amounted to -0.3 percent. Changes in length and width were considerably less than 0.1 percent, the probable errors being of the order of ±0.03 percent and ±0.1 percent, respectively. Thickness changes ranged from -1.5 to +2.0 percent, but were mostly positive, the probable error being ±1.5 percent. It appears, then, that the volume growth of approx. 0.4 percent (Fig. 10) occurred largely in the thickness of the specimen, but was too small to be measured directly in this dimension. The density changes in the depleted alloys were much smaller, and of the order of the degree of accuracy of the measurements.

One of the two density scales in Fig. 10 and Fig. 11 refers to the "sandwich" as a whole, measured directly, whereas the other scale for the density changes in the alloy core was obtained by multiplying the former by a factor 5.5 ± .2, *i.e.*, the ratio of the total volume of the "sandwich" to that of the core. This conversion is based on the assumption that no radiation effects were produced in the aluminum cladding, and that no or little diffusion of fission fragments into the cladding took place.

The most significant result of the measurements of density and dimensions of the third series of specimens is, then, the qualitative confirmation of the results of the second series at least with respect to the order of magnitude of the volume increase. With an integrated flux of 10^{20} neutrons per square centimeter in the adjacent metal, 2.3 percent in the sandwich type specimens (Fig.10) seem to represent not quite a maximum effect, whereas 3.0 percent in the rod specimens of the second series do seem to represent a maximum value of damage in the 15 percent alloys. An explanation for the remaining difference in the rate of damage, and particularly for the lower rate in the presumably cooler specimens of Series III is not immediately apparent. It may be significant, however, that in the five percent alloys of the second series a maximum effect does not seem to have been obtained even in a volume increase of 3.5% with 2×10^{20} neutrons per square centimeter in the adjacent metal. The same seems to be true for another series of measurements reported in a memo[7] by E. C. Miller on 4.25% U-Al alloy, W-type, 4 in. slugs. An increase in diameter from 1.436 ± 0.003 inches to 1.443 ± 0.003 inches in 93.8 days, and

(7) Miller, E. C., *Dimensional Changes Resulting from Pile Irradiation of Aluminum Enriched Uranium Alloys*, ORNL CF-49-11-216, (Nov. 22, 1949).

to 1.447 ± 0.003 inches in 152 days, at Hanford, was reported there, corresponding to volume increases of 1.5 and 2.3 percent, respectively, if isotropic expansion is assumed. It thus appears as though not only low uranium 235 content, but also efficient cooling tends to delay the saturation of the radiation effect.

THERMAL CONDUCTIVITY OF URANIUM-BEARING MATERIALS*

R. G. Berggren, J. M. Kincaid, W. Primak**

A number of thermal conductivity samples are currently being irradiated in fuel channels in the X-10 pile and additional ones are currently being prepared. These are described:

(a) The *low temperature graphite* sample[8] described previously[9] is still being irradiated. The behavior of this sample to date is shown in Fig. 12. The peculiar shape of the ΔT curves is interpreted as the result of annealing as the temperature distribution in the sample was altered by the changing thermal conductivity. The central thermocouple failed during this period after five and one-half months of operation. Another sample is currently being prepared for irradiation.

(b) The irradiation of the *high temperature graphite* sample[10] described previously[11] was continued through this period. The behavior of this sample is shown in Fig. 13. The damage rate of this sample cannot be directly compared with those of previous samples because the neutron flux in this hole is probably different from what it was for previous samples.[12] However, the damage rate seems to be rather low, comparable to the previous samples which were pitch-impregnated following UNH impregnation. These samples have been termed "high density" samples.

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* Sponsored by Argonne National Laboratory.

** Consultant, Argonne National Laboratory.

(8) The terms "high temperature" and "low temperature" graphite samples refer to the apparatus in which the irradiations are performed. The apparatus is shown in Figs. 6-7, ORNL 171, *Power Pile Division Quarterly Report for June, July, August, 1948*, p. 27-28 (Nov. 9, 1948), and in Fig. 2, MonN 442, *Progress on High Temperature Thermal Conductivity Measurements* by L. P. Hunter, p. 7b (Dec. 17, 1947). See also ORNL 576, "Thermal Conductivity of Uranium-Bearing Materials" by W. Primak *et al.*, *Physics Division Quarterly Progress Report for Period Ending December 15, 1949*, p. 12, section b (Feb. 7, 1950).

(9) Primak, W., *et al.*, "Thermal Conductivity of Uranium-Bearing Materials," *Physics Division Quarterly Progress Report for Period Ending December 15, 1949*, ORNL 576, 12-15 (Feb. 7, 1950).

(10) See Ref. (8).

(11) Primak, W. *et al.*, *op. cit.*

(12) *Ibid.*

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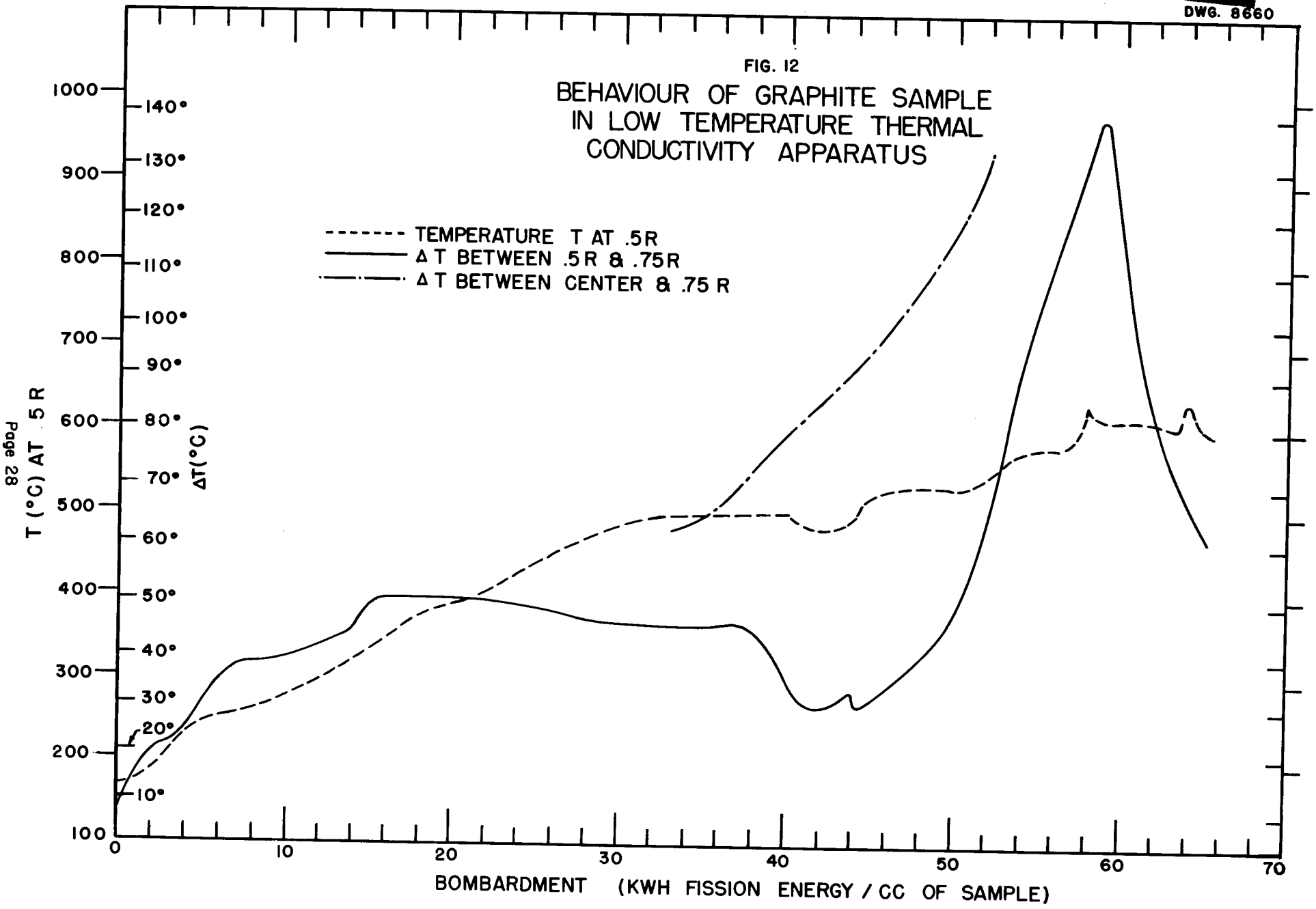
DWG. 8660

FIG. 12

BEHAVIOUR OF GRAPHITE SAMPLE IN LOW TEMPERATURE THERMAL CONDUCTIVITY APPARATUS



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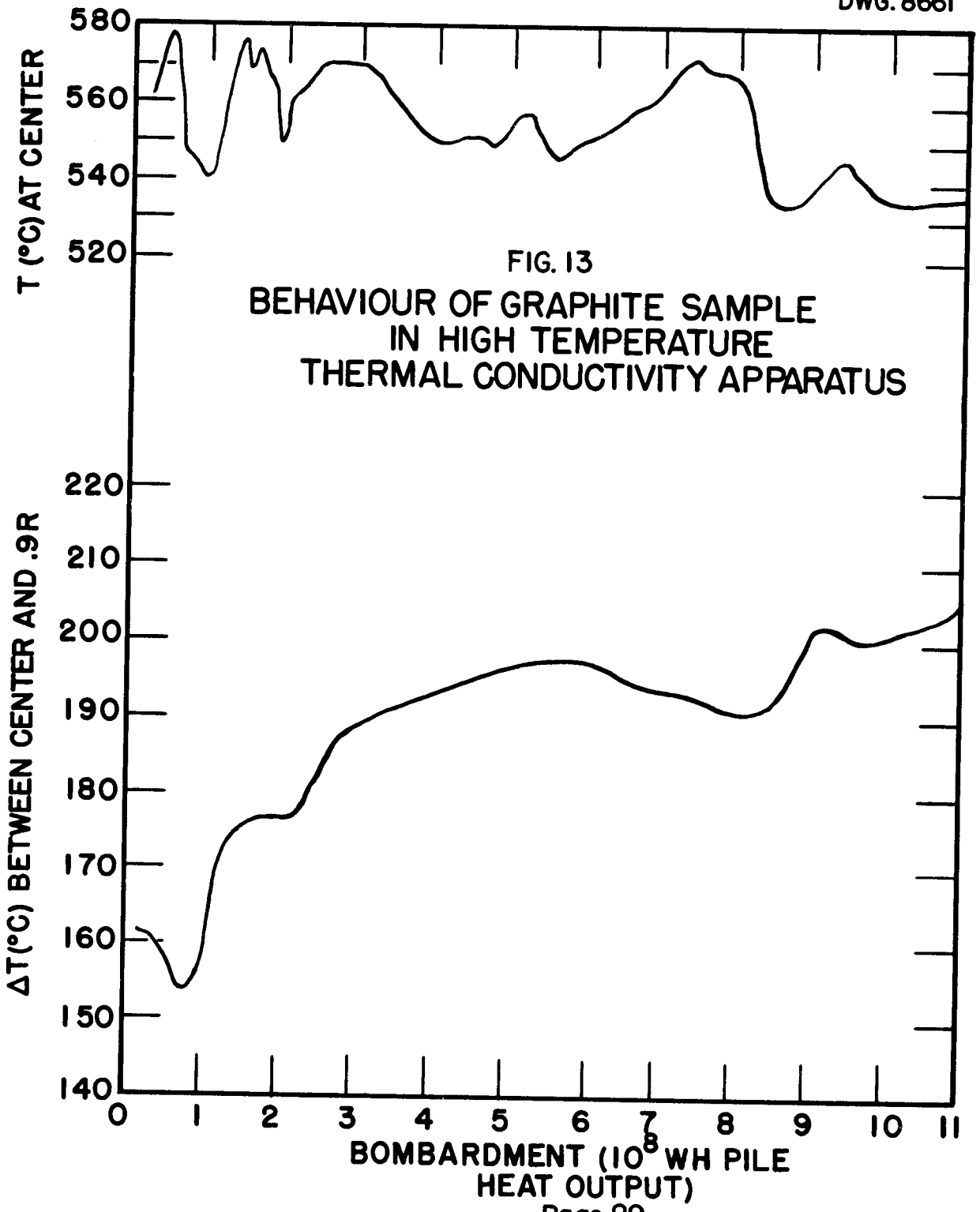

FIG. 13

BEHAVIOUR OF GRAPHITE SAMPLE IN HIGH TEMPERATURE THERMAL CONDUCTIVITY APPARATUS

(c) The irradiation of the *beryllium-uranium alloy* sample previously described[13] was continued through the present period. There are some indications of a small increase in thermal conductivity during the first month of the irradiation. Another sample is currently being prepared for insertion into the pile, and this behavior will be checked. The thermal conductivity of the second sample has been checked and is reported below. These BeU samples were prepared by A. R. Kaufman (MIT) by methods of powder metallurgy and contained 20% U (93.14% enriched). An X-ray radiograph[14,15] showed the uranium was present in small grains and uniformly dispersed.

(d) A sample of *zirconium-uranium alloy* is currently being prepared for irradiation in the X-10 pile. The alloy was made at the Battelle Memorial Institute and contains 5.92 atomic percent U (93.17% enriched).[16] The thermal conductivity of this sample has been measured and is reported below.

(e) A *carbon sample* is currently being prepared for irradiation in the X-10 pile. This material was obtained from the National Carbon Company and is described by them as follows: ".....SA-45, apparent density 1.53 g/cm^3. These blocks are molded from a lampblack pitch mixture, with the molding direction perpendicular to the largest face. They are nearly isotropic in the three directions, as contrasted to material made from graphite or petroleum coke." [17] The density of the sample which is being prepared for irradiation was checked and found to be 1.47 g/cm^3. Its thermal conductivity was checked by the method previously described[18] and found to be .04 - .06 cal/cm^2(°C/cm) sec. The sample was examined by X-ray diffraction (M. A. Bredig) and gave a typical carbon pattern.[19] A drying procedure[20] has to be developed which

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(13) Primak, W. *et al.*, *op.cit.*

(14) Hunter, L. P., *Progress on High Temperature Thermal Conductivity Measurements*, MonN-442, App. 1 (Dec. 17, 1947), and subsequent reports.

(15) Hunter, L. P., "Thermal Conductivity of Uranium-bearing Material Under Irradiation at High Temperatures," *Journal of Metallurgy and Ceramics*, TID 66, 41 (Jan., 1949).

(16) Analysis by B.M.I., private communication.

(17) H. S. Patten (NCC), private communication.

(18) Morgan, J. G., "Thermal Conductivity of Graphite*," *Physics Division Quarterly Report Ending June 25, 1949*, ORNL 365, 17 (July 27, 1949).

(19) Biscoe, J. and Warren, B. E., "An X-ray Study of Carbon Black," *App. Phys. J.* 13, 364-371 (1942).

(20) Hunter, L. P., *op. cit.*, MonN 442.

would give a uniformly impregnated sample with SA-45 carbon. A satisfactory sample was obtained with the following procedure: The sample was placed in a furnace connected to a variable auto-transformer and the muffle temperature raised uniformly from 80°C to 200°C at a rate of 1°C/min by manually controlling the transformer. The resultant sample contained 3.65 g uranium oxide in 23 g of carbon after firing at 800°C.

Difficulties encountered in the use of the thermal conductivity apparatus previously described[21] led to the development of a new apparatus. It is shown in Fig. 14. In this apparatus the thermal conductivity of the unknown sample (A) is determined relative to a known sample (B). The thermal conductivity calculated for different heat inputs was plotted against the temperature drop across one of the samples and extrapolated to zero drop. The results seem good to ±5%. This is as good as can be expected with the present thermocouple arrangement and the present cooling arrangement. Using a nickel bar as a standard (assumed k = 0.142 cal/cm^2(°C/cm) sec, a value k = 0.26 was obtained for BeU alloy [see (c) above] and 0.29 for ZrU alloy [see (d) above]. The latter result may be compared with a result, 0.30, indicated by data obtained at Battelle Memorial Institute.[22]

IRRADIATION CREEP EQUIPMENT*

W. E. Brundage and W. Primak**

The creep equipment previously built and tested[23] was designed to obtain engineering creep data under irradiation in a pile. From the results reported[24] and a recent summary,[25] it seems likely the apparatus developed could not yield creep data of better than 50 - 100% precision. A very large change in creep rate would therefore be required before it could be observed in this apparatus. Tests thus far made on metals in present piles have not

(21) Morgan, J. G., *op. cit.*

(22) Private communication.

(23) Brundage, W. E. and Primak, W., "Irradiation Creep Experiment," *Physics Division Quarterly Progress Report for Period Ending December 15, 1949*, ORNL 576, 8-12 (Feb. 7, 1950).

(24) *Ibid.*

(25) Sully, A. H., *Metallic Creep and Creep Resistant Alloys*, New York: Interscience Publishers, Inc. (1949).

* Sponsored by Argonne National Laboratory.

** Consultant, Argonne National Laboratory.

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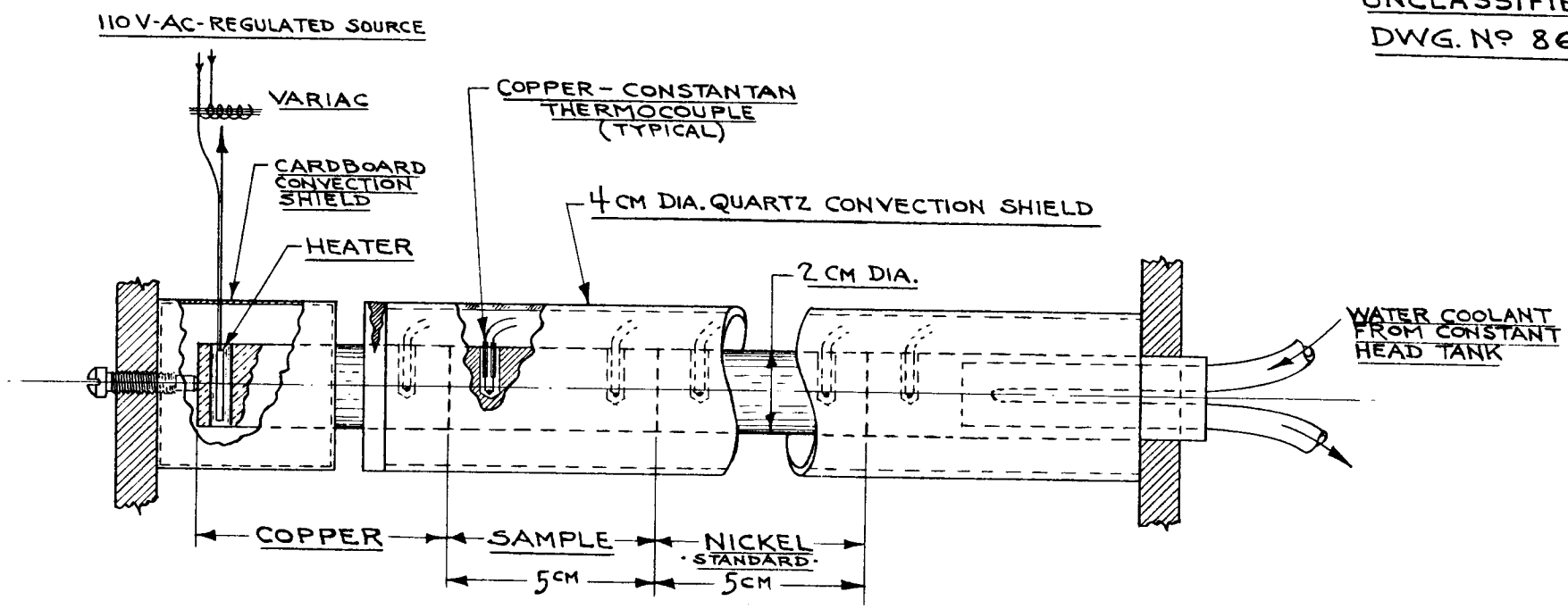

FIGURE Nº 14
THERMAL CONDUCTIVITY
APPARATUS

indicated that any such change might be expected.[26] It is important to look for a change, however small, which might be proportional to the flux since it is planned to build piles with 100 - 1000 fold increase over present pile fluxes. In these tests, an increase in precision of measurement could greatly exceed in value a slight enhancement in fast flux by a neutron converter. The fission heaters previously used which prevented satisfactory temperature control were therefore abandoned. To increase the precision to the highest possible degree it was decided to use the same specimen to determine the normal creep rate and the irradiation creep rate. In the initial experiments the attempt would be made to make the comparisons in the second stage of creep where the creep rates could be more easily established. Although this is the region in which previous experiments indicated a minimum of effect,[27] it is the region of major engineering concern. If the comparisons in this region are successful, an attempt will be made to extend them into the earlier stages of creep. To attain the desired precision in the creep rate measurements, the temperature and loading would have to be controlled with the highest possible precision. It would also be desirable to perform the experiment in a pile having the highest possible flux. A facility ideally suited to the experiment has been found in the Hanford pile and the apparatus is being designed for use in this facility.

Tests on the sylphon bellows used in previous designs indicated that with their use the loading on the specimen might change by several percent over the range of creep of a specimen because of the spring force of the bellows. The effect of extension and sustained load on the mean effective area of the bellows has not yet been determined, and would have to be checked before using them for any precise experiment. It was therefore decided to discard the bellows and use dead weight loading of the specimen. It was estimated that frictional losses would not be more than several ounces in several hundred pounds of load, thus keeping the load constant to a fraction of a percent.

Various methods of temperature control were considered. The long distance from the experiment to the control instruments (over 100 ft) and the exposure

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(26) Siegel, S. and Billington, D., "Effect of Reactor Radiation on Metals," *Journal of Metallurgy and Ceramics*, TID 66, 67 (Jan., 1949).

(27) Andrade, E., "Effect of Alpha-Ray Bombardment on Glide in Metal Single Crystals," *Nature* 156, 113 (1945).

in the pile precluded the use of electrical resistance thermometers. It was therefore decided to attempt to control the temperature by means of thermocouples. It would seem that control beyond one microvolt would be impractical. This would be a little less than 0.1 degree for usual thermocouple materials. It is the limit attained in usual potentiometric measurements. It is beyond the usable range of commercial controlling devices. It was therefore decided to build a controller which was operated by the reflected beam of the potentiometer galvanometer. In the first device tried, the galvanometer beam was permitted to fall on a photo cell. The output was connected to a vacuum tube relay. Considerable difficulty was encountered in obtaining enough light to operate the relay. Satisfactory operation could be obtained only when a small fraction of the total power input was controlled. This was a very unsatisfactory arrangement since thermal losses might alter considerably during the operation of the equipment. A proportional controlling device based on the circuits described by Hull[28] and Waring and Robison[29] was then tried. With this device, it was found possible to keep the temperature of the control couple to within several thousandths of a degree during an initial test. The modified circuit which was found most satisfactory for the thyratron control is shown in Fig. 15. It is a slight simplification of the Waring and Robison circuit and is very much easier to adjust. It was found to operate satisfactorily with a number of phototubes, but vacuum phototubes were found to be best. Only a very weak galvanometer lamp was necessary. This circuit has the advantage over Hull's circuit that a special transformer is not required. Keeping the potentiometers adjusted for long periods of time remains a major problem. Tests have been started to determine how well this can be done. It is planned to keep the batteries and potentiometers in a regulated temperature box maintained at ±2°C and the cold junctions in a regulated oil bath maintained at ±.05°C. It is planned to adjust the potentiometers against a standard cell daily. It is also planned to heat the specimen by three independently regulated heaters, one on each end of the specimen and one over the gauge length. The temperature along the specimen is being determined in tests currently in progress. Tests will be made to determine how constant the arrangement will keep the temperature over a period of weeks.

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(28) Hull, A. W., "Hot Cathode Thyratrons. Part II. Operation," *Gen. Elec. Rev.* 32, 394 (1929).

(29) Waring, C. E. and Robison, G., "Experimental Details for a Precision High Temperature Control Utilizing the Hull Circuit," *Rev. Sci. Instruments* 14, 143 (1943).

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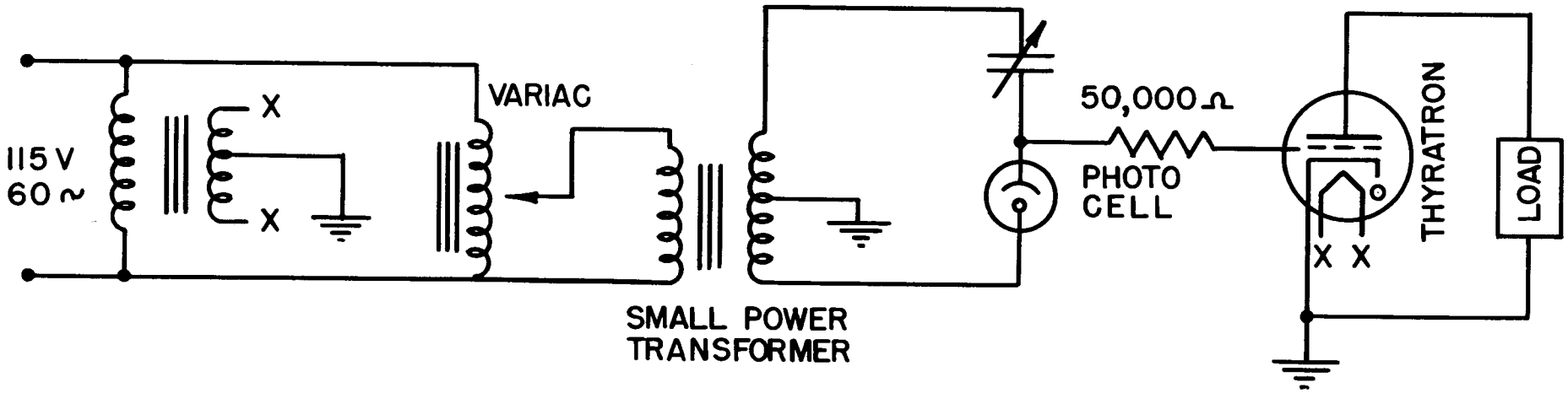

FIG. 15

CREEP UNIT HEATER CONTROL CIRCUIT

It is hoped that the equipment will permit the detection of an alteration of 1% in creep rate caused by radiation.[30]

(30) *Acknowledgement:* We are indebted to K. Smith, S. H. Paine (ANL), A. G. H. Anderson, D. S. Billington (ORNL), R. E. Nather, A. Carson, J. Lambert, and Snoebeger (HW) for suggestions and device concerning design and procedure; we are also indebted to F. Beyer (ANL) for design work and supervision of construction of samples, test rigs and creep equipment, and to C. Stilson (NEPA) for advice, assistance, constructing and testing of the temperature control equipment.