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MAGNETIC DETERMINATION OF IMPURITIES
IN URANIUM FUEL SLUGS

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MAGNETIC DETERMINATION OF IMPURITIES IN

URANIUM FUEL SLUGS

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ABSTRACT

A technique has been devised for the non-destructive determination of the uranium hydride and/or iron content in uranium fuel slugs of Hanford size. The technique, an adaptation of the Gouy method for measuring magnetic susceptibilities, is based on the ferromagnetic properties of the hydride below 173°K. A large electromagnet and a pan balance are utilized in measuring the magnetic forces on a vertically suspended slug at liquid nitrogen temperatures and at room temperature. It was found possible to test as many as 250 slugs per eight-hour day in this manner

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I. INTRODUCTION

The Sylvania method for producing experimental uranium slugs for the Hanford pile included the hydriding of uranium in the powder making procedure. Chemical analysis of several experimental uranium slugs had revealed the presence of hydrogen, presumed to be hydride, in small random amounts. This hydride was found concentrated toward the center of the slug as a coring. It remained if insufficient time was allowed for decomposition of the hydride after heating and before pressing, since the operation sealed the hydride core into the center. This was a matter of concern because a decomposition of the hydride at high temperature or flux might lead to the release of the gas, resulting in the formation of microcracks in the slugs. Since uranium hydride has been shown to be ferromagnetic at temperatures below 173°K , a magnetic test was envisioned as a nondestructive means of determining the hydride content. The use of a magnetic test suggested the possibility of concurrently testing for iron, an undesirable contaminant. This report deals with the technique derived to determine the uranium hydride and iron content.

II. FOUR METHODS

It was apparent that iron content could be determined in a slug by measuring a magnetic property of the slug at room temperature. The uranium hydride content could be determined by the change in the same property on cooling the slug to below the hydride Curie temperature, 173°K . Four methods were considered and partially investigated. These were based on the following:

- A. The inductance of a coil is proportional to the permeability of the material it surrounds.
- B. The electromotive force, as detected by a ballistic galvanometer, developed in a coil by changing an applied magnetic field also depends on the permeability of the material in the coil.

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- C. The torque exerted on a bar of magnetic material in a uniform field depends on the shape and the susceptibility of the material.
- D. The force exerted on a bar of magnetic material in a non-uniform field depends on the distribution and the susceptibility of the material.

It is necessary at this point to evaluate the effects to be expected. The susceptibility of a mixture is obtained from

$$k_m = k_1 f_1 + k_2 f_2 + \dots$$

where k_1 represents the susceptibility and f_1 the fraction of component 1 present in the mixture. Tabulated below are the susceptibilities of interest at various temperatures.

	<u>300°K</u>	<u>190°K</u>	<u>90°K</u>
U	1.67×10^{-6}	1.68×10^{-6}	1.75×10^{-6}
UH ₃	25×10^{-6}	180×10^{-6}	1
Fe	200	200	200

From these, it is apparent that the presence of one part of iron in one million parts of uranium (concentrations of this order of magnitude will be of interest) causes a 100-fold change in the susceptibility at any temperature. However, the effect of the same concentration of uranium hydride will not become apparent till liquid air temperatures are reached. The exact magnitude of the hydride effect cannot be estimated, but concentrations of the order of a part per million will be discernible in the k_m value. The same can not be said of the permeability, which is defined as

$$\mu = 1 + 4 \pi k$$

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Since the average susceptibility will never be much over 10^{-3} , techniques which measure permeability will not be sensitive. A change of an order of magnitude in concentration will cause only a very small change in permeability.

On the basis of this reasoning, A and B were judged to be too insensitive. A short investigation of method C indicated it to be unsatisfactory, since the shape factor, which is unknown for the hydride core, makes the technique inaccurate. Method D was therefore adopted.

III. EXPERIMENTAL PROCEDURE AND APPARATUS

The technique used was an adaptation of a method of Gouy (1,2,3) for the determination of the susceptibility of para- or diamagnetic materials. The Appendix contains an abbreviated derivation of the forces acting in the Gouy method.

Fig. 1 is a schematic representation of the technique. The slug to be analyzed is suspended from a balance slightly above the pole pieces of an electromagnet. The force necessary to counterbalance the additional pull found on applying a non-uniform magnetic field is given by equation (4) of the Appendix

$$F = A (k_m - k_a) H^2$$

where k_m is the average susceptibility of the slug

k_a is the average susceptibility of air

H is the magnetic field at the bottom of the slug

A is a constant of proportionality

and where

k_m is the variable which is evaluated in terms of the composition of the slug.

This can be rewritten as:

$$F = A (f_u k_u + f_{Fe} k_{Fe} + f_{hydride} k_{hydride} - k_a) H^2$$

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At room temperature k_{hydride} is small enough so that $(f_{\text{hydride}} k_{\text{hydride}})$ can be neglected. The force exerted by the field is thus equal to a constant plus a factor proportional to iron content.

At liquid air temperature, k_{hydride} becomes very large, so that $(f_{\text{hydride}} k_{\text{hydride}})$ becomes important and the force is equal to the room temperature value plus a factor proportional to hydride content*.

The experimental set-up is indicated in Figs. 1, 2 and 3. The slugs, all 4.00" long and 1.35" in diameter, were suspended between the pole pieces of a magnet and the downward forces were obtained using an ordinary pan balance.

An Arthur D. Little electromagnet, with pole pieces tapered to a one-inch diameter, was used. The spacing between the pole pieces was chosen as four inches, this spacing allowing for ease in handling the suspended slugs.

The vertically measured distribution of field strength, midway between the pole pieces, is shown in Fig. 4.

The basket for holding the slugs had to be non-magnetic, had to be subjected to extremely low temperatures, and had to support the 2-kilogram weight of each slug. All available metals (e.g. copper, brass) were rejected inasmuch as their high electrical conductivities would tend to prevent any motion in high magnetic fields because of eddy current effects.

*Strictly speaking, since the uranium hydride is not uniformly distributed, the above arguments are not quite true. If, however, one assumes the hydride core is always in the same region in the slug, in which regions the magnetic field is H' , the following equation can be written and an argument similar to the one above followed:

$$F = A (f_u k_u + f_{\text{Fe}} - k_1) H^2 + A' (f_{\text{hydride}} k_{\text{hydride}} - k_1) H'^2$$

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The design which was finally evolved for the basket (Fig. 3), used lucite, allowed each slug to be slipped in and out rather easily, and gave a high degree of reproducibility.

The lucite basket was suspended from a rigid brass rod (effectively out of the magnetic field) leading to a bronze turnbuckle which could be adjusted to give different heights to the slugs. Thus the motion of the slug in its holder was just about limited to the vertical degree of freedom.

The turnbuckle was attached directly to the pan of a double-beam Harvard trip balance, of capacity 2 kg and sensitivity 0.1 gram.

A long wooden trough with a welded sheet metal liner was used to cool the slugs in liquid nitrogen (Fig. 5). The trough itself sat in another wooden box, packed with dry ice. In addition, the sliding cover of the trough was constructed so as to hold dry ice. The box was long enough to hold about 24 slugs lying side by side.

The normal testing required a minimum of two men. Man "A" placed each slug in the holder, found the balance point, and announced the reading to man "B", who recorded it. It was the duty of "B" to check slug numbers against the test sheet, feed slugs to "A", remove slugs from the holder, place slugs in the liquid nitrogen bath, take differences on test sheets, separate rejects, and store finished slugs.

During the test, hot and cold slugs are tested alternately. A cold slug is removed from the exit end of the trough and handed to "A" as a warm slug is finished. This warm slug is then placed in the entrance end of the trough. After being tested, the cold slug is stored and a new warm slug is tested and the cycle repeated. The slugs slowly roll from the entrance to exit end of the trough. The process is continuous, but control is maintained by using groups of twenty slugs. Each test sheet records, and the trough holds, twenty at a time. Man "B" always

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has two test sheets active. On one he records data of warm slugs, on the other data of cold slugs. As soon as all data (warm and cold) are accumulated on a sheet, it is retired; the sheet on which warm data had been just recorded is ready to receive cold data, and a new sheet is started for warm data. A finished test sheet is reproduced as Fig. 6.

The test sheets which came with the slug batches, already contained the weights of the slugs. The "recording" man took the weight differences and separated the rejects as the test proceeded. A slug rejected for iron was not usually put in the liquid nitrogen bath.

Since the trough contained twenty slugs at all times, thirty-nine slugs were placed on the balance while a slug moved from entrance to exit end of the liquid nitrogen bath. This insured that each slug had a minimum of thirty minutes in the bath.

IV. RESULTS AND DISCUSSION

A total of 7000 slugs were tested in the above manner in a three-month period. When half this number had been examined using arbitrary reject figures, a group of 100 slugs (primarily rejects) were re-examined more accurately. From the Fe, two groups which had a large range of magnetic effects were chosen for destructive quantitative analysis to determine a calibration curve for iron and hydride. A vacuum fusion technique was used to determine hydrogen content and a spectrographic technique to measure iron concentration in the slugs. These techniques had an accuracy of 5 to 10%.

One group of 21 slugs had magnetic "pulls" at room temperature ranging from eleven to 127 grams. These slugs, primarily iron rejects, were analyzed for iron content using pie-shaped sections cut from various areas of the slugs. The other group of 21 slugs had magnetic "pull" differences (room temperature to liquid air temperature) ranging from one to 39 grams. These slugs, primarily hydride rejects, were analyzed for hydrogen. Small punch-

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ings taken from the center third of the slug were used.

The results of these analyses are plotted in Figs. 7 and 8. The iron analyses are directly equal to the average iron concentrations in the slug, since this iron is assumed to be uniformly distributed. The hydrogen results represent only the average concentration of hydrogen in the punchings of one slug. Since the extent of the core is unknown, the true hydrogen concentration cannot be given. One can only assume that the core size was related to the hydrogen concentration in it and the punchings fully averaged both effects.

The straight line shown in Figs. 7 and 8 represent the "best fit". A "t" test shows a high degree of significance for each slope. From an analysis of the data one can say that over 92% of the variance in iron content among the slugs is explainable by the straight line. However, only 64% of the variance in hydrogen content among the slugs is explainable. Considering the limitations pointed out above in sampling for hydrogen analysis, this difference is understandable.

Because time is consumed as the slug is transferred from the liquid nitrogen bath to the magnetic field, the slug is actually being tested at a temperature higher than 90°K. In order to ascertain the effect of warming up and to be sure that the transformation temperature for uranium hydride of 173°K was not exceeded, slugs of varying hydride contents were allowed to remain in the holder in the magnetic field while measurements were taken as a function of time. In no case was a detectable change found for at least five minutes. Since the operation was being performed in two minutes, no extra precautions were necessary.

As indicated previously, the magnetic pull depends on the position of the material in the field. In order to check the magnetic homogeneity of the slugs, 50 slugs of high iron content and 50 of high hydride content were tested with first one end up and then with the other.

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Not one of the high iron content slugs showed a marked difference, while three of the high-hydrogen slugs showed a significant variation. This result further explains the lower accuracy in the hydride determination but also indicates that more consistent results might be obtained by averaging tests taken with each end in the field.

It was suggested that other compounds with low Curie temperature might interfere with the hydride determination. A short survey indicated only UFe_2 , with a Curie temperature of 151°K , as likely, but further analysis showed that the compound would not be expected to form at the low temperatures used in the slug fabrication.

The sensitivity of the method is limited also by the pan balance and the fact that the magnetic pull is only a small fraction of the weight of the slug. To avoid this it should be possible to perform the test with a slug on a long pendulum suspended with its axis horizontal. The pendulum would support the slug's weight while a horizontal spring could be used to measure the magnetic pull.

The field strength in the electromagnet was found to become stable after a one-half hour warm up. The motor generator set used to excite the magnet acted as a stabilizer of input power fluctuations.

These factors lead to the conclusion that the overall accuracy of the technique as here calibrated is no better than 10% for iron and 20% for hydrogen. The reproducibility is, however, much better, being 5% for iron and hydride.

V. SUMMARY

A non-destructive magnetic method has been developed for the determination of uranium hydride and iron in hot-pressed uranium slugs. This method has been developed to the point where mass production testing is feasible.

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APPENDIX

Gouy's Method for Determinating Susceptibilities

A technique for the determination of k in paramagnetic and diamagnetic materials was proposed in 1889 by Gouy(1,2,3).

If a cylindrical rod is vertically suspended (see Fig. 4) between the pole pieces of a magnet so that it is in an inhomogeneous field, then the magnetic force acting on an element of this material of length dx and volume dV is:

$$(1) \quad dF_x = \frac{(k_m - k_a)}{2} \left(\frac{d(H_x)^2}{dx} + \frac{d(H_y)^2}{dx} + \frac{d(H_z)^2}{dx} \right) dV$$

where k_m and k_a are the respective susceptibilities of the specimen and the surrounding air, and H_x , H_y , and H_z are the rectangular components of the magnetic field intensity in the region of dx . In our experiment, the pole pieces of the magnet are such that the greater part of the magnetic field is directed along the y-direction, so that H_x and H_z , and hence $(H_x)^2$ and $(H_z)^2$, will be small. Thus the force acting at dx can be approximated as:

$$(2) \quad dF_x = \frac{(k_m - k_a)}{2} \frac{d(H_y)^2}{dx} dV = \frac{(k_m - k_a)}{2} \frac{d(H_y)^2}{dx} \alpha dx$$

where α is the cross-sectional area of the specimen. If we integrate

$$(3) \quad F = \int_{x=0}^{x=L} \alpha \frac{(k_m - k_a)}{2} \frac{d(H_y)^2}{dx} dx$$

$$= \frac{\alpha}{2} \int_{H_y=H_0}^{H_y=H} (k_m - k_a) d(H_y)^2$$

Two assumptions are made at this point: one, that the material be homogeneous in composition, so that k_m is not a function of H_y , and thus can be taken outside of the integral; and two, that the specimen is sufficiently long, so that the upper end of the bar is well removed from the field, or $H \gg H_0$. Then equation (3) becomes, setting $H_0 = 0$:

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$$(4) \quad F = \frac{a}{2} (k_m - k_a) \int_{H_y}^H d(H_y)^2 = \frac{a}{2} (k_m - k_a) H^2$$

Thus, knowing H , the magnetic field intensity, a , the cross sectional area, and k_a , the susceptibility of air, the susceptibility of the specimen, k_m , is obtained when the downward force F is measured.

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Fig. 1 - Gouy's Method for Determining Susceptibilities

Plate No. 208-2

Fig. 2 - ADL Electromagnet, Showing Pan Balance

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Plate No. 208-1

Fig. 3 - Magnetic Testing Area, Showing Pan Balance, Slug
in Lucite Holder, and Pole Pieces.

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Fig. 4 Field strength vs distance above the center of the ADL electromagnet

Fig. 5 Cooling box

FIG. 6 LIST OF SLUGS FOR MAGNETIC TESTING

Date Shipped to Bayside 4-14-54

Date Rec'd at Hicksville _____

Date Magnet Tested 4-15-54

Circle Rejects

Slug No.	Weight in Air** Room Temp.	Weight in Field** Room Temp.	Weight in Field** Cold	Iron Pull	Hydride Pull	Comments*
706B3	94.0	109.0	110.0	15	1	
6	95.0	111.2	119.3	16	8	
707B1	95.0	108.5	109.6	14	1	
2	91.8	105.1	105.4	13	0	
3	94.5	107.8	108.9	13	1	
4	94.9	109.6	109.7	15	0	
5	94.0	109.5	111.3	16	2	
6	89.1	100.9	103.2	12	2	
708B1	89.7	101.1	103.3	11	2	
3	87.5	98.5	100.0	11	2	
6	94.5	103.4	105.1	9	2	
709B1	92.3	122.6		30	-	
3	94.4	107.1	108.5	13	1	
4	92.2	105.6	108.9	13	3	
5	93.5	105.4	105.7	12	0	
6	93.7	106.9	107.2	13	0	
710B1	94.3	107.2	108.3	13	1	
2	92.6	104.8	106.0	12	1	
3	92.5	104.0	104.7	12	1	
4	93.1	106.1	106.7	13	1	

* Please note if slug appears light in weight, poor surface, etc.

** All weights less 1700 gms.

Fig. 7 Calibration for iron content

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Fig. 8 Calibration for hydrogen content

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