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USE OF A WATER BOILER REACTOR AS A PRODUCTION TEST FACILITY

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ABSTRACT

The feasibility of the use of a water boiler type reactor as a production test facility for making reactivity tests on various forms of uranium and uranium compounds has been investigated. The study indicates that the reactor should be very useful for production testing of the material with the danger coefficient technique. A suggested production procedure is outlined.

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The feasibility of the use of a water boiler type reactor as a production test facility for making reactivity tests on various forms of uranium and uranium compounds has been investigated. This investigation was initiated as a result of a letter from G. W. Wunder of National Lead Company of Ohio to C. Starr, North American Aviation, Inc., dated March 27, 1953. This letter and its enclosure outlined the types of measurements which should be possible with a test reactor which would be used at the FMPC. The measurements would be performed by the danger coefficient method in the core of the water boiler type reactor. The results reported here are based on analyses and preliminary measurements made in the North American Aviation Water Boiler Neutron Source currently in operation in the Atomic Energy Research Department at Downey.

The measurements in the Water Boiler Neutron Source indicate a limit of detectability of $\pm 10^{-3} \text{ cm}^2$ absorbing cross section. However, this limit can only be attained by making a precision type measurement which would probably require a larger expenditure of time than would be efficient for a production type facility. Therefore, in considering the types of measurements desired by National Lead Company a lower limit of detectability has been set at 0.005 cm^2 of absorbing cross section. This would then indicate that with a 1 kilogram sample of natural uranium, a parasitic absorption cross section of 0.005 cm^2 would be readily detectable. This has been experimentally checked by performing measurements on natural slugs $3/4$ inch in diameter by 4 inches long, which weigh approximately 500 grams. The equivalent of parasitic absorption cross section was obtained by "loading" the uranium slug with a small quantity of gold foil and/or aluminum foil to make a neutron absorption cross section equivalent to $0.004 \text{ cm}^2/\text{kilogram}$ of uranium. Measurements made on these slugs indicated that with one slug in the central exposure facility of the WBNS the rapid detection of the loaded parasitic absorption cross section was questionable. However, using two of these slugs with the same amount of loading, the parasitic absorbing cross section was readily detectable.

The various types of tests as listed in the enclosure to the above mentioned letter will be considered in order of their presentation there.

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1. Uranyl nitrate (liquid, molten, or crystalline) in which the total cross section of impurities may not exceed 0.05 to 0.5 cm^2 per kilogram of uranium. From the measurements outlined above, it appears that the detection of 0.05 cm^2 per kilogram of uranium in a sample of uranyl nitrate could be easily detected in 166 grams of the uranyl nitrate (this amount of uranyl nitrate would contain 100 grams of uranium). This amount of uranyl nitrate would only occupy a volume of approximately 59 cm^3 . Insertion of this volume of material in the central exposure facility of the WENS is not at all prohibitive.
2. Anhydrous uranium trioxide, uranium dioxide and uranium tetrafluoride. The tests of these compounds of uranium can be accomplished in the same manner as those of the uranyl nitrate outlined above. In order to detect 0.05 cm^2 of parasitic absorption cross section per kilogram of uranium only 120 grams of UO_3 and 113 grams of UO_2 would be required. The volumes of these amounts of the two compounds would be approximately 17 cm^3 and 11 cm^3 respectively. Approximately 132 grams of uranium fluoride would be needed to detect the 0.05 cm^2 of impurity per kilogram of uranium. It would not be possible to isolate an impurity of 10 parts per million of iron in any of these compounds. This amount of iron corresponds to an absorption cross section of $2.6 \times 10^{-4} \text{ cm}^2$ per kilogram of uranium and hence would be completely masked by the gross impurity of 0.05 cm^2 per kilogram.
3. Cast uranium metal samples. These metal samples might have impurities of the order of 0.05 cm^2 per kilogram of metal plus 30 ppm of carbon and 40 ppm of nitrogen. The gross impurity of 0.05 cm^2 per kilogram would be readily detectable with only 100 grams of the metal. The impurities of carbon and nitrogen will again be masked by any gross impurity of the order considered so far. This is evident since a concentration of 40 ppm of nitrogen amounts to an approximate cross section of only $3 \times 10^{-3} \text{ cm}^2$ per kilogram of uranium while the concentration of 350 ppm of carbon amounts to the entirely negligible amount of $6.8 \times 10^{-5} \text{ cm}^2$ per kilogram of uranium.

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4. Uranium slugs of the following dimensions:

1.4" x 4.1"; 1.4" x 9"; 1.1" x 9" .

These slugs contain approximately 1.9 kilograms, 4.2 kilograms, and 2.6 kilograms of uranium metal, respectively. With slugs of these sizes, impurity concentrations of 0.003 to 0.002 cm²/kg of uranium will be readily detectable.

5. Thorium metal in which the following impurities are considered: cadmium, 0.5 ppm; uranium, 25 ppm; beryllium, 500 ppm; iron, 200 ppm; nitrogen, 100 ppm; oxygen, 500 to 1000 ppm; carbon, 600 to 1000 ppm; boron, 1 ppm. These amounts of impurity correspond to the following concentrations of parasitic absorption cross section: Cd, 6×10^{-3} cm² per kilogram of thorium; Be, 3×10^{-4} cm² per kilogram of thorium; iron, 5.2×10^{-3} cm² per kilogram; nitrogen, 7.6×10^{-3} cm² per kilogram of thorium; oxygen, less than 7×10^{-6} cm² per kilogram of thorium; carbon, 1.4 to 2.2×10^{-4} cm² per kilogram of thorium; boron, 4.2×10^{-2} cm² per kilogram of thorium. Thus, it is seen that the boron impurity will entirely mask all of the rest of the impurities listed above. In order to maintain a limit of impurity of 0.05 cm² per kilogram of metal only about 100 grams of thorium metal would be required for a test sample. The use of kilogram samples of thorium will not be prohibitive because of volume limitations (these occupy a volume of approximately 90 cm³), but the problem of reactor control will probably be the limiting factor. The use of a one kilogram sample, which control limitations will not prohibit, would permit the detection of impurity concentrations of 0.005 cm²/kg of metal.
6. Thorium fluoride as anhydrous powder. The thorium fluoride would have the same impurities as those listed above for thorium with the exception of carbon, oxygen, and nitrogen. The tests on the thorium fluoride could be made in the same manner as those on the metallic thorium, but a larger volume of the fluoride would be needed in order to obtain comparable sensitivity with that of the thorium metal.
7. and 8. These two items consist of cast uranium-zirconium and uranium-chromium alloy. These would be metallic samples which could be machined,

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and hence the problems of testing are the same as those encountered in item (3) discussed above. The small amounts of zirconium and chromium present in the alloys contribute very little to the reactivity change of the sample, as both zirconium and chromium have low absorption cross sections, and the amounts present in the alloy are only a very small weight per cent of the total metal present. Thus, the accuracy which could be achieved on these alloys would be essentially the same as that available with the cast uranium samples.

9. Special uranium shapes. These shapes include hollow cylinders and uranium flats. The testing of the hollow cylinders offers no problem different from that presented in the test of the uranium metal slugs. Actually, one would expect the impurity detection to be somewhat better with the hollow cylinders because of the decrease in the self-shielding of the material. The uranium flats which are suggested to be 4 inches wide by 1/2 inch thick with lengths ranging up to several feet present a unique problem. A special facility would be necessary in order to insert metal of these geometries into the core of the reactor where the statistical weight is high enough to obtain measurements of the desired precision. However, if it is possible to cut samples from the flats, these could be inserted in the central exposure facility and tested in the same manner as the uranium metal slugs.
10. Assay of U^{235} in enriched and depleted uranium. Experimental tests to determine the feasibility of this type of measurement have been made in the NAA WENS on metallic samples of uranium 3/4 inch in diameter by 1 inch long in which the enrichments of U^{235} were 0.49 per cent, 0.71 per cent, (natural uranium) and 0.90 per cent. The measurements on these samples indicate that one can determine the per cent enrichment of U^{235} in the sample with an uncertainty of approximately ± 0.003 per cent. It might be noted here the accuracy quoted above is that obtained with a precision type measurement, and this accuracy might not be readily obtainable in a production type facility where one did not desire to spend the time required for this precision type measurement. However, there is no reason to believe that a precision of approximately ± 0.01 per cent could not be very easily obtained.

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11. The testing for trace elements in uranium and various uranium compounds by activation methods. It appears that this particular type of test cannot be efficiently conducted by activation in a reactor. It appears that any activation produced by the impurity in the samples to be tested would probably be completely masked by the natural radioactivity of the uranium plus the fission product activity produced by even a very brief activation in the reactor.

A brief summary of the above outlined characteristics is given in Table I.

It must be emphasized that this type of a test procedure will not permit one to determine the type of impurity causing the decrease in reactivity in the uranium. Rather, the measurement will show that some gross amount of parasitic absorption cross section is present in the sample which is being tested, and that this amount of absorber has the indicated effect on the reactivity of the uranium. Hence, it is not at all possible to isolate various elemental impurities such as the iron, nitrogen, carbon, etc., as outlined in the various sections of Wunder's letter to Starr. A problem of which one must be cognizant is the control problem in the reactor. This is most important in the testing of thorium metal and thorium compounds, since the thorium has a much greater effect on the reactivity than a corresponding amount of uranium.

The results of the earlier mentioned experiments with thorium and with different enrichments of uranium are shown in Fig. 1. These measurements were made with thorium and uranium slugs $3/4$ inch diameter by 1 inch long. Three enrichments of uranium were used, 0.49 per cent, 0.71 per cent (natural uranium), and 0.90 per cent. The reactivity change associated with each sample was determined relative to the condition with graphite in the core of the WBNS in place of the sample. In addition, measurements were made on the equivalent of natural uranium slugs $3/8$ inch diameter by 8 inches long. Extrapolations from these measurements indicate that four kilograms of natural uranium and one kilogram of thorium would produce negative reactivity changes of approximately 0.25 per cent and 0.7 per cent, respectively, when inserted in the core of the WBNS. These numbers are rather uncertain because of lack of knowledge of the self-shielding effects and the resulting effects on the reactivity changes.

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The control problem must also be considered in the assay of enriched uranium. The results given in Fig. 1 indicate that uranium with an enrichment of about 1 per cent would have no effect on the reactivity of the reactor when inserted in the central exposure facility. This means that in the assay of material of greater than 1 per cent enrichment a positive reactivity change would be obtained. Thus the control rods and the loading of the reactor must be such as to adequately compensate for both this positive reactivity change and the negative reactivity changes obtained in the testing of natural and depleted uranium metal and compounds. However, sufficient control for the above conditions could be very easily obtained.

A proposed test facility would be a water boiler reactor consisting of a spherical core encased in a graphite reflector with suitable shielding for operation at power levels of up to about 10 watts. No cooling facilities would be required. The central exposure facility in which the samples would be inserted for testing would be a tube through the spherical core. Specially designed sample holding and changing equipment would be incorporated in the reactor. This would include an auxiliary control rod which would permit rapid, safe, and efficient sample changing without shut-down of the reactor. The control rod would be calibrated, and charts, nomograms, or graphs could be used to present the data in the most useful form.

With the above type of facility the following is presented as a schedule for production testing of samples:

- 1) Start the reactor and bring it up to power with the auxiliary control rod in place.
- 2) Insert a standard slug or sample of material of the type to be tested in the reactor with the sample changing equipment.
- 3) Obtain a reactor period measurement by letting the reactor power drift without changing the control rod position for about 10 minutes.
- 4) Determine period of reactor.
- 5) From calibration of control rod and period measurement above, obtain critical position.
- 6) Set control rod on exact critical position.

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- 7) Insert test sample with sample changing equipment, making use of the auxiliary control rod and keeping the reactor control rod at the critical position for the standard. Allow about 3 to 5 minutes for this change and for transients to disappear.
- 8) Observe reactor drift for about 10 minutes.
- 9) From period of reactor as determined in (8) and from control rod calibration determine effect of test sample. This effect can be given either in terms of reactivity or parasitic absorption cross section.
- 10) Insert next test sample and follow through steps (7), (8), and (9).
- 11) Periodically (it is suggested every 2 hours) re-standardize the reactor by following through steps (2) through (6). This will compensate for any slow variations in the reactivity of the reactor during the period required for the measurements.

If this type of procedure were used for routine testing, about 7 test samples and one standardization of the reactor could be completed every two hours. Thus, it appears that even if one allows 2 hours out of each 24 hour day for contingencies, approximately 75 routine tests could be completed each working day. However, this figure is perhaps optimistic because of the necessity of some rechecking of samples in which the measurement has not been of a positive enough character. Thus, 50 determinations would probably be a more reasonable number to plan for each 24 hour working day. These 50 determinations per day are consistent with the production figures outlined in the original reference letter.

Tests on the NAA water boiler at an operating level of 0.2 watt indicate that a typical value for the slow drift in reactivity of the reactor is about 0.1 inh. per hour. This would correspond to a drift of approximately $5 \times 10^{-3} \text{ cm}^2$ of cross section per hour. Thus, if this drift were to be experienced during a 2 hour run as outlined above, the change in reactivity would amount to approximately 0.01 cm^2 of cross section. However, this does not mean that the limit of detectability of impurity cross section would necessarily be increased to this 0.01 sq. cm. It is believed that by

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comparing two successive standardization measurements a correction can be applied for the drift of the reactivity during the period between the two measurements.

It would be desirable to operate the water boiler reactor for such a test facility at the lowest power feasible with obtaining sufficient detection signals to permit accurate measurements of the reactor periods. The measurements with the NAA water boiler have all been made at a power level of approximately 0.2 watt. With the production facility it might be desirable to increase this operating power level to perhaps the order of one watt. It is recommended, though, that power levels no greater than about one watt be used, so that changes in the reactivity resulting from temperature effects, power coefficients, and other variations in the activity associated with high power operation be minimized. Also, the operation at this low power level results in small build-up of fission product activity in the uranium samples undergoing the test. There appears to be no indication that suitable instrumentation cannot be designed to give adequate signals at power levels of one watt and less.

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TABLE IRESPONSE OF WATER BOILER TYPE REACTORTO IMPURITIES IN URANIUM AND THORIUM

MATERIAL	FORM	DENSITY gms/cm ³	AMOUNT kg	VOLUME cm ³	AMOUNT OF U IN SAMPLE kg	CONCENTRATION OF IMPURITY WHICH IS DETECTABLE cm ² /kg
UO ₂ (NO ₃) ₂	liquid, molten, crystalline	2.81	0.67	236	0.4	0.012
UO ₃	anhydrous powder	7.29	1.20	165	1.0	0.005
UO ₂	"	10.9	2.26	208	2.0	0.003
UF ₄	"	6.7	1.32	197	1.0	0.005
U Metal Slugs	1.4" x 4.1"	18.7	1.93	103	1.93	0.003
	1.4" x 9"	18.7	4.25	227	4.25	0.002
	1.1" x 9"	18.7	2.62	140	2.62	0.003
U-Zr	metal alloy 3-5% Zr	~17.7	2.1	118	2.0	0.003
U-Cr	metal alloy 0.1-0.3% Cr	18.7	2.0	108	2.0	0.003
Th	metal	11.2	1.0	89	1.0	0.005
Th F ₄	anhydrous powder	3.8	0.85	225	0.64	0.008

AMOUNT OF Th
in Sample

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REACTIVITY CHANGE IN NAA-WBNS
FOR CYLINDERS OF U AND TH IN THE
CENTER OF WBNS CENTRAL
EXPOSURE FACILITY