GENERAL DYNAMICS

GA-1934

**MARITIME GAS-COOLED
REACTOR PROGRAM**

FISSION-PRODUCT PLATEOUT FROM A HELIUM GAS STREAM

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This is a preprint of a paper submitted for
publication in Nucleonics.

GENERAL ATOMIC DIVISION

Contract AT(04-3)-187 with the
U. S. Atomic Energy Commission
and the Maritime Administration

March 10, 1961

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FISSION-PRODUCT PLATEOUT FROM A HELIUM GAS STREAM

One of the problems associated with high-temperature gas-cooled reactor systems is related to the behavior of the fission products which may escape from fuel elements through pin-hole leaks or through a fuel cladding failure. The gaseous fission products, xenon and krypton, that escape from the fuel elements will be circulated throughout the system in the coolant stream until they decay to their daughter products, cesium and rubidium. Nongaseous fission products that are volatile at the fuel temperature may also be expected to escape through fuel cladding defects. These nongaseous isotopes will be deposited from the gas coolant stream at points in the system where temperature and surface conditions are favorable. Thus, a knowledge of the behavior of the nongaseous isotopes is useful in the design of fission-product collection systems and of the plant radiation shielding.

Exploratory tracer experiments were performed to study the plateout of nongaseous fission products from helium as part of the Maritime Gas-cooled Reactor (MGCR) program. This paper describes the experimental procedures used and the results obtained in the exploratory studies.

A 1-g sample of finely powdered, natural-enrichment UO_2 was irradiated for one hour at a thermal-neutron flux of approximately 1×10^{12} nv in General Atomic's TRIGA reactor to provide tracer quantities of fission products. The irradiated UO_2 powder was placed in a porcelain boat, which was inserted in a 1-in. -diameter quartz tube. The tube was heated by four tube furnaces placed end to end along the tube. The furnaces were operated at successively lower temperatures of 1800°F , 1400°F , 1000°F , and 400°F . The temperature profile along the length of the tube was determined with a thermocouple probe prior to the insertion of the boat containing the UO_2 powder. The UO_2 sample was located at the

hottest point in the system. A train of metal coupons extended from the porcelain boat through the temperature gradient to a point outside the last furnace, where the temperature was approximately room temperature.

Bureau of Mines grade tank helium was fed into the quartz tube at the high-temperature end of the apparatus at a rate of approximately $1 \text{ ft}^3/\text{hr}$. The helium in the tube was at atmospheric pressure. The heating and the helium flow were continued for 2-1/2 days, and the fission products that were swept out of the quartz tube were collected in suitable traps. Oxidation of the metal specimens occurred in each of the two tests that were performed. The concentration of impurities in the helium gas stream is not known.

At the conclusion of the heating cycle the metal coupons, the Nichrome wire links that were used to connect the metal coupons together, and the quartz tube were analyzed by gamma-spectrometry techniques. Gross counting of gamma photons of greater than 10 kev energy and gamma-spectrometry analysis for specific isotopes were performed using a counting geometry of approximately 2π .

In the first experiment, Type 302 stainless steel coupons connected together with Nichrome wire links were used. The results of the experiment are presented in Fig. 1. The gross activity levels are reported in counts per minute per square inch of surface area for the metal coupons and for the wire links. The gross activity level for the quartz tube is only reported in counts per minute, since the surface area being scanned could not be defined accurately for the experimental arrangement that was used.

From Fig. 1 it may be observed that the gross activity for each of the three materials that were exposed to the fission-product vapors follows the same general pattern. Peaks in the activity profiles were located at approximately 1200°F , 800°F , and 500°F . In addition, it may be noted that the specific gross activity of the Nichrome (80% nickel, 20% chromium) was markedly greater than that of the Type 302 (18% chromium, 8% nickel, balance iron) stainless steel coupons. This behavior appears to indicate

Fig. 1--Fission-product gamma activity for specimens from first plateout experiment

that high nickel content in the metal may tend to promote plateout of volatile fission-product species. Radioactive isotopes that were identified as being deposited on the metal specimens were I^{131} , I^{133} , Mo^{99} , Ru^{103} , and Te^{132} . Other species were probably present but could not be identified in the gamma spectrum. The location of the maximum intensity of the gamma radiation from the above isotopes was found to coincide approximately with the peaks in the gross gamma activity. These locations are indicated in Fig. 1.

The second experiment was designed to determine if the composition of the metal coupons affected the amount of activity plated out. Metals were selected to provide different concentrations of nickel, and included (1) nickel, (2) Nichrome (80% nickel), (3) Type 310 stainless steel (20% nickel), and (4) Type 405 stainless steel (0% nickel). Four parallel trains of coupons were used, with each tier in the train containing one coupon of each of the above materials. The results of the experiment are presented in Fig. 2. During the course of the 2-1/2-day heating cycle the helium supply was exhausted and the flow stopped. However, the location of the gross gamma activity peaks remained essentially unchanged when compared with the results of the first experiment. Some indication that high nickel content promotes plateout at high temperature was also obtained. For temperatures below about $1200^{\circ}F$ it was not clearly demonstrated that the composition of the coupons had a marked effect on plateout.

In each experiment approximately 5% to 6% of the total fission-product activity contained in the UO_2 source could be accounted for as having been plated out on the test specimens. Also, the gamma activity of specimens that had been maintained at or near room temperature during the experiments was markedly lower than that of specimens that had been heated to $400^{\circ}F$.

The results of these experiments may be summarized as follows:

1. It is concluded that significant quantities of high-activity fission products are volatilized from powdered UO_2 at $1800^{\circ}F$.
2. Plateout of nongaseous fission products from a helium stream

Fig. 2--Fission-product gamma activity for specimens from second plateout experiment

occurred on all of the materials exposed to the stream over the temperature range from 1700°F to 400°F.

3. Fractionation of ruthenium, molybdenum, tellurium, and iodine was observed.
4. Some indication of the influence of metal surface composition on the plateout process was obtained. However, additional data are required in order to definitely establish such a relationship.