The Behavior of Fission Products Captured in Graphite Powder by Nuclear Recoil. III. Diffusion of Iodine-133 and Tellurium-132 in Graphite at High Temperatures

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(Received September 3, 1960)

One of the distinctive features of a semi-homogeneous reactor is its use of a fuel element of the mixture of graphite and uranium dioxide. It was stated in the previous paper[1] that most of the fission products were captured in graphite by recoil energy. The diffusion of fission products captured in graphite at high temperatures is an important subject from the standpoint mentioned below. The release of fission products from a fuel element causes contamination of the coolant and its facilities. On the other hand, it is desirable from the viewpoint of neutron economy to remove fission products to some extent.

But the high velocity of diffusion results in the release of the fission products emitting delayed neutrons that are important in reactor control.

This paper describes the diffusion of iodine and tellurium, following the description of the diffusion of xenon in the previous paper[1].

In operating a reactor, the behavior of iodine-135, precursor of xenon-135, must be taken into consideration as well as that of xenon-135, which has an abnormally large neutron absorption cross-section. Because of the short half-life of iodine-135, iodine-133 was used as a tracer in this experiment. On the diffusion of tellurium which shows a chemical behavior similar to iodine, tellurium-132 was used as a tracer.

Experimental

Preparation of Samples.—A mixture of 1 g. of graphite powder and 50 mg. of uranium dioxide was irradiated with thermal neutrons (thermal neutron flux, about 5×10^{11} neutron/cm^2/sec.) for 2 hr. in an experimental hole of a JRR-1 reactor. After irradiation the mixture was treated with hot 4 N nitric acid solution to dissolve the uranium dioxide and neptunium-239 produced. The graphite was washed with distilled water and dried at 100°C. It was already reported that the fission products captured in graphite were not leached out by being treated with 4 N nitric acid solution in a short period of time[2]. In order to allow short-lived

TABLE I. PARTICLE SIZES OF GRAPHITE SAMPLES AND THE ANALYTICAL DATA OF IMPURITIES

	Sample		
	AG (Artificial graphite)	NG-1 (Natural graphite-1)	NG-2 (Natural graphite-2)
Paticle size			
μ	30	1	50
Impurity			
Ash (total)	0.07%	0.3%	0.8%
SiO_2	250 p.p.m.	220 p.p.m.	3700 p.p.m.
Al_2O_3	210	60	3500
Fe_2O_3	130	24	300
MgO	15	24	100
CaO	70	36	300
V_2O_5	—	0.12	0.5
MnO	—	—	—
Na_2O	20	12	25
K_2O	15	12	20
NiO	—	0.06	—
ThO_2	—	2.4	—
CuO	—	—	—
B_2O_3	0.1	0.02	0.5

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1) T. Nakai, S. Yajima, K. Shiba, J. Osugi and D. Shinoda, This Bulletin, 33, 497 (1960).

2) T. Nakai, S. Yajima, K. Shiba, J. Osugi and D. Shinoda, ibid., 33, 494 (1960).

fission iodine nuclides to decay, the irradiated graphite samples were cooled for about forty hours before use. The particle sizes of graphite samples used in this experiment and analytical data on impurities are given in Table I.

Flow System.—*Diffusion of Iodine-133.*—A flow system is illustrated in Fig. 1. The furnace consisted of a heater of silicon carbide. The temperature of the furnace was measured with a platinum-platinum rhodium thermocouple with a regulator control. The whole assembly was made of silica. The sponge titanium heated at 800°C served to remove a trace amount of oxygen in helium to avoid oxidation of the graphite surface. The silica tube from the diffusion cell to a copper trap was heated at over 300°C with a nichrome winding to prevent fission iodine from deposition on the wall of the tube. A copper trap was heated at 430°C in a furnace, because it was experimentally confirmed that fission iodine was completely captured in a copper net at 400 to 450°C.

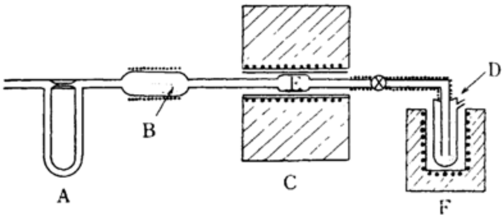

Fig. 1 Flow system.
- A, Flow meter
- B, Sponge titanium
- C, Furnace
- D, Copper trap
- E, Diffusion cell
- F, Heater

The graphite powder mentioned above was placed in the diffusion cell. Helium gas was introduced into it. After confirming the fact that gas took the place of air, the diffusion cell with the graphite powder was slid into the preheated furnace, when the temperature was kept constant at 400, 800 and 1200°C (or 1100°C). Helium gas swept over the sample at a constant flow rate (100 ml./min.) and carried the volatile fission products diffusing out of graphite. The iodine-133 used as a measure of diffusion was separated from the fission products and was captured in a copper trap together with other fission iodine nuclides. The trap was exchanged for a new one at appropriate intervals.

Diffusion of Tellurium-132.—The apparatus was almost similar to that for diffusion of iodine-133. A copper trap was used to capture tellurium-132. It was found in advance that fission tellurium was completely captured in copper at 700 to 720°C. Therefore, the copper trap was heated at 710°C.

The silica tube from the diffusion cell to a trap was heated at about 700°C with a nichrome winding to avoid the deposition of fission tellurium on the wall of the tube.

Determination of Iodine-133 and Tellurium-132 Activities.—Activities of iodine-133 and tellurium-132 were measured with a RCL 256 channel gamma-ray spectrometer with a well type sodium iodide scintillator (2 inch diameter × 1.75). The gamma-ray spectra of the fission products in the graphite sample, iodine-133 and tellurium-132 in a copper trap, all cooled for forty hours after irradiation, are illustrated in Fig. 2. Iodine-132, the daughter of tellurium-132, exists in a trap in the state of transient equilibrium. Because the height of a photoelectric peak is proportional to the activity of the iodine-133 released and thus to the amount of iodine-133, the percentage of iodine released was calculated by comparing the height of the gamma-ray spectrum of the graphite sample, which was not heat-treated, with that of a trap. It was impossible to measure directly the activity of tellurium-132 in the graphite sample. Therefore, radiochemically pure tellurium was separated from the sample by the following radiochemical procedure. After burning the sample the residue was dissolved in nitric acid. The tellurium released from the sample on burning was caught in nitric acid solution and the resulting solution was combined with the solution with the residue. A tellurium carrier was then added to the solution, reduced with sulfur dioxide and precipitated as a metal. Then the activity of the tellurium metal was measured. The percentage of tellurium released was calculated as described in the case of iodine.

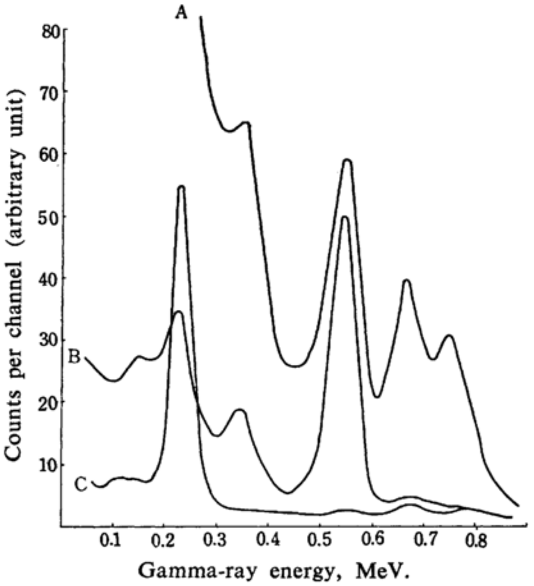

Fig. 2. Gamma-ray spectra of (A) fission products in graphite sample and (B) iodine-133 and (C) tellurium-132 in copper trap.

Results and Discussion

Isothermal curves of the diffusion of iodine-133 and tellurium-132 in four kinds of graphite are illustrated in Figs. 3—10. These figures show that with these nuclides the respective curve apparently consists of a rapid diffusion lasting for about 30 min. and a slow diffusion

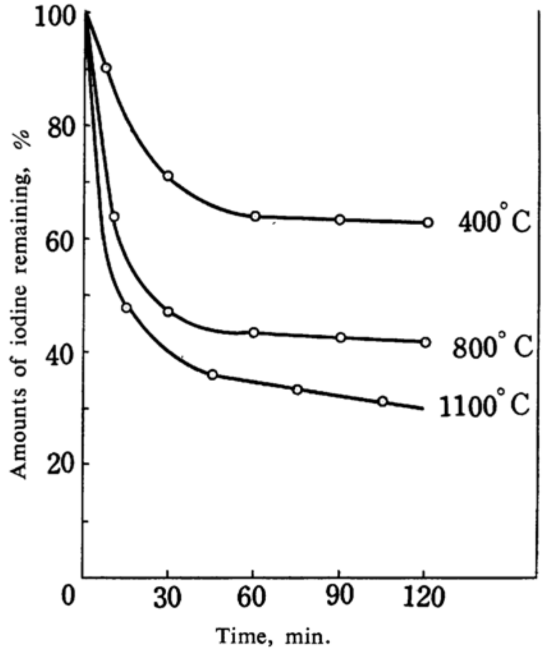

Fig. 3 Isothermal curves of diffusion of iodine-133 in natural graphite-1.

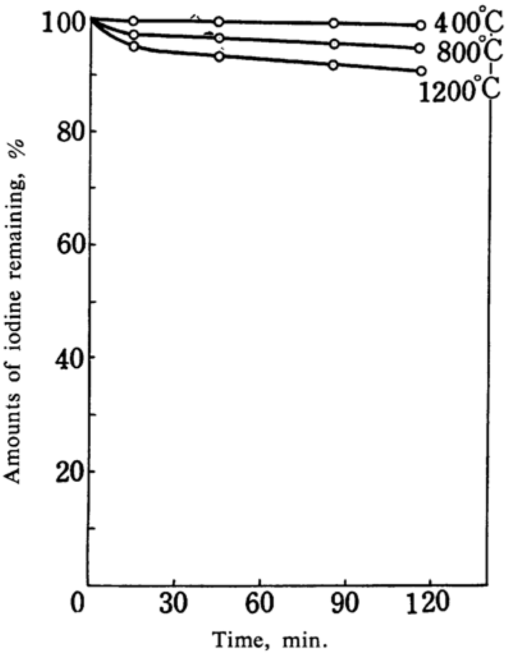

Fig. 5 Isothermal curves of diffusion of iodine-133 in artificial graphite.

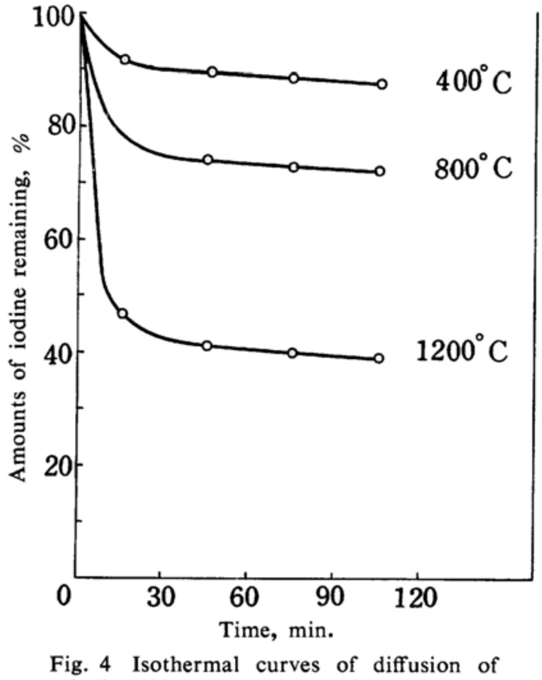

Fig. 4 Isothermal curves of diffusion of iodine-133 in natural graphite-2.

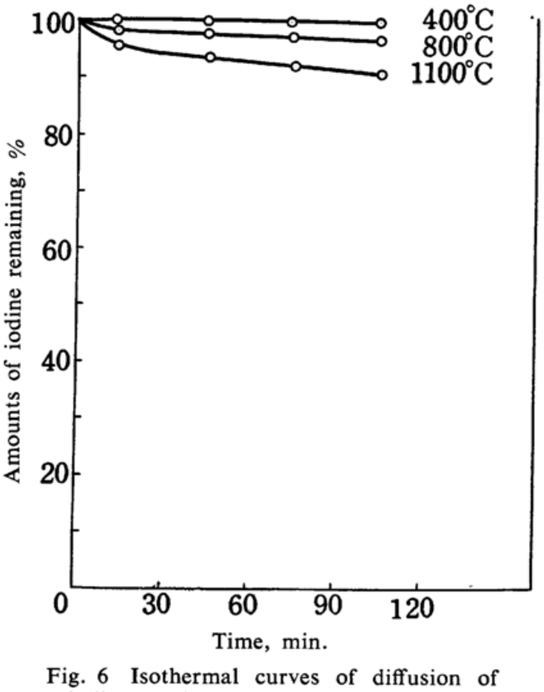

Fig. 6 Isothermal curves of diffusion of iodine-133 in amorphous carbon.

following it. This tendency is the same as that observed in the diffusion of xenon-135, where the activation energy of interlaminar diffusion of xenon-135 in graphite was calculated from a slow diffusion[1].

It has, however, been expected that there are several activation energies in diffusion in graphite. In fact, it is difficult to interpret the diffusion in graphite only from the interlaminar activation energy. A study is being continued on this subject with the use of a new method, and interesting information has

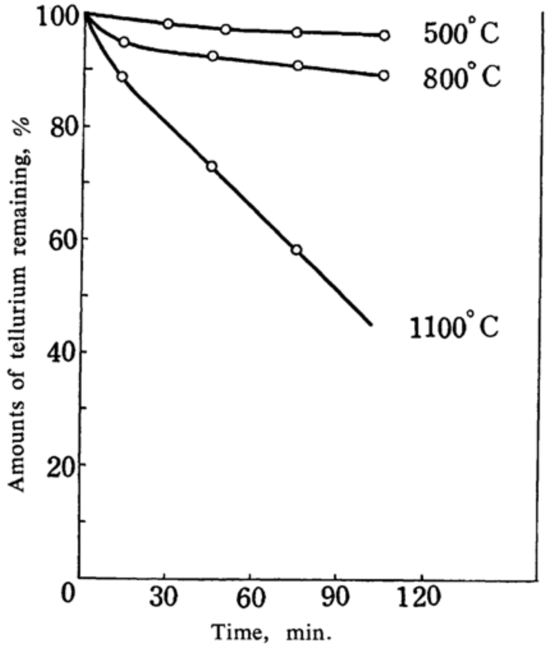

Fig. 7 Isothermal curves of diffusion of tellurium-132 in natural graphite-1.

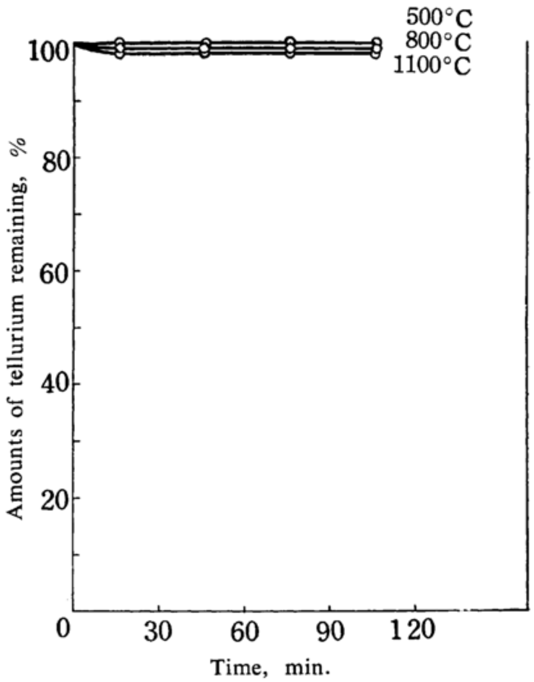

Fig. 9 Isothermal curves of diffusion of tellurium-132 in artificial graphite.

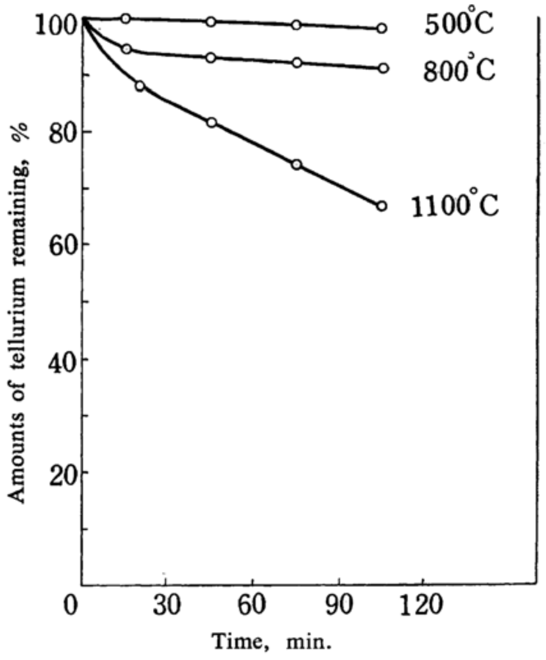

Fig. 8 Isothermal curves of diffusion of tellurium-132 in natural graphite-2.

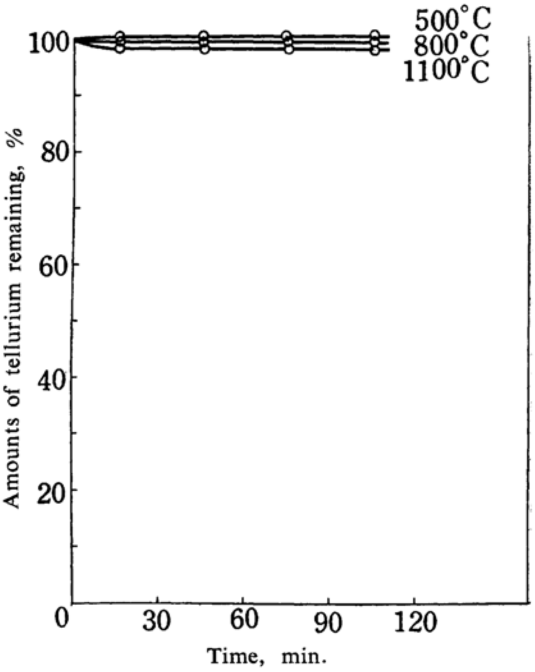

Fig. 10 Isothermal curves of diffusion of tellurium-132 in amorphous carbon.

been obtained. Details of the study will be reported later[3].

The rates of diffusion of iodine-133 and tellurium-132 in natural graphite are greater than those in artificial graphite. This result is similar to the diffusion of xenon-135 in graphite observed in the previous work[1]. According to the X-ray diffraction and electron diffraction studies, natural graphite has a large

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[3] S. Yajima, S. Ichiba, Y. Kamemoto and K. Shiba, This Bulletin, in press.

mosaic structure and artificial graphite has a small one. From this observation, it will be concluded that natural graphite has a lower density of dislocation than artificial graphite and that the trapping centers which are distributed along the dislocation line are less in natural graphite than in artificial graphite. Consequently, it is expected that the rates of the diffusion of iodine-133 and tellurium-132 in natural graphite, which has only a few trapping centers, are greater than in artificial graphite which has many trapping centers.

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