Short-range Dating of Fresh Nuclear Debris by the Activity Ratio of Iodine-133 to Iodine-131

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In order to investigate the radiochemical composition at time-zero of short-lived radionuclides in fallout particles and its time variation shortly after a nuclear test explosion, it is necessary to determine as accurately as possible the age of the fallout particles. Fresh nuclear debris particles usually consist mainly of short-lived nuclides, such as ^{97}Zr, ^{99}Mo, ^{143}Ce, ^{132}Te-^{132}I, ^{133}I and ^{239}Np. Recently we proposed a simple method for the short-range dating of fresh nuclear debris;[1] this method was successfully carried out by the determination of the activity ratio of ^{97}Zr to ^{95}Zr at a given time 2—7 days after a nuclear explosion.

Results and Discussion

According to the radiochemical composition of short-lived fission products present in hot fallout particles, the particles can be grouped roughly into two classes. The first is enriched with refractory nuclides, such as ^{97}Zr, ^{99}Mo, and ^{144}Ce, and depleted in volatilely-behaving ones, such as ^{132}Te-^{132}I and ^{133}I. The second shows the reverse trend regarding the enrichment of these nuclides; that is, refractory nuclides are depleted and volatile ones, enriched. In many cases these two different types of particles were not found at the same time among hot particles or in rain samples resulting from a given nuclear test explosion, but one of the two types was detected shortly after a nuclear detonation. The first type of particles was recently found among the fallout particles derived from the May 9, 1966, Chinese nuclear test explosion[2] and the second, in the particles (Fig. 1) and rain samples from the December 28, 1966, Chinese nuclear test. Figure 1 shows three γ-ray spectra of a hot particle collected on December 30, 1966; the spectra, A, B, and C, were measured 9, 28 and 37 days after the explosion, respectively. From these spectra it is obvious that this particle consisted mainly of ^{103}Ru, ^{132}Te-^{132}I, ^{133}I, and ^{239}Np, and that it contained no detectable amounts of ^{95}Zr-^{95}Nb or, hence, of ^{97}Zr-^{97}Nb.

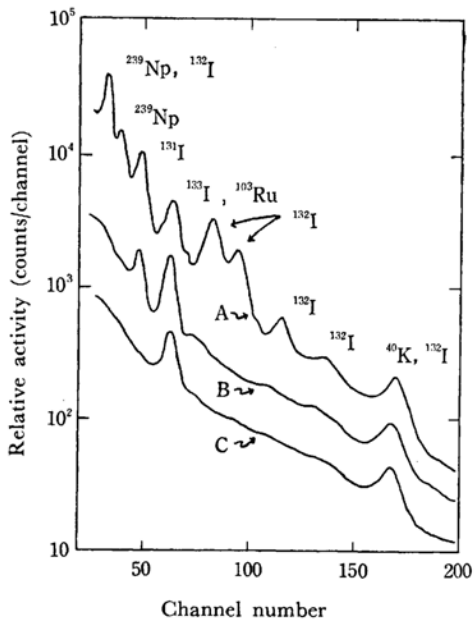
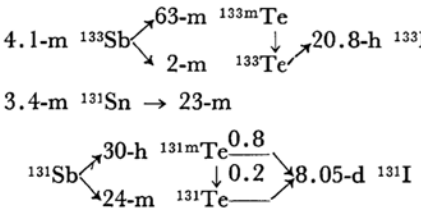


Fig. 1. Gamma-ray spectra of a fallout particle from the December 28, 1966, Chinese nuclear explosion. The spectra A, B and C were measure on January 6, 25 and March 3, 1967 respectively.

1) T. Sotobayashi, T. Suzuki and S. Koyama, *Nature*, **215**, 728 (1967).
2) T. Sotobayashi, T. Suzuki and S. Koyama, This Bulletin, **40**, 1555 (1967).

The $^{97}Zr/^{95}Zr$ dating method is not suitable for the radioiodine-rich particle shown in Fig. 1 because of the very great trouble needed to collect a number of fallout particles large enough for the activity ratio to be determined with sufficient accuracy. In the present work, therefore, we have especially examined two isotopes, ^{133}I and ^{131}I, present in large quantities in the particles in question. The ^{133}I and ^{131}I isotopes decay, with 20.8-h and 8.05-d half-lives, to ^{133}Xe and ^{131}Xe, respectively. Thus, we can expect the $^{133}I/^{131}I$ activity ratio to provide a radioactive clock very sensitive to small differences in time. The two mass chains, 133 and 131, are relatively complex compared with those for 97 and 95:



At the earlier stages of the formation processes of fallout particles, it seems that the two nuclides, ^{133}I and ^{131}I, still remain in the forms of ^{133}Sb and ^{131}Sn, respectively; this is on the assumption that the time taken for fission products to condense and to form a fallout particle is 40 sec or less after an explosion.[3] The melting points of antimony and tin are 630 and 230°C, respectively, and the solidification temperature of the carrier material (silicate soil) of fallout particles can reasonably be considered to be ~1700°C.[4] Taking these points into consideration, we will show here that the requirements for a successful dating are fulfilled by the ^{133}I and ^{131}I fission fragments extracted from the particles or rain samples resulting from the December 28, 1966, explosion.

Highly-radioactive fallout particles and rain-water samples were collected at Niigata between December 30, 1966, and January 2, 1967. One particle and 3 rain samples were submitted to preliminary treatment for chemical separation. The sample solutions thus obtained (1 M HNO_3) were then analyzed by the HNO_2-CCl_4 extraction method. The activities of the purified radioiodines in the form of silver iodide were measured using a $1\frac{3}{4}$ in $\times$ 1 in NaI(Tl) scintillation detector with a 200-channel pulse-height analyzer, and were determined by measuring the areas of the photopeaks at 0.36 and 0.53 MeV.

Figure 2 shows the activity ratios of ^{133}I to ^{131}I as a function of the time 2—10 days after the explosion. The solid line in this figure was drawn

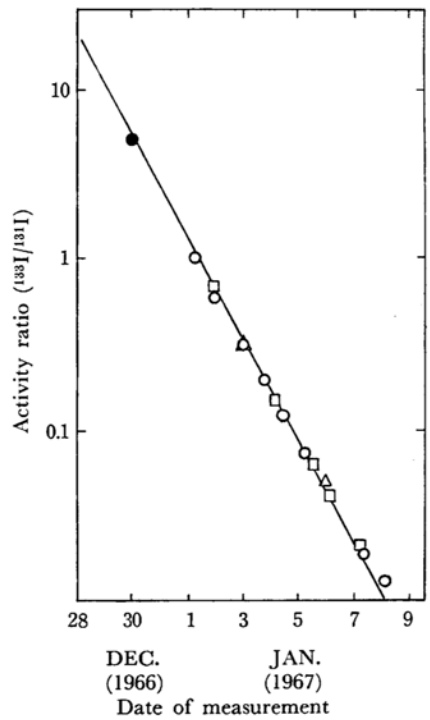


Fig. 2. Activity ratios of ^{133}I to ^{131}I as a function of time 2-10 days after the explosion.
●, a particle sample collected on Dec. 30, 1966
○, a rain sample collected on Dec. 31, 1966
□, a rain sample collected on Jan. 1, 1967
△, a rain sample collected on Jan. 2, 1967

against the time on the assumption that the cumulative fission yields of ^{133}I and ^{131}I are 6.9 and 3.1, respectively.[5] Further, the Chinese nuclear bomb was estimated to have been blasted at 14 : 30 h on December 28, 1966 (Japanese time), by the $^{97}Zr/^{95}Zr$ method.[1] We regarded this estimated time of explosion as the time-zero of the nuclear fission event because the exact time of the explosion was not announced by the Chinese agency responsible. It can clearly be seen in Fig. 2 that most of the measured ratios lie nearly along the assumed line. This finding suggests that, in spite of some differences in the physical and nuclear properties of each first member for the two mass chains, 133 and 131, no significant fractionation occurred between ^{133}I and ^{131}I during the formation of fallout particles. As a result, the dating method by the activity ratio $^{133}I/^{131}I$ is well suited for the short-range dating of fresh nuclear debris of the second type.

In conclusion, when any kind of fresh nuclear debris is found shortly after a nuclear explosion, either of the two dating methods will enable us to determine readily the age of this fresh nuclear debris with sufficient accuracy.

3) R. S. Clark, K. Yoshikawa, M. N. Rao, B. D. Palmer, M. Thein and P. K. Kuroda, *J. Geophys. Res.*, **72**, 1793 (1967).
4) S. H. Cassidy and G. R. Crocker, USNRDL-TR-67-70, 8 (1967).
5) S. Katcoff, *Nucleonics*, **16**, 78 (1958).