Elements 99 and 100 from Pile-Irradiated Plutonium*

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THIS note[1] describes some nuclear and chemical properties of elements 99[2] and 100. The transcalifornium elements were produced in a pile irradiation of plutonium (integrated flux of 1.0×10^{22} neutrons) in the Materials Testing Reactor. The purified transcurium elements were separated from each other with a Dowex 50 cation citrate column.[3] The final element-99 peak (Fig. 1) contained a total of 3.3×10^4 alpha disintegrations/minute. The energy of the prominent alpha group was measured by pulse analysis to be 6.64 Mev. The alpha spectrum of this isotope appears to exhibit fine structure which is characteristic for odd-even nuclides, e.g., similar to Np^{237} and Am^{241}. In the final californium peak were 2.3×10^5 alpha disintegrations/minute as well as 4.6×10^3 spontaneous fission disintegrations/minute. Alpha pulse analysis of the californium fraction showed at least two alpha groups of 6.01 and 6.10 Mev. These alpha groups chemically follow the large spontaneous fission activity as shown in Fig. 1 and are in the ratio of forty-five californium alpha disintegrations to one spontaneous fission disintegration. The presence of short spontaneous fission half-lives in the californium fraction is supporting evidence for the recently published spontaneous fission systematics.[4]

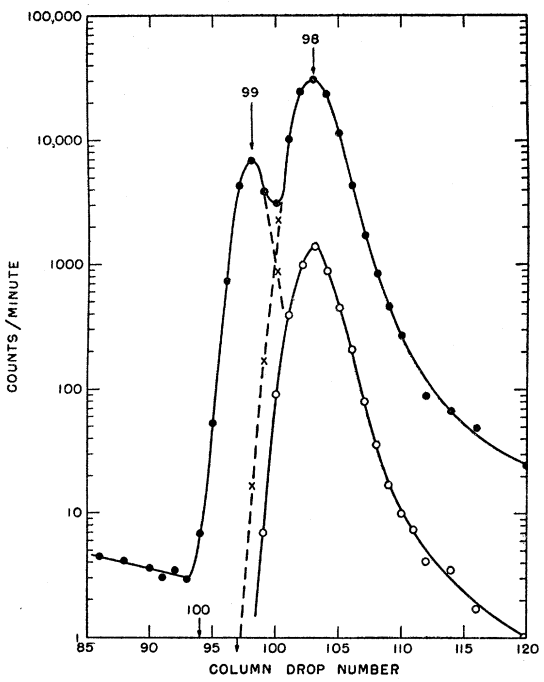

FIG. 1. The solid points represent the total activity in counts/minute (alpha particles plus spontaneous fissions) of successive drops from the Dowex 50 cation citrate column. The open circles give the spontaneous fissions in counts/minute as a function of drop number. The dashed lines give the respective amounts of californium and element-99 activities in the drops which contain alpha activity from both elements. The curium began to elute at drop 170 in this column run.

The small amount of spontaneous fission activity on plates containing the 6.64-Mev alpha particles is a measure of the excellent separation of element 99 from 98 (see Fig. 1). Drop 97 contained 8600 disintegrations/minute of 6.64-Mev alpha particles and about 0.004 ± 0.002 fission disintegrations/minute. Assigning all of the fissions to californium gives a limit of 0.2 disintegrations/minute of californium alpha particles on the above plate.

The observed half-life of the 6.64-Mev alpha-emitting isotope of element 99 is 20 ± 2 days (the decay was over a period of 8 days). By using a 20-day decay half-life for the above element-99 isotope, the spontaneous fission half-life is calculated to be greater than 10^5 years. This half-life limit can be increased if the spontaneous fissions do not decay with the 6.64-Mev alphas. Such a long spontaneous fission half-life (compared to even-even nuclides) for a 99 isotope is consistent with expectations for an odd-Z element. The 6.64-Mev alpha has grown in drop 109 from 2 counts/minute (at the time of the first alpha pulse analysis) to 13 counts/minute in six days, establishing its genetic relationship to californium. A number of arguments[1] indicate that the probable mass assignment of the element-99 isotope is 253.

Alpha pulse analysis showed the presence of an energy group of 7.20 Mev and possibly another group (one-third the abundance of the 7.20-Mev group) approximately 170 kev lower in energy in the element-99 fraction. From the energies of these groups one may deduce that they represent isotopes of $Z>99$ in equilibrium with β^--emitting element-99 isotopes. The ratio of 6.64-Mev activity to the 7.20-Mev activity is about 5000. An alpha group of approximately 7.1 Mev was shown to elute from a Dowex 50 cation citrate column ahead of the 6.64-Mev alpha group, which is evidence for assigning at least one of the energetic groups to element 100. The short half-life of the ~7.1-Mev alpha group which eluted ahead of element 99 prevented a possible resolution of this group in the absence of the element-99 activity.

Several chemical experiments on elements 99 and 100, in addition to the previously mentioned cation citrate elution behavior, have been performed. Both elements carry on LaF_3 and $La(OH)_3$, and solvent extract into tributyl phosphate from a highly salted, dilute nitric acid solution. In concentrated hydrochloric acid, element 99 behaves like curium with both Dowex A-1 anion and Dowex 50 cation resins.

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* These elements (99 and 100) have previously been discovered in other work at Argonne National Laboratory, University of California Radiation Laboratory, and Los Alamos Scientific Laboratory, not yet published.

[1] A manuscript containing details of these experiments as well as a number of additional experiments is in preparation.

[2] The production of lighter isotopes of element 99 by nitrogen-ion bombardment of uranium has recently been described: Ghiorso, Harvey, Rossi, and Thompson, Phys. Rev. **93**, 257 (1954).

[3] B. H. Ketelle and G. E. Boyd, J. Am. Chem. Soc. **69**, 2800 (1947); K. Street, Jr., and G. T. Seaborg, J. Am. Chem. Soc. **72**, 2790 (1950).

[4] J. R. Huizenga, Phys. Rev. (to be published).

The Nuclear Spin of Si^{29}†

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SEVERAL attempts have been made to determine the spin of Si^{29} with either inconclusive,[1] conflicting,[2] or rather indirect[3] results. The consensus favors the value 1/2. Nuclear shell theory gives 1/2, 3/2, and 5/2 as the only possibilities.[1] We have eliminated values greater than 1, thereby establishing the spin of Si^{29} as 1/2. This was accomplished by resolving the multiplet structure produced in the fluorine nuclear magnetic resonance by the electron coupled interaction[4] of the Si^{29} spin with the F^{19} spins, using isotopically enriched $Si^{29}F_4$.

In pure liquid or gaseous $Si^{29}F_4$, the fluorine resonance should be a multiplet, with $2I(Si^{29})+1$ equally spaced components of the same intensity.[4] The SiF_4 actually used was prepared[5] from a sample of SiO_2 whose composition was given as 30, 69, and 1 percent, respectively, of Si^{28}, Si^{29}, and Si^{30}. The enriched sample of SiO_2 was supplied by the Oak Ridge National Laboratory. The