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I. Zvara, L. K. Tarasov, M. Krzhivanek, Su Hung-Gui, T. S. Zvarova

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Abstract

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I. Zvara, L. K. Tarasov, M. Krzhivanek, Su Hung-Gui, T. S. Zvarova

Formation of $Zr^{97}Cl_4$ during the slowing down of fission fragments in chlorine-containing gases

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Methods of radiochemical separation and purification based on differences in the volatility of the elements being separated or of their compounds are distinguished by their fundamental simplicity, the high efficiency of a single separation, and also by rapidity, which is often of special interest. However, such methods have not become widely used. Up to now, practically only the volatility of certain simple substances, oxides, and nonhydrolyzing halides has been employed. Apparently, the "development" of the rather numerous group of comparatively readily volatile higher halides of transition metals is very promising. Their disadvantage is their easy hydrolyzability.

The use of gaseous compounds to accelerate radiochemical separation should give an especially large effect in the case when these compounds themselves can be obtained by slowing down recoil atoms ("hot atoms") in a gaseous medium of suitable composition and transporting them by a gas stream.

The purpose of the present work was to study the conditions necessary for obtaining Zr^{97} of fission-fragment origin in the form of $ZrCl_4$ by slowing down fission fragments in a gas, and to carry out the transport of this substance in the gaseous state.

The experiments were carried out in an apparatus whose main part was a flow ampoule made of fluoroplastic-4, passing into a tube of the same material. A target was inserted into the ampoule; it consisted of a layer of highly enriched uranium protoxide-oxide deposited on a mica backing rolled into a tube. The layer of U_3O_8 was covered with a thin fluoroplastic film transmitting a sufficiently large fraction of fission fragments. The ampoule with the tube was placed in a thermostated oil bath. The target was irradiated with standard Po–Be neutron sources. A gas, usually containing a certain amount of vapors of inactive zirconium chloride carrier, was continuously blown through the ampoule. In the end part of the tube, emerging through a seal from the bath, the carrier condensed. The condensate was analyzed radiochemically for its Zr^{97} content. The duration of an experiment was 5-7 h.

To study the question of the formation of $Zr^{97}Cl_4$ during the interaction of fission fragments with a gaseous medium, we carried out a series of experiments with variation of the gas composition and temperature. The cylindrical shape of the target, combined with the use of an inert film to cover the U_3O_8 layer, ensured constancy of the geometrical efficiency of the target when the composition and temperature of the gas were changed, despite the fact that the dimensions of the gas space bounded by the target were comparable with the range of fission fragments in gases (1).

The results of experiments on the formation and transport of $Zr^{97}Cl_4$ are shown in Table 1. The activity of Zr^{97} in the condensate is reduced to identical irradiation and measurement conditions. The transport time represents the time of motion of the gas from the middle of the cavity with the target to the place where the carrier condensed. The vapor pressure of the $ZrCl_4$ carrier was 10–20% of its saturated vapor pressure at the experimental temperature (2,3). In the outgoing gases, Zr^{97} was not observed in a single case.

As is seen from these data, at temperatures $> 170°$ in gas mixtures containing $ZrCl_4$ vapors, fission-fragment Zr^{97} is stabilized in the form of zirconium tetrachloride.

Table 1.

Formation of $Zr^{97}Cl_4$ in gas mixtures of various composition and temperature

Principal components of mixture, vol. %	Vapor pressure of $ZrCl_4$, mm Hg	Bath temperature, °C	Activity of Zr^{97} in condensate, pulses/min	Transfer time, sec
Cl_2 10	0	20	0	30
Cl_2 10	10^{-4}	150	0	35
Cl_2 10	0.12	178	1550	33
Cl_2 47, N_2 53	0.04	173	1490	14
Cl_2 22, N_2 73	0.12	173	1810	20
Cl_2 17, N_2 88	0.08	173	1830	7
N_2 100	0.31	184	1760	18
N_2 100	0	174	0	24
Cl_2 52, air 48	0.16	173	1840	17
Cl_2 22, air 79	0.08	173	1650	20
Cl_2 3, air 97	0.03	173	1720	28

Total activity of Zr^{97} in Al foil: 2300.

An unexpected fact is that $Zr^{97}Cl_4$ is formed in gases of very different composition, even in the absence of free chlorine and with a high oxygen content in the mixture. This most likely indicates that the process of formation of $Zr^{97}Cl_4$ is based on exchange of a hot atom (ion) of Zr^{97} with a $ZrCl_4$ molecule. The data obtained do not permit a definite conclusion concerning the mechanism of formation of the labeled molecules. This question is the subject of our further investigations.

The "chemical yield" of the process of conversion of Zr^{97} fragments into $Zr^{97}Cl_4$ can be calculated only with some uncertainty. The total amount of Zr^{97} obtained from the target as a result of emission of fragments was found by absorbing all fragments in aluminum foil. The scatter of the data on the yield of $Zr^{97}Cl_4$ obtained at temperatures $> 170^\circ$ for diverse gas mixtures is apparently within the limits of measurement error. The mean value from these determinations is $\sim 75\%$ of the amount of Zr^{97} found in the aluminum foil. The independent yield of Zr^{97} should be somewhat less than half its cumulative yield in fission ([4]). Consequently, $Zr^{97}Cl_4$ is formed both from primary fragments of Zr^{97} and from atoms of Zr^{97} arising in the β-decay of Y^{97}. There are as yet no data for deciding with which of the two processes of formation of Zr^{97} some incompleteness of its conversion into $Zr^{97}Cl_4$ is associated.

Fig. 1. Decay curve of the activity of the $ZrCl_4$ condensate. Solid curve—$T_{1/2} = 17$ h.

The $ZrCl_4$ condensate contains Zr^{97} of very high radiochemical purity. This is evidenced by the typical decay curve of the activity of condensate that had not been subjected to radiochemical purification (see Fig. 1). Only contamination with short-lived activity with a half-life of ~ 20 min is clearly observed. Its nature has not yet been established.

It is known that, under short irradiation, the relative activity of Zr^{97} in the sum of the fission fragments reaches a maximum, 8%, about ~ 30 h after irradiation ([5,6]). On the basis of this figure, one can estimate the lower limit of the purification coefficient of Zr^{97} from fission products in our experiments. It is equal to ~ 300. From the data on the composition of fission fragments it follows that,

that, by the proposed method, Zr^{97} is effectively purified from the isotopes Sr, Y, La, Ce, Pr, Te, J, Tc, Xe, and other elements.

The minimum transfer time in our experiments was 7 sec. In this case the chemical yield did not decrease. This fact is direct proof that the formation of $Zr^{97}Cl_4$ occurs within no more than 1-2 sec after the emission of the fragment or the decay of Y^{97}. If the assumption of a hot mechanism for the formation of $Zr^{97}Cl_4$ is correct, then the time for extracting Zr^{97} from fission products can be further substantially shortened.

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