ULTRA-FAST CHEMICAL SEPARATIONS ("PLASMA CHEMISTRY") WITH A HELIUM-JET RECOIL-TRANSPORT SYSTEM

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The transport efficiency through a long-capillary He-jet recoil-transport system can be changed for different elements by altering the nature and/or the concentration of the impurities in the He. Thus, it appears that a form of "plasma chemistry" is taking place in the formation of the "macromolecules" that transport the radioactive recoils through the system and the attachment of the recoils to chemically specific sites on these macromolecules. We describe some experimental results and point out the advantages of such a system; also, we note that it is possible to obtain information about the Z of a nuclide from these experiments.

1. Introduction

In the course of our recent work developing a helium-jet recoil-transport system (HeJRT)[1–4], we have made an interesting observation that may have important practical ramifications. It is generally accepted that the mechanism of transport of activities through long organically doped HeJRT systems (systems in which small amounts of benzene, methane, ethane, chloroform, or pump oil, etc. are added to the bulk helium supply) requires the combining of the activities with macromolecules generated from the organic material[5]. However, the nature of this combining or attachment is not generally understood.

We have some evidence that the chemical nature of both the recoil activity and the macromolecule are important, suggesting some form of chemical bond, as opposed to other mechanisms such as nucleation which do not reflect chemical specificity. We have further observed that this chemical specificity can be used to transport preferentially through the HeJRT system different components of a mixture of chemically differing recoil activities produced from the same target. In effect, in at least some cases, it is possible to achieve a partial chemical separation in the course of the thermalization and transportation process.

One possible explanation of this effect could be that the chemical natures of the recoil and macromolecule are effective in determining the macromolecule's ability to pick up and hold the recoil atoms. If this be the case, that component of a mixture of chemically different recoils which is preferentially picked up and held by macromolecules will, as a result, also be that component which is preferentially transported through the system. Those remaining components that are not readily picked up and held by the macromolecules will not have the benefit of the macromolecules' high transport efficiency through the system and will tend to become lost on the walls of the capillary leading to the counting area.

It is perhaps worth pointing out that, even if this explanation is in gross error, and the chemical nature of neither recoil nor macromolecule is the source of the effect, the effect is still real and can be used to achieve a preferential enhancement in the transport yield of chemically differing nuclear recoils in the HeJRT system. Even though only a partial chemical separation may be obtained, it can in many cases be used to determine the Z's of the nuclides being produced in a complex nuclear reaction. Coupling these effects with an on-line isotope separator following an HeJRT system[6,7] could thus produce a powerful means of identification of both the Z and A of short-lived nuclei far from β stability without even having to resort to information from X-rays, etc. Also, since this "plasma chemistry" is ultra-fast, depending only on the transit time of the HeJRT system (in our present system ≈ 200 ms), one can use it with very short-lived nuclei.

2. Experimental

Our experimental results fall into three classes: First, the transport efficiencies for *physically similar* species differ significantly; second, one could change, even reverse, the relative transport efficiencies of different elements by using differing dopants in the He; and, third, the absolute and relative efficiencies of different elements could be altered by varying the concentration of a dopant in the He. Our most significant results to date for these effects follow.

2.1. Transport efficiencies for physically similar recoils

Our first observation was simply that the experimental transport efficiency differed substantially for physically similar recoils. For example, for Co, Cu, and Zn, the total transport efficiencies in our system were 90, 75, and 50%, respectively. These efficiencies were determined on the basis of a direct comparison of the HeJRT performance (collecting on paper tape; 20 ppm benzene by volume in helium) with activities left following the irradiation of thin foils.

The reactions of interest were $^{65}Cu(p,3n)^{63}Zn$ $(t_{1/2} = 38.5$ min), $^{62}Ni(p,3n)^{60}Cu$ and $^{60}Ni(p,n)^{60}Cu$ $(t_{1/2} = 23.0$ min), and $^{58}Ni(p,\alpha n)^{54m}Co$ $(t_{1/2} = 1.5$ min). In each case a 30-MeV beam was used. The Cu foils (0.0025-mm thick natural Cu) were irradiated with, as nearly as possible, constant currents for 30 s (the currents were integrated), the Ni foils (0.0025-mm thick natural Ni) for 60 s. The targets were retrieved using a pneumatic tube ("rabbit") system. Eeach source was counted in the same geometry, for the same duration after the same time of bombardment, and with dead-time correction. The only uncertainty in the determination lies in the corrections for the range of the recoils in the target, taken from Northcliffe and Schilling[8]).

Following the experiments made using the rabbit, the same activities were collected using the HeJRT system. Similar copper and nickel foils were placed in the HeJRT target assembly. The pressure in this assembly was maintained at 1.0 atm using helium with an initial purity of > 99.995% which was doped with approximately 250 ppm benzene vapor. The capillary was made of polyethylene, had an inside diameter of 1.4 mm, and was just under 15 m in length. The activities were collected on a stationary piece of paper mounted in the chamber pumped to < 1000 μm pressure. As in the case of the rabbit runs, the targets were bombarded for 30 and 60 s for the copper and nickel foils, respectively, with a nearly constant current

of 30-MeV protons. After the end of the bombardment, the collection of the activities on the paper was continued for an additional minute to assure complete sweeping of activities from the target area. [Experiments carried out in this laboratory and elsewhere indicate that one obtains essentially 100% sticking of the deposits on paper tape, even when an acute angle of incidence is used[1, 5]).] Five minutes after the end of the bombardment a 15-min count of the γ radiation was taken using the same detector in the same geometry. Also, as in the case of the rabbit run, the photopeak areas were corrected for integrated current and analyzer dead time.

In both the rabbit and the HeJRT runs 0.0025-mm foils were used. However, for the efficiency determination the thickness of the target used in the HeJRT system was taken to be the maximum range of the recoils, i.e., the target thickness from which recoils were able to leave the target and enter the helium atmosphere. For the rabbit targets it was necessary, of course, to correct for the extra residual activity that could not have left the targets.

The efficiencies were determined to be 24% for ^{63}Zn, 49% for ^{60}Cu, and 60% for ^{54m}Co. However, it was subsequently learned that the efficiency of the HeJRT system increases as we proceed from 1 atm to 3 atm of helium, reaching 50% for ^{63}Zn, 75% for ^{60}Cu, and 90% for ^{54m}Co. It is difficult to explain these differing total transport efficiencies on the basis of the physical properties of the recoils (mass, ionic radius, ionization potential, etc.).

2.2. Changing transport efficiencies by changing impurities

The second observation was that the absolute transport efficiencies for a pair of simultaneously generated activities could be shifted in opposite directions upon changing the major macromolecule-generating component in the helium supply. In this experiment a natural aluminum target (100% ^{27}Al) was bombarded with 30-MeV protons to produce (2.1-s) ^{26}Si and (12-s) ^{23}Mg from (p,2n) and (p,αn) reactions, respectively. Gamma-ray spectra of the products were recorded with Ge(Li) detectors when the primary impurity in the helium was 1) benzene vapor, 2) compressed air (the active impurity probably being pump oil), and 3) when the level of impurities was very low. In each case there was a small amount of water vapor present. The concentration of benzene vapor used was roughly 0.01% to 0.1% by volume. The concentration of compressed air used was roughly 5%, and the water vapor was added by passing the helium

through a gas-washing bottle filled with water. Each time an impurity change was made the target assembly was pumped out and refilled two to three times with helium doped with the new impurity. However, when the main component of the impurities was changed from run to run it should be presumed that a small (and possibly significant) amount of the impurities from the previous run remained in the target assembly. This was particularly true in the runs where pure helium was used; here the transport efficiency did not drop nearly so much as it would have if no macro-molecule-generating impurities had remained.

The results of this experiment are shown in fig. 1. Error bars shown are the average deviations of the points. It is quite apparent that the absolute efficiencies of the HeJRT system for transporting different activities is dependent on the type of impurities in the helium. Magnesium is transported best when compressed air plus water vapor is used (the ratio of Si to Mg activities transported changes by nearly 40% on changing the types of impurities used to produce cluster molecules). Since the activities were made at the same time, under the same conditions, from the same target, the efficiency changes cannot be explained away as a half-life effect or as a reflection of any changes (other than the source of macromolecule-generating components) in the HeJRT system during the experiment.

2.3. Effects of concentration of impurities on transport efficiencies

Most recently, when operating the HeJRT system using ethylene as the primary impurity in the helium,
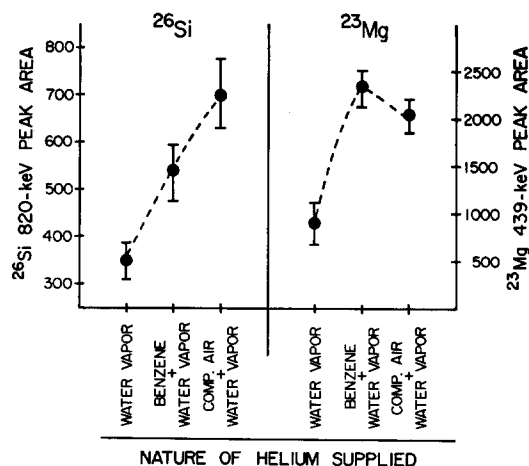
we have observed a third effect. Again using 30 MeV protons on aluminum to produce ^{26}Si and ^{23}Mg, we found that the ratio of ^{26}Si to ^{23}Mg transported through the system was dependent on ethylene concentration. Results are shown in fig. 2. Both ^{26}Si and ^{23}Mg efficiencies increase with increasing ethylene concentration in the range from 25 to 300 ppm by volume; however, the ^{23}Mg total transport efficiency increases much more rapidly in the low concentration range. The 7 points are the averages of a total of 29 determinations. The error bars in the plot of the ratio of peak areas to ethylene concentrations are just the average deviations of the points. The two points without error bars are points for which fewer than three determinations were made.

This behavior for the HeJRT system using ethylene as the source of macromolecule-generating material is particularly striking when it is contrasted with the system behavior when run using benzene. With benzene, the efficiency for transporting both ^{26}Si and ^{23}Mg through the system is constant to within the accuracy of the measurement throughout the entire range of
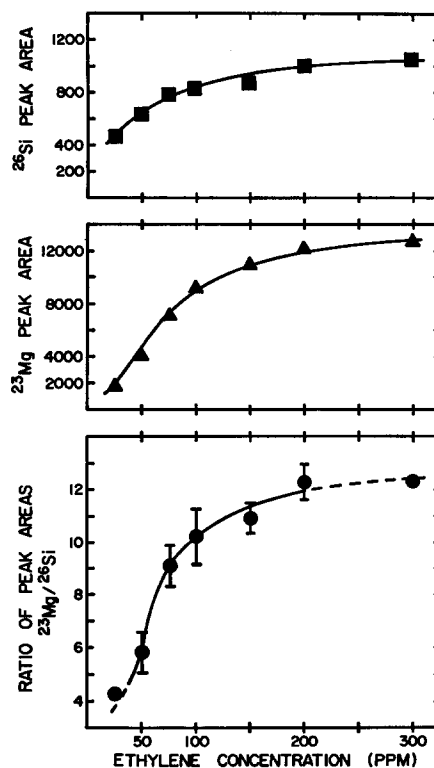

Fig. 1. Relative transport efficiencies for ^{26}Si and ^{23}Mg both made simultaneously from the same ^{27}Al target. By changing the major "macromolecule"-generating dopant, the relative transport efficiencies can be made to change.

Fig. 2. Effect of varying the ethylene concentration in the He supply on the total transport efficiency for ^{26}Si and ^{23}Mg, made simultaneously from the same ^{27}Al target.

benzene concentrations from $\lesssim 5$ to $\gtrsim 500$ ppm by volume[1]).

3. Discussion – Plasma chemistry

For the purposes of this discussion "plasma chemistry" is taken to mean chemistry occurring between thermalized recoils and macromolecules in the plasma in the target assembly for the purpose of achieving chemical separations. The evidence described in section 2 suggests it may be possible to accomplish separations of this nature for a fairly wide range of elements.

This possibility of being able to perform plasma chemistry gives some information about the mechanism through which the recoil becomes a part of the cluster molecule. It seems likely that the macromolecules are formed when impurities present in the helium condense out of the plasma in the target assembly (where "condense out" is not clearly defined and can be taken to be polymerization, nucleation, etc.). If during the process of condensation, the thermal recoil, because of its physical presence in the region of condensation, becomes trapped in the macromolecule, then plasma chemistry would not be possible. If, on the other hand, the thermal recoil becomes bound to some chemically active site on the macromolecule during or after the condensation process, the possibility of controlling the nature of these chemically active sites and thereby possibly achieving a separation of chemically differing recoils can be considered. That is to say, if it is possible to control the types of recoils that become attached to macromolecules, a separation of the recoils will be accomplished as a result of the poorer transport efficiency of the HeJRT system for those recoils not attached to macromolecules.

We feel this "chemically active site" hypothesis is supported by the observations reported above and

suggest that it would be profitable to follow this preliminary work with an in-depth study of performing plasma chemistry. Further, if this does become a reality, it will extend the realm of ultra-fast chemistry beyond those elements that easily form gaseous products in the HeJRT system, and the difficulty of collecting or trapping these gaseous products would be eliminated, since virtually any surface could be used for collecting the activities[1,5]). This type of chemical separation would also be compatible with several methods of on-line mass separation (identification) discussed in the literature[1,5,6]). This provides a possibility for both Z and A determination in low-resolution isotope-separator systems.

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