STUDIES OF SHORT-LIVED PRODUCTS OF SPALLATION FISSION REACTIONS AT TRIUMF

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Abstract. The gas-jet recoil transport technique has been used to transport products from spallation and fission reactions from a target chamber to a shielded location for nuclear spectroscopic studies. These involve X-β-γ coincidence measurements and (shortly) time-of-flight mass spectroscopy. It has been deduced that the proton beam at present intensities has no appreciable effect on the ability of ethylene and other cluster-producing gases to transport radioactivity. Preliminary results will be presented for shortlived fission products from uranium, and for spallation products of iodine and argon. The latter were obtained from the bombardment of gas and aerosol targets mixed with the transporting gas in the target chamber, which appears to be a generally useful technique.

1. Apparatus

A target chamber has been installed in Beam Line 4A at TRIUMF for the production of short-lived reaction products for nuclear spectroscopy studies. Proton beam currents up to 100 nanoamperes are currently available routinely, while the design limit for this line is 10 microamperes.

In the chamber, metal foil targets may be moved into the beam remotely, upstream of a variable gas volume in which recoiling product nuclei are thermalised. Gases, appropriately chosen for radioactivity transport properties, are fed into the chamber, the gas flow being directed so as to flush efficiently the region adjacent to the down-stream face of the target.

The gases leaving the chamber are conducted through a 25-metre long, 4-mm inside diameter polyethylene tube to a collection chamber. Here the gas stream impacts on to a plastic (computer) tape, the chamber being exhausted to 500 microns of Hg by a high capacity pump. Turnover time in the target chamber plus transit time in the capillary tube is between 1 and 2 seconds.

The radioactivity spot is moved on a predetermined schedule to a position between assemblages of X-, β- and γ- ray detectors, with appropriate Pb shielding to inhibit radiation scattering between the detectors. Signals from the detectors are measured in coincidence, and as a function of delay time, by a data acquisition system based on a PDP 15 computer, and recorded event-by-event on magnetic tape. Coincidence spectra are later obtained by computer analysis of the data.

Identification of the nuclei, responsible for the decay radiations observed, is at present effected by means of the X-rays emitted (giving Z) and the half-lives for decay (giving A for known nuclei). Commissioning is presently underway of a time-of-flight mass spectrometer system, patterned on a published design [1]), which will be applied to mass-measurement of nuclei recoiling from the decay events being studied.

2. Performance of the Gas-Jet System

Previous measurements have shown [2-6]) that the necessary ingredient for transport of radioactivity by a gas-jet system is the presence of molecular clusters in the gas stream. In the system

installed at TRIUMF, it is deduced that these clusters must have a diameter roughly in the range from 1 to 5 microns [7]). The lower limit is deduced from the observation that the radioactivity does deposit efficiently on surfaces on which the gas stream impinges; the upper limit is deduced from the observed absence of sedimentation in the capillary tube.

The first studies in this program had been made on products from the fission of ^{238}U induced by 14-MeV neutrons. These indicated a transport efficiency essentially constant for a constant cluster density in the gas stream, regardless of the gas or liquid used as the source of clusters. When the experiments were to be extended to use of TRIUMF proton beams, it was not evident in advance whether the same constancy would be observed, or whether plasma-production by the beam might reduce the cluster concentration or otherwise lead to a selective chemical effect on transport efficiency.

In the measurements with neutrons, it had been shown [7]) that the variation of yield of radio-activity at the exit from the gas-jet system was related to the gas flow rate by:

$$n = n_o [1 - \exp(-\alpha\phi)] \qquad (1)$$

where ϕ is the flow rate of the gas carrying the clusters and n_o and α are constants. This relationship is shown in Figure 1 as the top curve, the corresponding data being shown as solid points.
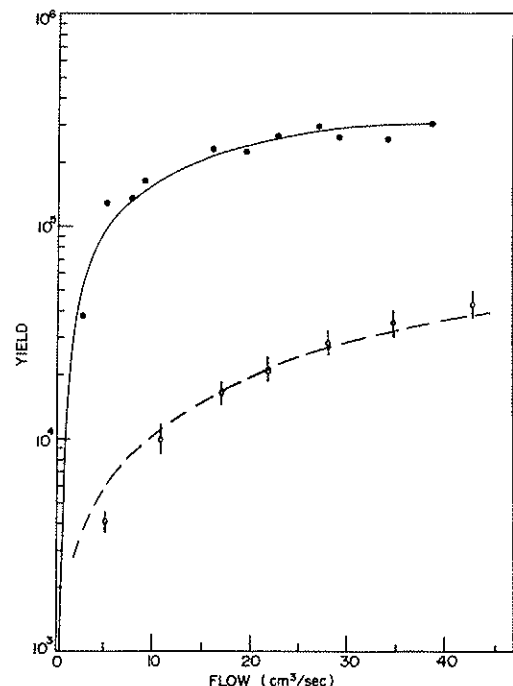

Figure 1. Yield of fission product radioactivity transported by the gas-jet as a function of gas flow-rate. Solid curve - equation (1) from the text. Solid points - experimental data from $^{238}U + 14$-MeV neutrons. Broken curve - equation (2) from the text normalised to equation (1) at a flow rate of 22 cm^3. sec^{-1}. Open points - experimental data from $^{238}U + 500$ - MeV protons.

This functional dependence was demonstrated, while maintaining the pressures and total gas velocities constant, by means of two ethylene reservoirs (one at pressure and temperature conditions such that the gas contained clusters and the other not), the gases from which were mixed in appropriate ratios so that only the cluster concentration changed.

The same technique was employed in experiments on products from the proton-induced fission of uranium; the expected functional form for yield as a function of flow rate is now expected to be:

$$n = n'_0 \left[1 - \exp(-\alpha r \phi) \right] \qquad (2)$$

where r is the ratio of the volumes of the target chambers. The lower curve in Figure 1 shows this relationship, and the observations in this experiment are shown as open points. The good agreement is suggestive of a similar gas-jet behaviour in proton bombardments as in neutron-bombardments, and presumably no very destructive plasma effects.

Confirmation of this was found in delivered yields of fission product activities from a uranium metal target foil of a magnitude quite satisfactory for nuclear spectroscopy measurements. Observed yields of spallation products from aluminum or silver metal target foils were found, however, to be much lower. This is expected for the presently available beam currents; the effective target thickness from which recoils into the gas phase will be obtained is no greater than the recoil range of nuclei concerned. Such ranges are very much smaller for spallation products than for fission products.

If, however, the target were gaseous, or were composed of an aerosol of which the droplet diameter was much smaller than the recoil range in question, the effective target thickness would then be expected to approach the total thickness of the gas column in the beam direction in the chamber, which was in the order of 10 mg. cm^{-2}. Yields were expected then to be appreciable.

An experiment was therefore conducted with argon gas, which, before feeding into the target chamber, was loaded with clusters, either by being mixed with ethylene gas or be being blown through an atomiser [7,8] containing methanol. The proton energy was 500 MeV. The respective γ-spectra observed by a single Ge(Li) detector from the deposits collected briefly at the end of the gas-jet system are shown in Figure 2; it is seen that useful yields of a number of the expected spallation products were obtained. Some differences between the two spectra may be interpretable in terms of specific chemical effects.

In a second experiment, nitrogen carrier gas was blown through an atomiser containing either iodomethane (mixed with methanol) or diiodomethane without methanol. The proton energy was again 500 MeV. The respective γ-spectra are shown in Figures 3 and 4. Again useful yields of spallation products, this time of iodine, are observed; once more, spectrum differences other than of just total yield are indicative of chemical selectivity in transport, although more work is required on this.

This technique for obtaining good yields of spallation products may be one of general usefulness, via (say) compounds of elements of interest dissolved in methanol or benzene, atomised into the gas stream. This is being explored. A further extension, as yet

Figure 2. Gamma-ray spectra observed with a 30-cm^3 Ge(Li) detector for spallation products from Ar + 500-MeV protons. The respective gas mixtures irradiated are indicated on the figure.

Figure 3. X-ray and low energy gamma-ray spectra observed with a 5-cm^3 Ge(Li) detector for spallation products from I + 500-MeV protons. The respective compounds irradiated are indicated on the figure.
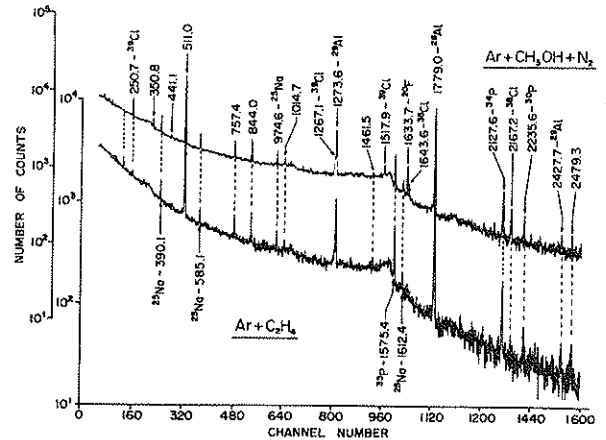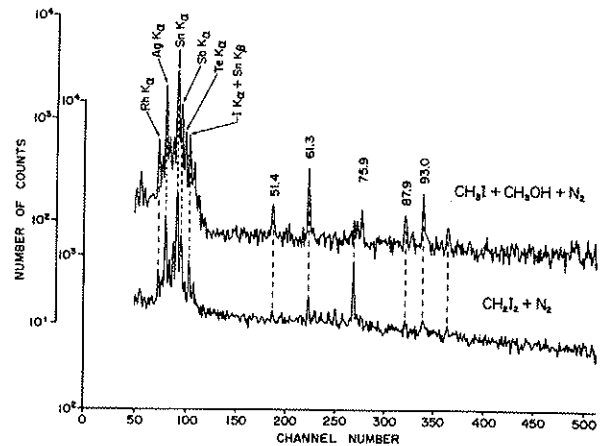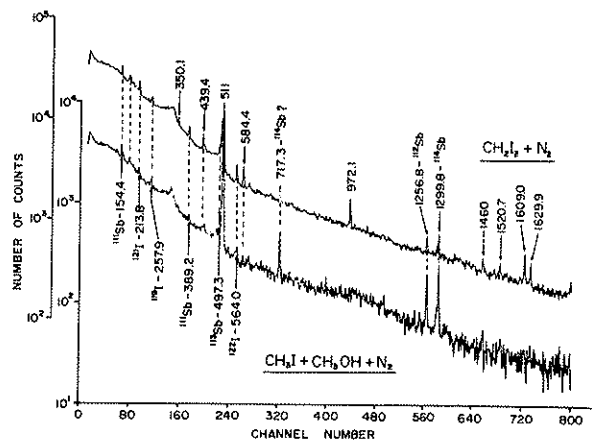

Figure 4. Gamma-ray spectra observed with a 40-cm^3 Ge(Li) detector for spallation products from I + 500-MeV protons. The respective compounds irradiated are indicated on the figure.

untried, might be to seed the gas stream with a fine powder, if one could be obtained with a particle size in the 1 to 5 micron range.

3. Time-of-flight Mass Spectrometer

The TOF spectrometer system is shown diagramatically in Figure 5. The gas stream from the exit of the gas-jet capillary will first impinge on a "skimmer" intended to separate the bulk of the gas load from the radioactivity-carrying clusters, which proceed on into the main chamber (where the gas pressure is 10^{-4} torr or better) and collect on an aluminised mylar foil held at a negative potential in the range of 15 to 20 kV.

Following a decay event (which will be detected by means of one or more radiation detectors), it has been shown [1] that a significant fraction of the recoiling nuclei leave the foil surface with very low energy and carrying a +1 charge. Such nuclei will be accelerated by the applied electrostatic field and detected with reasonable geometrical efficiency at the far end of the one-metre flight path, in the present case by means of a Chevron channeltron detector.

Figure 5. Time-of-flight spectrometer (schematic).

This system is presently being commissioned. Tests have been performed with a very thin specimen of ^{212}Pb activity and its daughters, prepared by recoil collection from the decay of ^{228}Th. A preliminary time-of-flight spectrum is shown in Figure 6, obtained in coincidence with emitted α-particles. The mass 208 recoils are clearly seen, together with those corresponding to A = 104 (ie: charge of +2), and also to A = 1, 16 and others – perhaps arising from the mylar support. A peak (indicated by an arrow) with a time-of-flight corresponding to an unaccelerated nucleus recoiling from α-decay is observed in addition.

Problems are anticipated in coupling the TOF system to the present gas-jet system, utilising relatively heavy transport gases (such as ethylene) rather than helium, for which the skimmer principle works more effectively. This and other problems are currently under study.

4. Preliminary Spectroscopic Data

A substantial series of measurements has been conducted on the fission products of ^{238}U bombarded by 14-MeV neutrons (from the SFU D-T neutron generator) and by 300-MeV and 500-MeV protons. X-ray and low energy γ-ray measurements were made by a 5-cm³

Figure 6. Time-of-flight spectrum from a ^{212}Pb source.

Ge(Li) detector, while higher energy γ-rays were measured by a 30-cm³ Ge(Li) detector. Conventional coincidence detection electronics with a resolving time of 300 nsec were used to detect coincidence events, which were then used to trigger data acquisition. In studies of particularly short-lived nuclides, radioactivity collection times were typically 20 seconds, and data were subsequently taken for 10 intervals of 2 seconds each.

Spectra have been analysed by conventional computer techniques; the decay in time of spectrum peak areas was analysed to extract half-lives via computer fitting to experimental decay functions.

Typical data from among the first results obtained only are presented here. Figure 7 shows single detector X-ray and low-energy γ-ray spectra obtained for the products of neutron-induced and proton-induced ^{238}U fission respectively.

Figure 7. X-ray and low energy gamma-ray spectra obtained for fission products from ^{238}U + 14-MeV neutrons and from ^{238}U + 500-MeV protons respectively.

The conspicuous differences between these spectra evidently arise from shifts in the yield patterns when bombarding energies are raised from 14 to 500 MeV, and particularly as the yields of products from near-symmetric fission increase.

About 150 or 70% of the peaks in such spectra have been identified on the basis of energy, half-life, and coincidence relationships. A typical γ-ray

spectrum in coincidence with an X-ray peak of
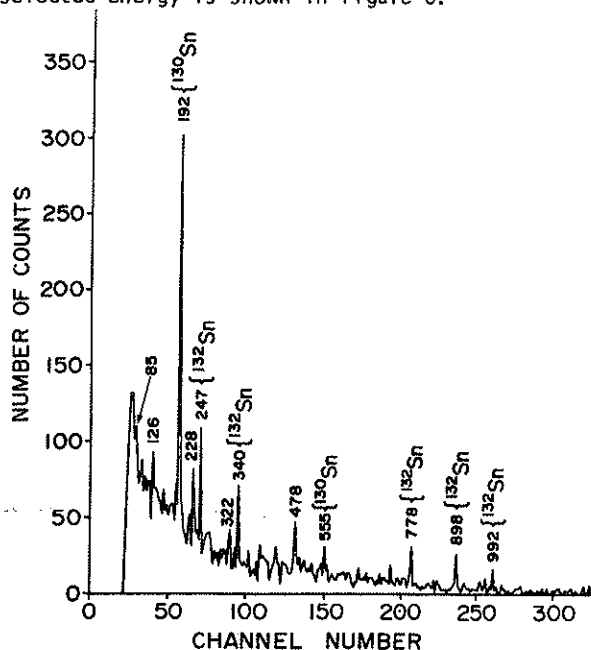selected energy is shown in Figure 8.

Figure 8. *Gamma-ray spectrum observed in coinci-
dence with Sb X-rays for fission products from*
^{238}U.

Most of the identifications are in good agreement
with recently reported results [9]. Particular
attention in the present study has been focussed on
the decay of neutron excess Mo, Tc and Sn nuclides,
(selected by X-ray emission from daughter nuclides)
and the analysis is continuing.

A second series of measurements has been pursued on
the decay of neutron deficient Te nuclides (also
via Sb X-ray emission) produced as spallation
products of iodine by the above mentioned
techniques. The data analysis is much less
advanced in this case, and results will be reported
elsewhere.

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