ON-LINE INVESTIGATION OF CYCLOTRON-PRODUCED FISSION PRODUCTS:

TARGET-ION SOURCE OPTIONS

ROBERT A NAUMANN

Dartmouth College, Hanover, New Hampshire, USA.

The properties of neutron-rich nuclei are of central interest to both nuclear structure physics and astrophysics. Experimental data concerning neutron-rich nuclei afford opportunities to test and refine current nuclear structure theories; such data are also needed to further the understanding of the r-processes – the last stages of stellar evolution.

The fission of a heavy nuclear target such as plutonium-242, uranium-238 or thorium-232 affords the only convenient method for synthesis and study of the shortest-lived neutron-rich nuclei.

Laboratories equipped with cyclotrons of moderate size providing light ion beams such as protons or alphas in the 30 to 50 Mev range, are particularly well suited to accommodate experimental investigations of a wide range of neutron-rich nuclei.

It is known that fission of uranium or thorium by light charged particles in this energy range results in a fairly uniform distribution of fission product yields. Figure 1, taken from the work of A. S. Newton¹, shows the isobaric yields of fission products when thorium is irradiated with 38 Mev helium ions. Unlike the sharp double-peaked distribution resulting from slow neutron-induced fission, the higher energy fission results in a broad, essentially flat, yield of products over the mass number range A=80 to A=155. This distribution includes isotopes of elements with atomic numbers lying between Z=31 to Z=60 (Ga to Nd).

Separation of this complex mixture of isotopes according to mass number is a necessary pre-requisite to meaningful nuclear investigations. Since the primary fission product nuclei have typical half-lives in the range of seconds, it is advantageous to operate a mass separator in a continuous on-line mode to accomplish this task.

A recently developed mass separation method² exploits the recoiling charged product ions formed by the nuclear reaction. However, this very fast technique provides lower activity levels due to the necessarily thin targets that are required.

The conventional technique typically involves a relatively non-volatile thick target held at high temperature. During bombardment, the more volatile radioactive product nuclei are vaporized from the target and pass to the continuously operating ion-source of the electromagnetic

mass separator. After mass dispersion, the ion beam corresponding to the radioactive isobars of interest impinges on a moving tape collector. The tape is advanced, in steps, to transport the collected samples to the measuring equipment – typically radiation spectrometers. The stepping-time is chosen to optimize observation of the isobaric activity of particular interest.

At least three qualities of the target-ion source combination are desirable if short-lived isotopes are to be mass-separated and studied:

First: The target—ion source should operate at the highest possible temperature. High temperature exponent-ially affects the diffusion rate of the trace product atoms within the solid target, their diffusion through the target boundary surface(s) and the probability that the product atoms will be in the gaseous discharge region for ionization.

<u>Second</u>: Two different substances are necessary for ion source construction; a conductor and an insulator. The limiting temperature at which the ion source may operate is primarily limited by the melting points of these two materials. It is also important that the components have a reasonably low vapour pressure (<2 exp-4 torr) at the operating temperature. This enables reasonable efficiencies for ionisation of the trace atoms by the ion source.

Third: The conducting and insulating materials must be chemically stable, i.e. no chemical reaction should take place between them at the ion source operating temperature.

The metal with the highest melting point³ is tungsten, M.P. 3422°C. This exceeds the melting point³ of thorium dioxide, ThO₂, M.P. 3220 \pm 50°C. We propose to use thorium both as the fission cyclotron target and the insulator.

A feasible high temperature appears to be 2800° C. At this temperature the vapor pressure of tungsten³ is 1.4 exp(-4) torr. Since this is 600°C below the melting point, structural integrity of the metal should be maintained. Thorium dioxide is used for fabrication of high-temperature crucibles, so production of thoria insulators with particular shapes appears reasonable.

The stability of the thorium dioxide components in contact with tungsten metal is important. At 2800°C, using the temperature data from ref. 3, one estimates the standard free energies of formation for tungsten dioxide as:

 $\Delta G^0_+(WO_2) = -51 \text{ kJ/mole}$ and thorium dioxide as: $\Delta G^0_+(ThO_2) = -670 \text{ kJ/mole}$. Accordingly, thorium dioxide should be stable in contact with tungsten metal at this temperature.

It is of interest to consider which elements should be volatilized at the suggested 2800°C operating temperature. Over the range of the fission product elements Z=31 to Z=60, all have listed normal boiling points below 2800°C, except for the refractory metals Z=40 tot Z=45, i.e., Zr, Nb, Mo, Tc, Ru and Rh. To separate isotopes of these six elements special chemical vaporization techniques appear necessary. As an example, one may cite the ionization of molybdenum using chlorine both as chlorinating agent and as ion source support gas⁴.

A schematic for the proposed high temperature ion source is shown in figure 2. It is based on the Nielsen oscillating electron ion source⁵. The tungsten metallic electrodes are shown in black; the thorium dioxide insulators in cross-hatched white. The inside diameter of the cylindrical tungsten anode is 2.5cm, while the overall length from tip to insulating back-plate is 8.5cm. The target, a thorium dioxide rod, is situated on the axis of the ion source and surrounded by the heated tungsten cathode-coil. The cyclotron beam enters through openings in the three tungsten heat shields. It then passes through a thin tungsten plasma-confining window and strikes the target rod. This arrangement insures that the thorium target is as the same temperature as the ion source. A major advantage is that the complications of a separate target and connecting transport tube are avoided.

Several hurdles can be foreseen in the implementation of such an ion source design. First and foremost, the fabrication of the thorium dioxide insulating components may prove difficult, given the present regulatory conditions governing even naturally occurring alpha-emitters. Secondly, one needs experience with susceptibility of thorium dioxide components to thermal stress and fracture. Finally, one must be concerned with sintering effects in the thorium dioxide targets at high temperatures and the consequent reduction in effusion rates for the volatile fission products.

Successful operation of such a high temperature source will be rewarded by many new experimental opportunities.

References

- 1. A S Newton, Phys. Rev. 75, 17 (1949).
- 2. J Arje et al., Phys. Rev. Lett. 54, 99 (1985).
- 3. Handbook of Chemistry and Physics 76th Edition, CRC Press (1995).
- 4. Alburger, D and Thulin, S, Phys. Rev. 89, 1146 (1953).
- Almen, O and Nielsen, K O, Nucl. Instrum. & Methods 1, 302 (1957).



Fig. 1 Yields of fission products from A. S. Newton (ref.1)





F = Filament Leads H = Heat ShieldsW = Tungsten Window for Cyclotron Beam