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A TARGET FOR PRODUCTION OF RADIOXENONS

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TRIUMF

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SUMMARY

A liquid cesium target has been developed which allows the production and separate identification of the neutron deficient isotopes of xenon. The present report describes irradiations utilizing 34 to 41 MeV protons to produce millicurie quantities of Xe-127 and Xe-129m. At higher energies, however, the target could be used without modification to produce xenon isotopes as light as 119.

INTRODUCTION

The practical application of radioxenons in nuclear medicine provides the motivation of a study of the targetry appropriate for their production using high current proton accelerators. Xenon-123 is medically the most important radioxenon, because it is the parent of I-123, which is used directly in thyroid studies and in labelling many pharmaceutical compounds. Xe-125 is similarly valuable as the parent of I-125, while Xe-127 is used directly in lung-imaging studies. If available, Xe-129m may also prove useful for such studies. In addition, Xe-129m is of interest in that it exhibits a Mossbauer transition. Previous Mossbauer studies (ref. 1) utilized rather weak sources produced by reactor irradiation of natural xenon, followed by mass separation.

TARGET CONSTRUCTION

In the present work a 6 cm diameter disk, 1.2 cm thick is filled with liquid cesium. This target was irradiated with photons from the Lewis Research Center's variable energy cyclotron. The proton beam enters normal to one of the flat faces of the target. For protons of the energy available here the target is thick, with the beam coming to rest in the target. This target has several obvious advantages relative to a solid target:

1) The liquid metal does not suffer radiation damage.
2) The heating caused by the beam can be carried away from the target by the evaporation of cesium.
3) The radioxenons can be removed from the target by a stream of carrier gas so that chemical separations are unnecessary.
4) The target is indefinitely useful.

EXPERIMENTAL PROCEDURE

The liquid cesium target has been bombarded by protons at selected energies between 28 and 40 MeV with beam currents from 2 to 5 microamperes. The

irradiations were short (approximately 5 minutes) so that the dynamic response of the target could be studied. The radioxenons were carried from the target by a continuous flow of helium gas (about 30 ml per minute). The experimental arrangement is shown in Fig. 1. The target is connected by 20 meters of 6.2 mm OD tubing to two cold traps, arranged in series.

Each trap consists of a section of tubing immersed in liquid nitrogen. In addition the second trap contained a molecular sieve. Two Ge(Li) detectors were used to observe the gamma radiation as radioactive products accumulated in each trap, for a period of several hours after each run.

Analyses of the gamma ray spectra were carried out using the cyclotron computer system to fit Gaussian line shapes to the peaks after an appropriate background subtraction. For more complex spectra, consisting of both Xe-127 and Xe-129m, (such as observed at 35.7 MeV) a pure Xe-129m spectrum (recorded at E_p = 32.0 MeV) was subtracted, with proper normalization, to permit a more accurate analysis of the Xe-127 radiations. This procedure is illustrated in Fig. 2.

RESULTS

The production of Xe-129m as indicated by the presence of a 196 keV line in the gamma ray spectra was observed in all bombardments (27.3 MeV $\leq E_p \leq$ 40.2 MeV). Similarly, at proton energies above 33 MeV, the presence of Xe-127, as evidenced by a 203 keV gamma, was detected. Both the Xe-129m and Xe-127 activities were observed to require more than one hour for complete removal from the target and accumulation in the first trap. Both activities accumulated with the same time constant. This is shown in Fig. 3, where counting rates for the 196 keV and 203 keV gamma rays are shown as a function of time after bombardment. It was observed that this collection time could be decreased by approximately a factor of two by raising the target temperature prior to irradiation from 100° to 260° C.

In all irradiations, annihilation radiation was observed to accumulate in the first trap. A positron emitter arrived at the trap promptly after the start of the irradiation, was retained there temporarily as shown in Fig. 3, and then appeared to diffuse into the second trap with a mean life time for retention of approximately 26 minutes. This activity was permanently trapped in the molecular sieve, where its nuclear decay lifetime was determined to be 21 minutes. This activity is presumed to be due to ^{11}CO, which arises from oxygen impurity in the target. The reactions would be:

$$^{16}O(p;\alpha,n,p)^{11}C$$

and

$$^{11}C + Cs_2O \rightarrow 2Cs + {}^{11}CO$$

The thick target yields of Xe-129m and Xe-127 are given in Fig. 4 in microcuries/microamp-hour for a number of proton energies. These data have been used to obtain cross sections for the reactions that yield Xe-129m and Xe-127. The difference in yields at two successive energies is assumed to be

due to reactions taking place in a thickness of the target ΔX, where ΔX is the difference in range of protons at the two energies. This ΔX can be converted to an areal density of target nuclei, which together with the number of incident protons and the increase in the yield of radioxenon permits the calculation of a cross section. These results are shown in Fig. 5, plotted at the midpoint of each energy interval.

The absolute values of the yield are estimated to have an uncertainty of about 30%. The relative uncertainty in yield from one energy to the next is estimated to be about 10%. The principle source of error is the uncertainty in solid angle between source and detector. The location of the radioxenon frozen on the walls of the cold trap could fluctuate several centimeters as the liquid nitrogen level changed due to boil-off during the experiment. The ratio of the yield of Xe-127 to that of Xe-129m at any one energy is the most reliable number reported. The uncertainty in this value is due to the possible error in the values used for the number of photons (196 and 203 keV) per disintegration. For the 203 keV gamma, the value was taken to be 0.66. For the 196.7 keV gamma of Xe-129m the value 0.043 was used. These are based on data given in Nuclear Data Tables (ref. 2).

DISCUSSION

The most interesting features of the data are:

1) Both Xe-127 and Xe-129m accumulate on the target with the same time constant.

2) The cross sections for production of Xe-127 and Xe-129m are comparable in magnitude and quite large. The yield of Xe-131m was not measured but was observed to be at least two orders of magnitude smaller.

3) There is a considerable difference in the rate of increase of the yield with increasing proton energy for the two isotopes.

4) The difference in the threshold for production of the two isotopes (differing by two neutrons) is approximately 10 MeV, even though the binding energy for a single neutron is approximately 9 MeV.

The fact that both isotopes have the same collection time indicates that this time is characteristic of the target and transport system, rather than the nuclear lifetimes involved in the production. In addition, if either isotope were produced from the beta decay of an intermediate parent, those lifetimes would be either 32 hours or 6 hours for Cs-129 and Cs-127, respectively. The delay in removal of the activity from the cesium target is presumed to be due to a chemical adsorption or combination of the radioxenon with cesium. The adsorption hypothesis is obviously consistent with the decrease in collection time which was observed at higher temperatures. If on the other hand the delay is due to the formation of a CsXe molecule, it must be of a nature such that its rate of dissociation increases with temperature. The possibility of the formation of the CsXe molecule is supported by the fact that both Cs_2^* and Xe_2^* molecules exist, although both species are stable only in an excited molecular state.

The fact that the isotopes are produced directly rather than through a long-lived precursor means that either a (p;xn,yp) or (p;α,xn) reaction must take

place. For Xe-127, the former reaction (x = 5, y = 2) has Q = -47 MeV, while for Xe-129m (x = 3, y = 2) A = -31 MeV. In both cases the proton energies employed are too low for such a reaction to be the dominant process in accounting for cross sections as large as those observed. Consequently, one must conclude that alpha particle emission is an important factor in the production of both Xe-127 and Xe-129m, whether the alpha particle is considered to preexist in the nucleus or whether it is considered to form after emission of its components. Reactions of the type (p;α,x) have been studied (ref. 3) for medium A nuclei at 39 MeV; however, x was not identified in the experiment. No information is available for the (p;α,n) and (p;α,3n) reactions. A recent study (ref. 4) of the (d,^{6}Li) reaction, an alpha particle pickup, showed a reduced cross section for nuclei with A = 130. This was attributed to subshell closure. The present study would indicate that such effects have disappeared by A = 134.

The difference in the shape of the cross section curves for the production of the two isotopes may be attributed in part at least to the vastly different natures of the final states. Xe-129m, with J = 11/2 is likely only formed through rather high spin intermediate states, while Xe-127, with J = 1/2 would be formed only through low spin states, with the yield curves reflecting the density of each type of intermediate state. The large cross sections observed for production of Xe-129m at 40.2 MeV is probably due to the opening of another reaction channel. At 43 MeV it is possible for the reaction (p;3n,2p) to occur. It is still, however, not far enough above threshold for this reaction to make a large contribution. A more reasonable possibility is that Ba-133 becomes the intermediate nucleus rather than Xe-130. That is, the neutron could be emitted before the alpha particle.

CONCLUSIONS

The thick target yields of Xe-127 and Xe-129m were measured for a number of incident proton energies. These data were also used to obtain cross sections.

It is concluded that a liquid cesium target is practical for routine production of radioxenons using high current proton accelerators. The temperature of the liquid cesium should be at least 300° C in order to obtain prompt removal of the xenon products from the target. This consideration would be particularly important for Xe-123 where the 2 hr half-life would give significant reduction in yield if allowed to decay to I-123 which would chemically combine with cesium and become unrecoverable.

The subtraction process of separating gamma ray spectra for analysis should prove even more useful at higher energies, where spectra will become more complex and will arise from a greater number of radioactive isotopes.

REFERENCES

1. J. S. Geiger, R. L. Graham and F. Brown, Can. J. Phys. 40, 1258 (1962).

2. Nuclear Data Sheets 8, 138 (1972).

3. F. E. Bertrand and R. W. Peelle, Phys. Rev. C 8, 1045 (1973).

4. F. D. Becchetti, et al., Phys. Rev. Lett. 34, 225 (1975).

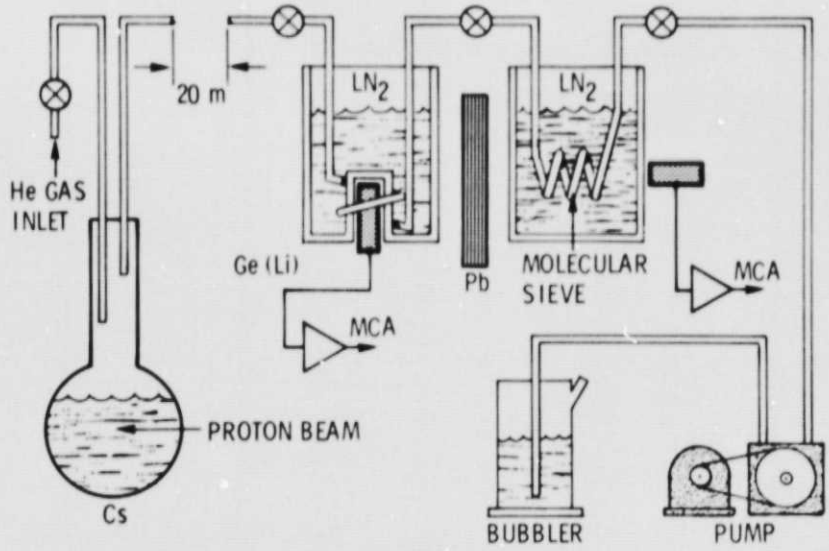

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Figure 1. - Cesium target system.

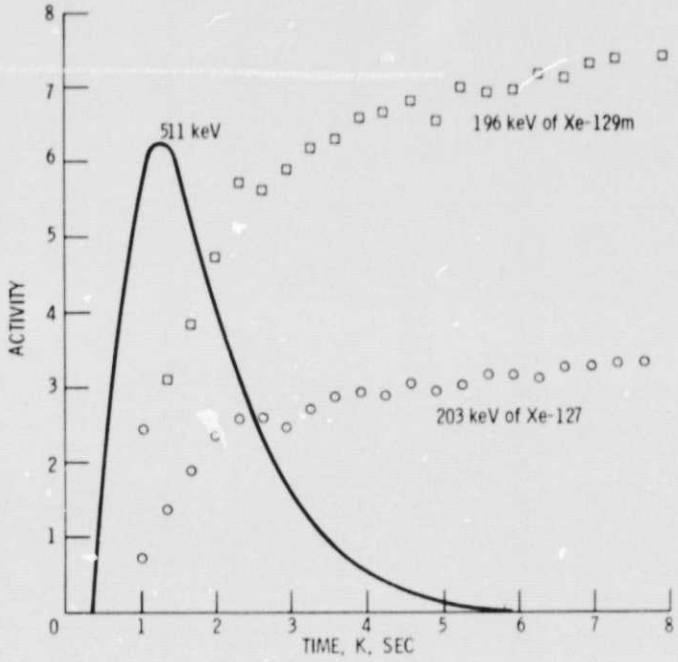

Figure 2. - Time dependence of photopeaks.

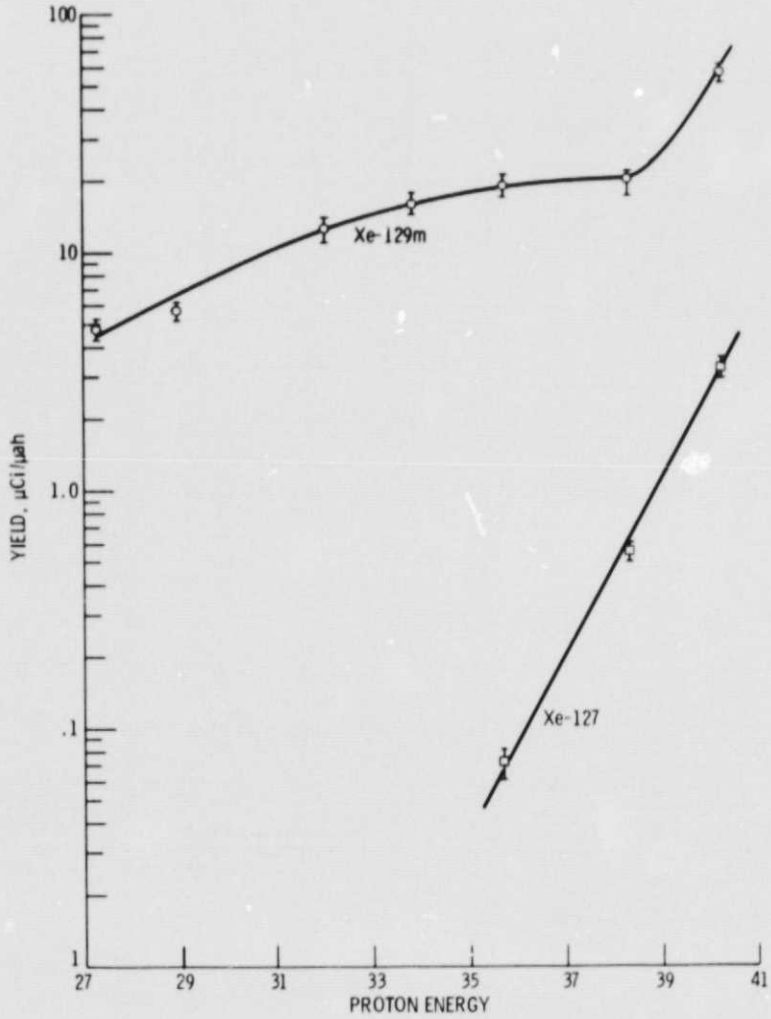

Figure 3. - Yield vs. energy.

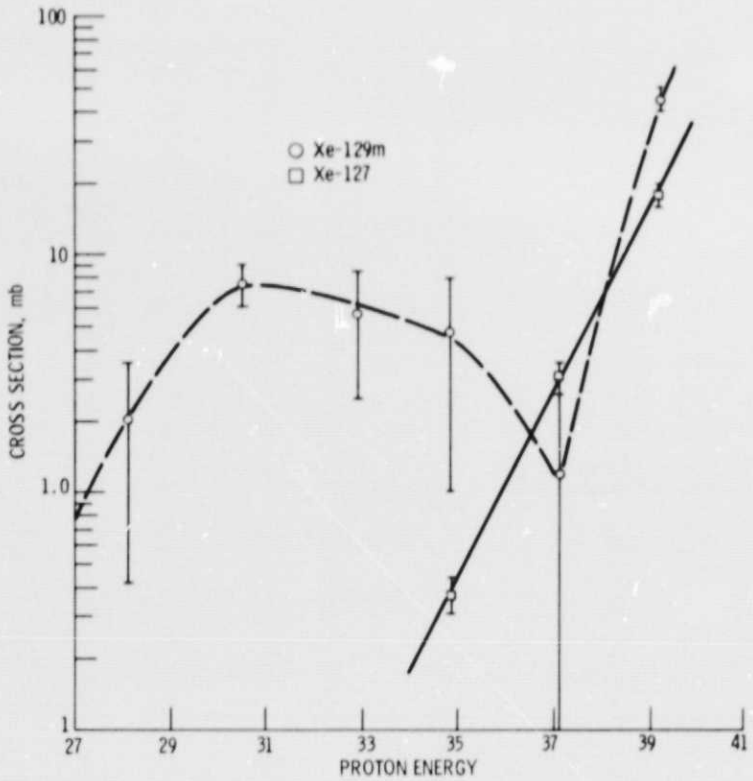

Figure 4. - Cross section vs. proton energy.

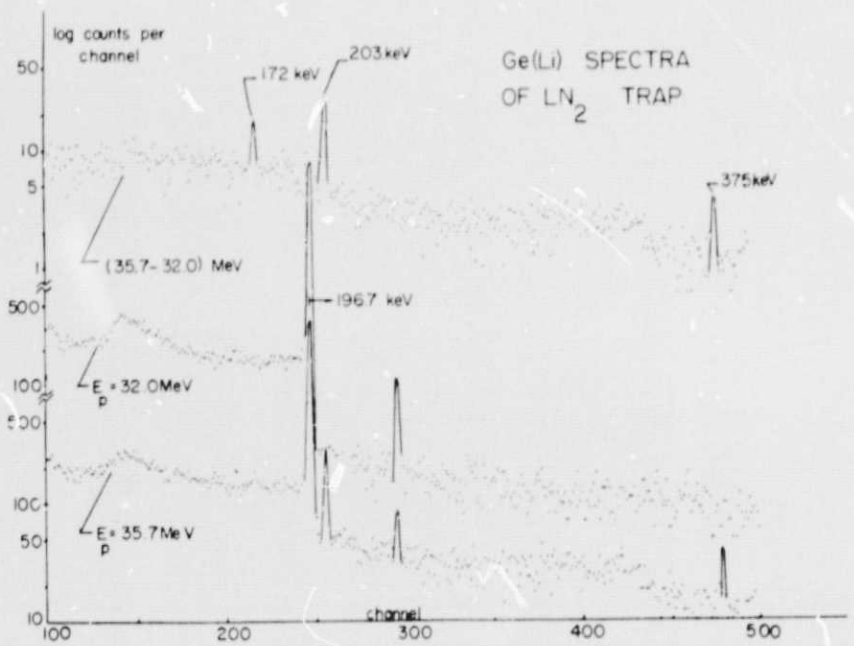

Figure 5.