in view of the absorption spectrum in Fig. 1(a). The α-ethanol radical shows a strong absorption at the 2537 Å. line of the mercury spectrum.

Acknowledgment.—We are indebted to a number of our colleagues for their extensive assistance during this investigation. In particular we wish to thank Mr. Douglas Harter for his technical assistance. Mr. C. Plucinski has kindly performed all the mass spectrometric analyses. The entire investigation was dependent upon Mr. B. E. Clifft in connection with the operation of the linac. It is a pleasure to acknowledge helpful discussions with Dr. A. O. Allen of Brookhaven National Laboratory concerning the material balance considerations.

[CONTRIBUTION FROM THE DEPARTMENT OF CHEMISTRY, UNIVERSITY OF ROCHESTER, ROCHESTER, N. Y.]

Excitation Functions of the (p,pn) and (p,2p) Reactions on Te^{130} at 60–233 Mev.[1a]

BY DEAN W. MAURER[1b] AND EDWIN O. WIIG

RECEIVED MAY 18, 1962

The cross sections of the reactions $Te^{130}(p,pn)Te^{129m}$, $Te^{130}(p,pn)Te^{129g}$ and $Te^{130}(p,2p)Sb^{129}$ have been determined for protons of 60, 120, 180 and 233 Mev. energy. The observed yields of Te^{129} and Sb^{129} are compared with those predicted by interpolation of Monte Carlo cascade calculations on Ru^{100} and Ce^{140} at 83, 238 and 368 Mev. followed by evaporation calculations based on data for Ce^{142}. There is good agreement in shape and absolute magnitude for the (p,pn) excitation function but poor agreement for the (p,2p) reaction. In the (p,pn) reaction the low-spin Te^{129g} isomer is found to be favored over high-spin Te^{129m}, in agreement with the predictions of Häller and Rudstam.

Introduction

The general shape of the excitation function for a given, simple spallation reaction brought about by moderate and high energy protons, results from the linear addition of the yields of the individual reaction steps which give rise to this product. The relative weight or importance of each reaction step and how this changes with the energy of the incoming particle depends upon various nuclear parameters such as the shape of the nuclear potential well, the distribution and density of nucleons in the nucleus, the interaction of the particles, etc. Recent Monte Carlo calculations[2] of the fast cascade phase of such reactions based on a crude model of the nucleus have made possible an estimation of the weight and variation with energy of the individual reaction steps. By comparing the resulting net excitation function for simple reactions, *i.e.*, those involving the loss of only a few nucleons, with experimental results, a much clearer picture of the reaction may be obtained. Furthermore, deviations from the experimental results may indicate changes to be made in the nuclear model.

An investigation of the (p,pn) and (p,2p) reactions on Te^{130} was undertaken to add to the meager data available[3,4] in the intermediate mass range for comparison of cross sections of such reactions with cross sections obtained from Monte Carlo and evaporation calculations. This region is of special interest since the only agreement in absolute value for the (p,pn) cross section was obtained here by Ware and Wiig[4] on Ce^{140}. Natural tellurium was used, since Te^{129} and Sb^{129} can arise only from the heaviest isotope, Te^{130}.

Experimental

Tellurium[5] in the form of metal powder was bombarded in the internal beam of the University of Rochester 130-inch synchrocyclotron. The targets, as described earlier,[6] were prepared by packing 75–100 mg. of Te powder into a 5 × 20 × 1 mm. box made of 0.75 mil high purity aluminum foil. Two 0.75-mil aluminum foils were placed in front and two in back of the target envelope to serve as monitors of the beam by the known cross section of the Al^{27} (p,3pn) Na^{24} reaction.[7] Only the two inner foils were used as monitors since the gain and loss of Na^{24} from these foils would be the same.

All irradiations were carried out for 1 hour after which the monitor foils were separated from the target and reserved for Na^{24} determination. In general, the tellurium target was dissolved in HCl and HNO_3 and a known amount of antimony carrier was added. The solution was boiled to dryness several times to expel HNO_3 and finally diluted to 100 ml. with 3 *N* HCl. A 25-ml. aliquot was taken and the remainder of the solution saved to determine the target weight. Holdback carriers of tin, cadmium, indium and palladium were added and tellurium precipitated with SO_2. The tellurium was purified by standard techniques, mounted as the metal and counted. The antimony, which had remained in solution, was then precipitated with H_2S, purified, mounted as the metal, and counted.

All the samples were counted on a methane-flow beta proportional counter. The decay was followed for a period of 12 to 18 months. Owing to the many isotopes of tellurium and antimony produced, the decay curves were quite complex. The routine resolution of these curves was done by the method of Perkel[8] after the composition has been determined by normal procedures. This method required the calculation of a large number of exponential terms. To facilitate the work an IBM-650 computer was programmed to do the tedious part of the calculation. The method allows one to resolve the 34-day isomer of Te^{129} from the tail of the decay curve, the 72-min. isomer from all the other activities and 4.6-hr. Sb^{129} from all the other antimony activities.

The aluminum monitor foils were counted directly on the proportional counter to determine Na^{24}. Resolution of the resulting decay curves was quite simple.

After the counting samples had decayed sufficiently and the decay curves had been analyzed, the samples were dissolved and analyzed spectrophotometrically to determine the chemical yield. The antimony was determined by the

(1) (a) The investigation was supported in part by the U. S. Atomic Energy Commission. This report is based on a thesis submitted by Dean W. Maurer in partial fulfillment of the requirements for the degree of Doctor of Philosophy at the University of Rochester, 1959. (b) Bell Telephone Laboratories, Incorporated, Murray Hill, N. J.

(2) N. Metropolis, *et al.*, *Phys. Rev.*, **110**, 185 (1958).

(3) (a) A. A. Caretto, Jr., and G. Friedlander, *Phys. Rev.*, **110**, 1165 (1958); (b) P. P. Strohal and A. A. Caretto, Jr., *ibid.*, **121**, 1815 (1961).

(4) W. R. Ware and E. O. Wiig, *ibid.*, **122**, 1837 (1961).

(5) Matthey spectrographically standardized Te powder was obtained from Johnson, Matthey & Co., Ltd., 73/83, Hatton Gardon, London, ECI.

(6) R. W. Fink and E. O. Wiig, *Phys. Rev.*, **94**, 1357 (1954).

(7) H. G. Hicks, P. G. Stevenson and W. R. Nervik, *ibid.*, **102**, 1390 (1956).

(8) D. H. Perkel, *Nucleonics*, **15**, No. 6, 103 (1957).

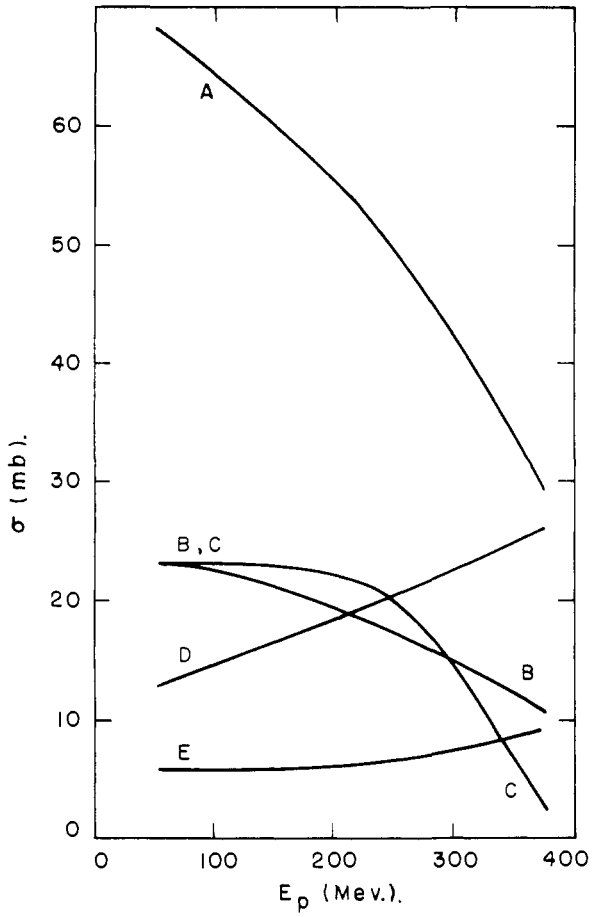

Fig. 1.—Cross sections of individual reaction steps predicted by the Monte Carlo cascade–evaporation calculations: A, neutron evaporation from Te^{130}; B, proton evaporation from Te^{130}; C, proton evaporation from I^{130}; D, (p,pn) by direct interaction; E, (p,2p) by direct interaction.

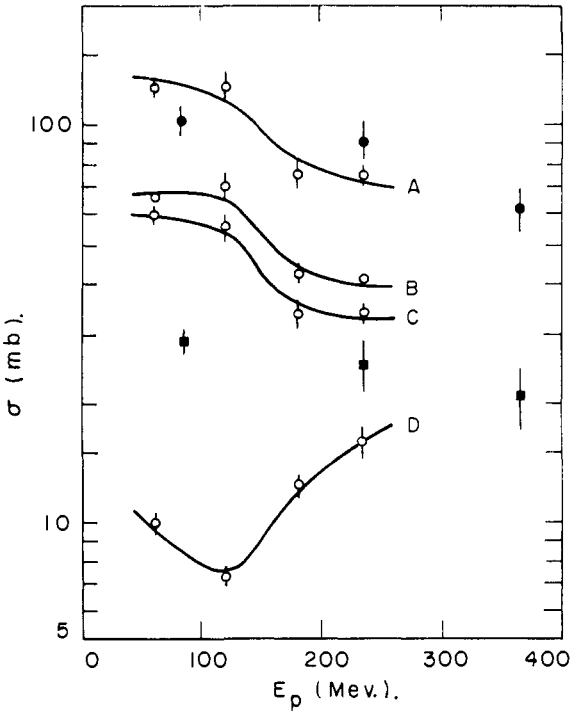

Fig. 2.—Experimental and calculated cross sections of the (p,pn) and (p,2p) reactions on Te^{130} as a function of energy: A, total (p,pn) reaction; B, (p,pn) reaction leading to the ground state; C, (p,pn) reaction leading to the metastable state; D, total (p,2p) reaction. The closed points are the calculated values; the open points are the experimental values. The errors on the experimental values are the standard deviations of the average of four determinations. The errors on the calculated points reflect only the statistics of the Monte Carlo calculation.

method of McChesney[9] and tellurium by the method of Hanson, *et al.*[10]

The cross sections for the reactions of interest were determined by taking into account the yield of Na^{24} from the monitor reaction, the growth and decay of Te^{129g}, Te^{129m} and Sb^{129} during bombardment, the contribution of Sb^{129} to Te^{129} before chemical separation, the chemical yields of tellurium and antimony, and the usual factors for calculation of the disintegration rate from the count rate.

The counter sample geometry factor was determined experimentally with a National Bureau of Standards calibrated P^{32} sample. The backscattering and air and window absorption factors were taken from curves of Weick[11] who determined these factors for a large number of different maximum energy beta groups on this particular counter. The self absorption factors were obtained from the curves of Nervik and Stevenson.[12]

Cascade–Evaporation Yields.—In order to compare the experimental cross sections with those to be expected theoretically, the contributions to the Monte Carlo cross sections[1] made by evaporation processes must be determined.

The (p,pn) reaction product can be obtained by several mechanisms: (1) by a direct knock-on or (p,pn) cascade process, (2) by a (p,p′) cascade reaction followed by the evaporation of a neutron, and (3) a (p,n) cascade reaction followed by the evaporation of a proton. The (p,2p) product can be obtained by a knock-on (p,2p) cascade process and by a (p,p′) cascade reaction followed by the evaporation of a proton.

Since Te^{130} was not one of the nuclides studied in the Monte Carlo calculations,[2] the residual excitation energy spectrum of the cascade products of interest for tellurium had to be interpolated from the data on Ce^{140} and Ru^{100}. This interpolation was carried out on the basis of mass number on the residual excitation energy spectrum for each product nucleus. This is a valid method because on the basis of the model used, the only difference between Ru and Ce is in the radius of the nucleus and the number of protons and neutrons. Since these are smoothly varying functions between the two nuclei, the change in the residual excitation energy should also be smooth. In view of the statistics on these spectra, no attempt was made to take into account the difference in proton to neutron ratio. The residual excitation energy data were kindly supplied by Dr. Turkevich.[13] As in the case of the (p,pn) and (p,2p) reactions[4] on Ce^{142} the evaporation calculation used was similar to that carried out by Caretto and Friedlander,[2a] except that 9 Mev. was used as the effective binding energy of the last nucleon instead of 10 Mev. and cases were considered for evaporation of one particle up to 49 Mev. of excitation energy rather than 40 Mev. The same barium-alpha compound nucleus results were also used for tellurium since these were the best data available for the evaporation calculation.

By summing the cross sections for all the contributing mechanisms shown in Fig. 1, the final calculated (p,pn) and (p,2p) cross sections given in Table I were obtained. The listed errors only reflect the statistics of the Monte Carlo calculation. No attempt was made to include possible errors arising from the barium-alpha data.

(9) E. W. McChesney, *Ind. Eng. Chem., Anal. Ed.*, **18**, 146 (1946).

(10) M. W. Hanson, W. C. Bradbury and J. K. Carlton, *Anal. Chem.*, **29**, 490 (1957).

(11) C. F. Weick, private communication.

(12) W. E. Nervik and P. C. Stevenson, *Nucleonics*, **10**, No. 3, 18 (1952).

(13) A. Turkevich, private communication.

TABLE I

CROSS SECTIONS OF (p,pn) AND (p,2p) REACTIONS ON Te^{130} FROM MONTE CARLO CASCADE–EVAPORATION CALCULATIONS

Reaction	Cross sections (mb.) 83 Mev.	235 Mev.	367 Mev.
$Te^{130}(p,pn)Te^{129}$	103 ± 8[a]	92 ± 10	61 ± 8
$Te^{130}(p,2p)Sb^{129}$	28 ± 3	25 ± 4	21 ± 4

[a] The errors listed are standard deviations and reflect only the statistics of the Monte Carlo calculation.

very similar ratios of 0.77, 0.67, 0.74, 1.06 and 0.70 for E_p = 49, 75, 103, 133 and 153 Mev., respectively, for the corresponding Zn^{69} isomers from $Zn^{70}(p,pn)$.

Discussion

Before discussing the detailed shape of the experimental excitation functions, the individual steps contributing to the over-all reaction mechanisms and their energy variation as predicted by the Monte Carlo calculation will be considered. In the direct interaction or knock-on reactions the incident proton interacts directly with one of the nucleons and both leave the nucleus with little or no further interaction. In order that the nucleus be left with too small an excitation energy to evaporate a particle, the nucleon hole left behind must not represent an excitation energy greater than the effective binding energy of the most loosely bound nucleon. The effective binding energy, *i.e.*, the energy below which particle evaporation is not very probable, was taken as 9 Mev. in the evaporation calculation. As the incident proton energy is increased, the probability of producing higher energy holes increases and the probability of a hole causing less than 9 Mev. of excitation energy is decreased.

The mean free path of nucleons in nuclear matter must also be considered. For low energy particles the struck nucleon must be close to the rim of the nucleus in order that both it and the incident particle escape without further interaction. As the incident energy is increased the mean free path increases and the impact parameter can be smaller and still allow the two nucleons to escape. In other words, there is a larger geometric volume in which the reaction can take place. This factor tends to cause an increase in the cross section, whereas the increase in the excitation energy deposited in the nucleus tends to cause a slight decrease in the cross section with increasing bombardment energy, the former being the stronger effect. The net result is a slowly increasing cross section with increasing energy as shown in curves D and E of Fig. 1.

In general, in the cascade plus evaporation reactions there is a single glancing collision of the incident particle with a nucleon that transfers an average of 20 Mev. of excitation energy to the nucleus, followed by the escape of the proton. Charge exchange may also occur, whereby a neutron is emitted in the cascade rather than a proton. The major factor governing the contribution of these steps to the total reaction is the probability of evaporating only one nucleon from the excited

TABLE II

EXPERIMENTAL CROSS SECTIONS OF (p,pn) AND (p,2p) REACTIONS ON Te^{130}

Reaction	Cross sections (mb.) 60 Mev.	120 Mev.	180 Mev.	233 Mev.
$Te^{130}(p,pn)Te^{129m}$	59 ± 4[a]	55 ± 4	33 ± 3	34 ± 2
$Te^{130}(p,pn)Te^{129g}$	66 ± 2	70 ± 5	43 ± 2	41 ± 1
$Te^{130}(p,pn)Te^{129}$ total	125 ± 4	126 ± 6	76 ± 4	75 ± 2
$Te^{130}(p,2p)Sb^{129}$	9.9 ± 0.5	7.2 ± 0.4	12.3 ± 0.6	15.8 ± 1.6

[a] The values are the average of four determinations. The errors are the standard deviations of the averages.

Results

The experimental cross sections obtained as a result of proton bombardment of tellurium are summarized in Table II and are plotted as a function of proton energy in Fig. 2. The value listed for each cross section is the average of four determinations and the errors are the standard deviations of the averages. Errors arise from a number of sources. There is usually an error of about 20% in the absolute cross sections caused by errors in determining counting corrections and in monitoring the beam. The error in these factors may be even greater since the decay schemes of the nuclides of interest are not completely known.

As can be seen from Fig. 2 the agreement between the experimental and calculated cross sections for the (p,pn) reaction is quite good, both in the general shape and in absolute values. A possible explanation for the detailed shape of the excitation functions is discussed later. The general shape predicted by cascade–evaporation calculations and found, for example, in the Cu^{65} (p,pn) excitation functions is a gradual decrease[14] in the cross section in going from 60 to 360 Mev. The decrease is not an order of magnitude. Agreement in absolute value is found in this work and in that[4] on Ce^{142}.

There is disagreement between the experimental and predicted cross sections for the (p,2p) reaction, both in general shape and in absolute value. The agreement in this case is somewhat poorer than was found[4] for Ce^{142} and there is an irregularity in the experimental curve.

In the present experiments the yields of isomeric[15] Te^{129m} (spin 11/2) and Te^{129g} (spin 3/2) were determined separately; the results are shown in Table II and Fig. 2. In agreement with the predictions of Häller and Rudstam,[16] the low spin isomer is favored for a simple (p,pn) reaction (σ_m/σ_g = 0.88, 0.79, 0.78, 0.83 at E_p = 60, 120, 180 and 233 Mev., respectively) and is essentially energy independent. Häller and Rudstam found

(14) Reference 2, p. 202.

(15) D. Strominger, J. M. Hollander and G. T. Seaborg, *Rev. Mod. Phys.*, **30**, 585 (1958).

(16) I. B. Häller and G. Rudstam, *J. Inorg. Nucl. Chem.*, **19**, 1 (1961).

nucleus. Single particle evaporation would be expected to have a relatively high cross section at low proton energies and fall off at higher energies owing to the larger amounts of excitation energy deposited in the nucleus and the consequent competition with multiparticle emission. This is the picture shown in curves A, B and C of Fig. 1. Neutron evaporation is, of course, much more probable than proton evaporation because of potential barrier effects and the fact that neutrons are in greater abundance than protons.

The result, then, of the gradual increase in direct interaction and the relatively rapid decrease in evaporation with increasing proton energy is a gradually decreasing (p,pn) cross section.

It is not so easy to generalize about the (p,2p) reaction because the Monte Carlo data do not agree with the experimental results. However, one would expect that a very gradual increase would occur in the direct interaction and that proton evaporation would not be nearly as prominent as neutron evaporation.

With the above picture of the changes in mechanism with variation in energy, the deviations of calculated curves from the experimental curves can be discussed. The normally expected curve would be a steadily decreasing function for the (p,pn) reaction and a steadily increasing one for the (p.2p) reaction with increasing energy. This is what was found by Ware and Wiig[4] for these two reactions on Ce^{142} and by Weick[17] for the (p,pn) reaction on I^{127}.

The (p,2p) product, Sb^{129}, in the present study contains a closed shell of protons plus one which would be expected to have a higher level density than would be the case without shell structure. In such a case the evaporation width for protons from excited Te nuclei should be greater than normally would be expected. At low energy where the evaporation process contributes a fairly large percentage of the total cross section this effect might become apparent. This seems to be the case in the (p,2p) excitation function. The evaporation mechanism is so predominant that the cross section falls off with increasing proton energy from 60 to 120 Mev. as it does for the (p,pn) reaction between 60 and 240 Mev. Above 120 Mev., however, the knock-on reaction becomes more important and the cross section rises with energy as is expected.

(17) C. F. Weick, Ph.D. Thesis, University of Rochester, 1959.

If the proton evaporation probability is higher than normal in this case, then the neutron evaporation probability will be less than normal. This effect on the (p,pn) cross section would be small, however, because the neutron evaporation width is considerably greater than the proton width. There is an indication of this in that the Te(p,pn) excitation function is flat between 60 and 120 Mev. while for Ce^{142} there is about a 14% decrease[4] in the same region. In the Ce^{142} case, however, the opposite effect is in operation. The neutron evaporation is going to a product nucleus with a closed shell (82) of neutrons plus one so that its level density would be higher than would be expected without shell structure and therefore neutron evaporation from Ce^{142} is more probable than would be expected without shell structure.

It is of interest to speculate on the effect on the Monte Carlo results of changing the model so as to have a nuclear potential which falls off gradually near the edge of the nucleus rather than a square well and a proton to neutron ratio near the edge which is smaller than in the center. Based on the discussion of the direct interaction given above, these changes would probably enhance the (p,pn) reaction, especially at higher energies, since there would be a greater volume in which this type of reaction could take place as well as a greater mean free path of the nucleons near the edge. For the same reasons the (p,2p) reaction cross section would be decreased. It would probably be decreased more at lower energies where the reaction zone would consist only of the outermost rim of the nucleus, where the proton to neutron ratio would be small. At higher energies the reaction zone would include larger volumes of the nucleus where the proton to neutron ratio is greater. The net effect would probably be a predicted (p,2p) cross section which is closer to the experimental results for tellurium. The enhancement of the direct interaction for the (p,pn) reaction and the corresponding decrease in the evaporation reactions may also give the gradual decrease in the cross section between 60 and 250 Mev., an increase between 250 and 400 Mev., and a gradual fall-off above that energy as a result of competition with other reactions as has been observed for Ce^{142} by others.[3a,4]

Acknowledgments.—The aid and coöperation of Professor S. W. Barnes and the crew of the Rochester 130-inch synchrocyclotron are gratefully acknowledged.