PREPARATION OF PURE CARRIER-FREE
^{123}Xe FOR RARE-GAS WASHOUT STUDIES

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The production of pure ^{123}Xe is considered for use as a tracer in rare-gas washout studies. The major contaminant in ^{123}Xe produced by the ^{122}Te(^{3}He,2n)^{123}Xe nuclear reaction was found to be ^{122}Xe, and the conditions for minimizing the ^{122}Xe impurity are discussed. The magnitude of this contamination is compared with that predicted from a compound statistical model calculation, and good agreement is obtained between the theoretical and experimental ratio of ^{122}Xe/^{123}Xe as a function of bombarding energy. The analysis is then extended to the production of ^{123}Xe by the alpha-particle bombardment of ^{122}Te. It is suggested that this model can be applied to estimate theoretically amounts of impurities when producing radionuclides by cyclotron irradiation in cases where the reaction cross sections are unknown.

Although ^{133}Xe is widely used for washout studies, there has recently been great interest in the production of rare-gas radioisotopes with more favorable decay characteristics than ^{133}Xe. Wagner (1) has listed 13 short-lived rare-gas radioisotopes which he suggests may be of biomedical interest. At this time, 36-day half-lived ^{127}Xe (2), 9-hr half-lived ^{135}Xe (3), 13-sec half-lived ^{81m}Kr (4), and 4-hr half-lived ^{85m}Kr (5) have been prepared in high radiochemical purity and in quantities sufficient for washout and ventilation studies.

These radioisotopes, with the exception of ^{127}Xe, which to date can only be prepared in limited quantities (2), must be used in tne vicinity of the cyclotron (4,5) or reactor (3) where they are produced. Two-hour half-lived ^{123}Xe has several advantages for use in washout and ventilation studies: This isotope decays in two major modes, by positron decay and by emission of a 149-keV gamma-ray, with 40 positrons being emitted for every 100 gamma rays (6). The first mode of decay allows direct comparison of a xenon washout with that of com-

pounds labeled with the short-lived positron-emitting isotopes ^{15}O, ^{13}N, and ^{11}C (7–9), without having to make corrections for different absorptions of gamma rays of various energies in tissue. The gamma ray emitted by the second mode of decay can be distinguished from the 511-keV annihilation radiation, and the use of the two types of radiation may allow corrections to be made for radiation emitted from the tracers in tissue overlying the organ of interest. However, corrections must be made to eliminate the effects of Compton scatter from the positrons which fall within the window set for the 149-keV gamma rays (10).

METHOD

Xenon-123 has been prepared for several years as a precursor to its daughter product ^{123}I. In this method of production, the aim has been to prepare the radiochemically pure ^{123}I daughter product. Our preparation method was developed to prepare pure ^{123}Xe.

Xenon-123 can be prepared by the following nuclear reactions (11): ^{122}Te(α,3n)^{123}Xe, ^{122}Te(^{3}He, 2n)^{123}Xe, and ^{123}Te(^{3}He,3n)^{123}Xe. Although the published yields (11,12) from the second reaction are lower than from the other two, this is the only one of the nuclear reactions feasible using a small biomedical cyclotron and was the reaction used in this study. Enriched ^{122}Te powder (>90% pure) is mounted in an aluminum pouch in a water-cooled target perpendicular to the ^{3}He beam of the Washington University 52-in. cyclotron. Following an irradiation of between 10 and 20 μA-hr integrated beam current, the pouch is placed in the reaction flask of the apparatus shown in Fig. 1. Thirty milliliters of 3M hydrochloric acid is then added to the reaction flask and the flask heated until the aluminum is dissolved. Two to 3 ml of 30% hydrogen peroxide is then added and the flask heated again until the
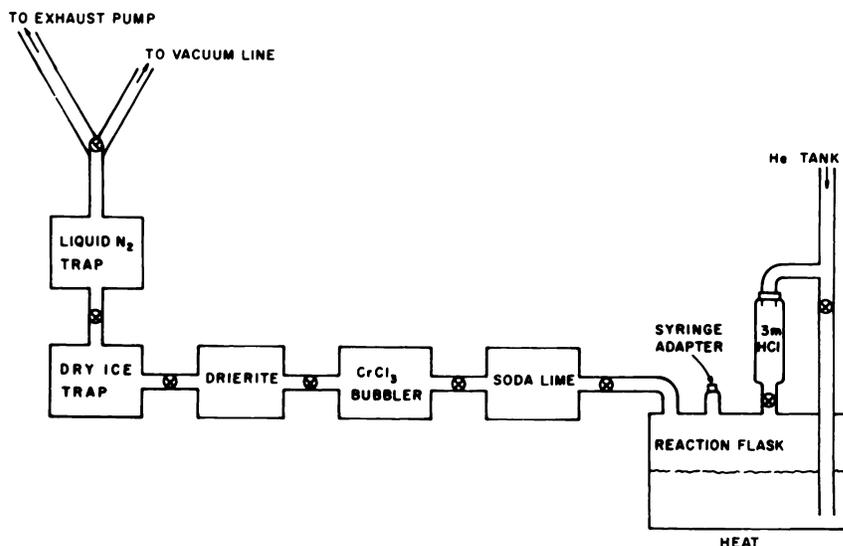
FIG. 1. Schematic of apparatus used for ^{123}Xe separation.

tellurium powder has dissolved. The ^{123}Xe and other xenon radioisotopes are carried out of the solution in the helium carrier gas, with HCl (gas) being removed by the soda lime trap and the oxygen formed by the decomposition of the hydrogen peroxide trapped in the chromous chloride bubbler. The Drierite and dry ice traps remove water and any other low boiling impurities, and the pure xenon is trapped at liquid nitrogen temperature.

Immediately before use, the trap containing the xenon is evacuated while still cooled to liquid nitrogen temperature; the trap is then warmed to $-80°C$ in a dry ice acetone bath, and the xenon is transferred either by Toepler pumping or freezing into an evacuated ampoule which has previously been heated to remove all traces of air (3). Saline is then added to the ampoule, and the xenon solution is injected no later than 10 min after introduction of the saline solution.

Because of the expense of the ^{122}Te target material, the tellurium target is recovered and reused. The tellurium is precipitated by addition of 15 ml of 1 M Na_2SO_3 and 5 ml of 15% hydrazine hydrochloride solution. The precipitate is then filtered and dried with the recovery of the tellurium being >97%.

RESULTS

Analysis of the ^{123}Xe prepared at 24 MeV, at which energy the work of Lebowitz, et al (12) suggests the yield would be greatest, showed that two radioisotopes were present. The impurity was a long-lived positron-emitting isotope, the half-life of which was found by allowing a sample to decay for 3 days and monitoring the decay curve. By analysis of this curve the half-life of the second radioisotope was found to be 20.0 ± 0.15 hr which corresponds to

the half-life of ^{122}Xe (13,14). Xenon-122 decays by electron capture, and the positron emission observed is in fact derived from the decay of the daughter radioisotope ^{122}I:

$$^{122} \text{Xe (20.0 hr)} \xrightarrow{\text{EC}} {}^{122}\text{I (3.5 min)}$$

$$\xrightarrow{\beta^+} {}^{122}\text{Te (stable)}.$$
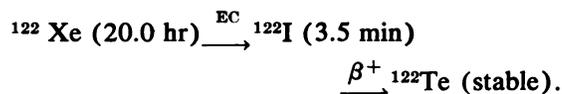
To confirm the identity of the positron emitter, a sample of the xenon was left in a flask for ≈15 min after preparation to allow the ^{122}I to grow. The flask was then cooled at $-80°C$ while the xenon was pumped from the vessel. The vessel was counted
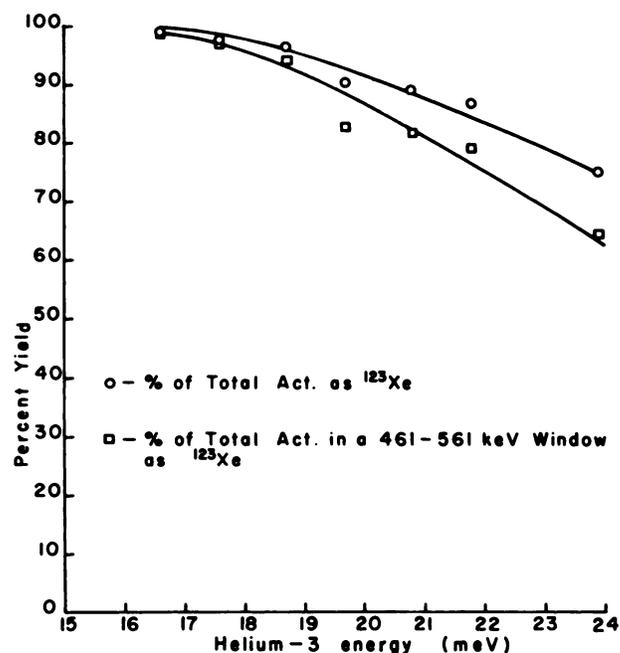

o – % of Total Act. as ^{123}Xe

□ – % of Total Act. in a 461–561 keV Window as ^{123}Xe

FIG. 2. Amount of ^{123}Xe present measured by half-life determination as function of energy.

over a 3-in. scintillation detector attached to a single-channel analyzer which monitored the positron annihilation radiation. The data were collected on line over a 40-min period using a classic LINC computer with the counting rate being integrated every 10 sec. Using a single exponential and a constant, a value for the half-life was determined to be 3.39 ± 0.01 min, which is in good agreement with the previously published values (15,16) of ^{122}I. Use of a mixture of ^{123}Xe and ^{122}Xe will invalidate rare-gas biomedical studies because of the presence of the positron-emitting ^{122}I, so the cyclotron beam energy was altered to maximize the percent of ^{123}Xe. These experiments were carried out by counting part of the xenon in a small flask for ≈36 hr and analyzing the positron decay into two components with half-lives of 2.08 and 20.0 hr. The amount of ^{123}Xe measured at various energies is shown in Fig. 2, where the percentage is plotted in two ways, as percent ^{123}Xe disintegrations present in the positron window of a sodium iodide detector and also as percent of decays which are ^{123}Xe. The spectra of the products at various ^{3}He energies measured with a Ge(Li) detector are shown in Fig. 3. It is seen that bombarding energies of ≦19 MeV ^{3}He must be used to reduce the ^{122}Xe contamination to acceptable levels. The recommendations of Lebowitz, et al (12) do not apply to the production of pure ^{123}Xe, since at the higher energy (24 MeV) high yields of ^{122}Xe are formed. Yields of ^{123}Xe obtained were very similar to those of other workers (11,12), with ≈7 μC/μA/hr/mg of target being obtained. Using a bombardment of ≈15 μA/hr on a 10-mg target, sufficient ^{123}Xe for several washout experiments has been produced.

DISCUSSION

From the data in Fig. 2 one can assess the importance of injecting the ^{123}Xe immediately after the removal of ^{123}I. Figure 4 shows the contribution of ^{122}Xe in the 511-keV window and ^{123}I in the 149-keV window for ^{123}Xe samples prepared at 18 MeV and at 24 MeV. If the ^{123}Xe is injected within 10 min after purification, then the impurity due to ^{123}I present at the time of injection is less than 1%. The growth of ^{123}I during the actual washout study contributes little additional activity. If one injects a sufficient quantity of ^{123}Xe to produce 7,000 cps in the 511-keV window, then the activity due to ^{123}I formed during the washout study is approximately 0.1 cps. The counts due to ^{122}Xe, however, are significant at higher energies.

Although large cyclotrons can produce greater quantities of ^{123}Xe through the ^{122}Te(^{4}He,3n)^{123}Xe
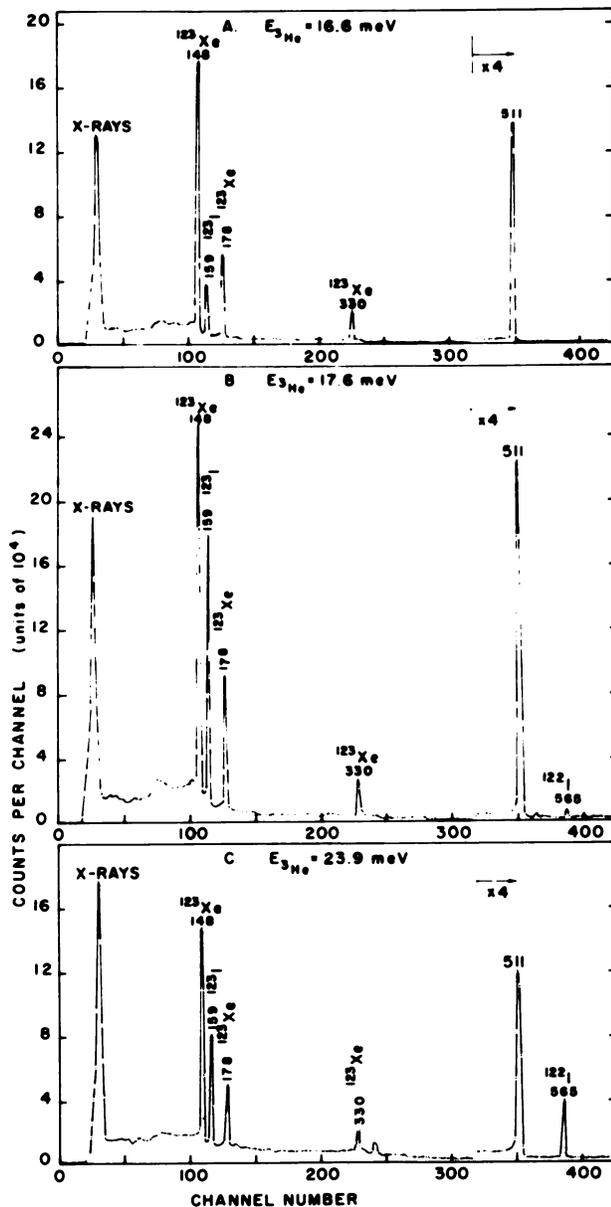

FIG. 3. Gamma-ray spectra taken with Ge(Li) detector of purified xenon reaction products from ^{3}He bombardment of 16.6, 17.6, and 23.9 MeV on ^{122}Te target. Spectra were taken 1.5 hr after end of bombardment.

nuclear reaction, the simultaneous production of ^{122}Xe must be considered.

Even though the excitation functions are not known for the ^{122}Te(^{3}He,xn) and ^{122}Te(^{4}He,xn) reactions, the relative production rates of the xn products can be estimated from the energetics of the system. This is accomplished by the following considerations: (A) The coulomb barrier for the entrance of the ^{3}He or ^{4}He particle, (B) the threshold energies of the reaction, and (C) the energy carried out of the compound nucleus by the neutrons to conserve energy and momentum.

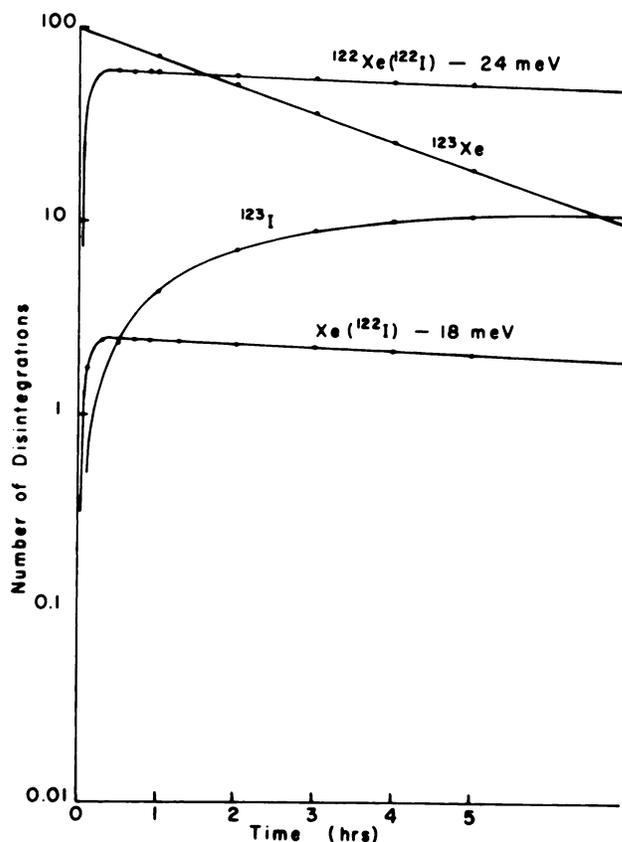The coulomb barrier and the threshold energies

FIG. 4. Impurities present in ^{123}Xe plotted as function of time after separation for irradiations carried out at bombarding energies of 18 MeV and 24 MeV.

TABLE 1. COULOMB BARRIER AND THRESHOLD ENERGIES

Reaction	Coulomb barrier (MeV)	Threshold energy (MeV)
^{122}Te(^{3}He,2n)^{123}Xe	14.65	—6.38
^{122}Te(^{3}He,3n)^{122}Xe		—14.88
^{122}Te(^{4}He,3n)^{123}Xe	14.33	—27.69
^{122}Te(^{4}He,4n)^{122}Xe		—36.25

for the various reactions are listed in Table 1. The coulomb barriers between the ^{3}He and ^{4}He particles and the ^{122}Te nucleus are 14.65 and 14.33 MeV, respectively. Even if the energy of the incident particle does not exceed the coulomb barrier, it can still enter the nucleus because of its wave nature which allows for quantum mechanical tunneling through the barrier. This process, however, lowers the probability of the reaction. On the other hand, if the threshold energy for the reaction is not satisfied, the reaction will not occur.

In considering the third factor, the energy carried out by the neutrons, a compound statistical model calculation can be performed. The ^{122}Te(^{3}He,xn) and ^{122}Te(^{4}He,xn) reactions at bombarding energies

of 5–60 MeV are very well described within the framework of the compound statistical model in which the input energy of the incident particles is randomly distributed throughout all the nucleons, until a nucleon or group of nucleons gains enough energy to escape the compound nucleus through an exit channel. This is referred to as evaporation and is analogous to the escape of a molecule from a hot liquid. Therefore the usual thermodynamic properties of energy and temperature are applicable. The energy and temperature terms that apply in the nucleus are the excitation energy, U, and the nuclear temperature, T. The relationship between these parameters has been shown (17) to be

$$U = (A/12)T^2 - T \tag{1}$$

in which A is the atomic mass number and U and T are in units of MeV. It has also been shown (17) that the average energy carried out by the neutron $(\overline{E}_n)$ is

$$\overline{E}_n \simeq 2T. \tag{2}$$

Solving for T in Eq. 1 and substituting into Eq. 2, one finds

$$\overline{E}_n \simeq \frac{1 + \sqrt{1 + 4(A/12)U}}{A/12}. \tag{3}$$

The excitation energies (U) and average neutron energies for the ^{122}Te(^{3}He,2n), ^{122}Te(^{3}He,3n), and ^{122}Te(^{4}He,3n), ^{122}Te(^{4}He,4n) reactions are listed in Table 2.

The energy available for the neutron leaving the compound nucleus comes from the difference between bombarding energy of the incident particle (^{3}He or ^{4}He) and the threshold energy for the particular reaction (the recoil energy of the xenon nucleus can be neglected). For the reaction to be very probable, this difference must be close to the average energy carried out by the neutron (or neutrons) calculated from Eq. 3. This comparison for the ^{122}Te(^{3}He,2n; 3n)^{123}Xe, ^{122}Xe and ^{122}Te(^{4}He,3n; 4n)^{123}Xe; ^{122}Xe reactions as a function of bombarding energy is shown in Figs. 5 and 6, respectively. It should be noted that Eqs. 2 and 3 are not exact and also refer to the average neutron energy carried out of the compound nucleus. In addition, the compound statistical model calculation refers to the production of atoms irrespective of their half-lives or activities. However, the theory should provide a representative index of the competition between the production of ^{123}Xe and ^{122}Xe in the above reactions.

From Fig. 5 at a ^{3}He bombarding energy of 16.6 MeV on a thin target, the ratio of ^{123}Xe/(^{122}Xe + ^{123}Xe) can be estimated from the difference between

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TABLE 2. AVERAGE NEUTRON ENERGY CARRIER OUT OF COMPOUND NUCLEUS

Particle	Bombarding energy (MeV)	Energy target	U (MeV)	$\bar{E}_n$ (MeV)	$\bar{E}_{2n}$ (MeV)	2n* (MeV)	3n* (MeV)
^{3}He	15	^{122}Te	26.6	5.86	7.0	8.6	0.12
	17		28.6	6.11	7.55	10.6	2.1
	19		30.6	6.37	8.04	12.6	4.12
	21		32.6	6.61	8.48	14.6	6.1
	23		34.6	6.83	8.89	16.6	8.1
	25		36.6	7.06	9.29	18.6	10.1
				$\bar{E}_{3n}$	$\bar{E}_{4n}$	3n*	4n*
^{4}He	28	^{122}Te	29.3	7.55		0.31	1.75
	34		35.3	8.86		6.3	
	38		39.3	9.63	10.54	10.3	1.75
	44		45.3	10.66	12.37	16.3	7.75
	46		47.3	10.97	12.87	18.3	9.75

* Average energy available for neutrons: Difference between bombarding energy and the reaction threshold energy.

lines 1 and 4 ($\sim$4 MeV in excess of the average energy carried out by two neutrons), and the difference between lines 2 and 3 ($\sim$5.8 MeV less than the average energy carried out by the neutrons). Therefore, since the threshold for both reactions (Table 1) has been exceeded, one would expect that ^{123}Xe would be favored, but that ^{122}Xe would also be produced. For a thick target of ^{122}Te the ^{123}Xe/ (^{122}Xe + ^{123}Xe) ratio is determined from the integral of production rates from 16.5 MeV down to the threshold of the ^{122}Te(^{3}He,2n)^{123}Xe reaction. As can be seen in Fig. 5, the radiochemical purity of ^{123}Xe increases as the bombarding energy is de-

graded. At a ^{3}He energy of 14.88 MeV, which is the threshold of the ^{122}Te(^{3}He,3n)^{122}Xe reaction, ^{122}Xe is no longer produced, and the production of ^{123}Xe will continue until the ^{3}He energy is below 6.38 MeV. However, because of the coulomb barrier, the reaction rate will begin to decrease at ^{3}He energies below 14.65 MeV. Therefore the thick target yield of ^{123}Xe relative to ^{122}Xe at a ^{3}He energy of 16.5 MeV should be high, but the total ^{123}Xe produced will be decreased as compared with the higher ^{3}He energy. As the ^{3}He energy is increased the percent of ^{123}Xe will decrease. This was validated by the spectra of the purified xenon reaction
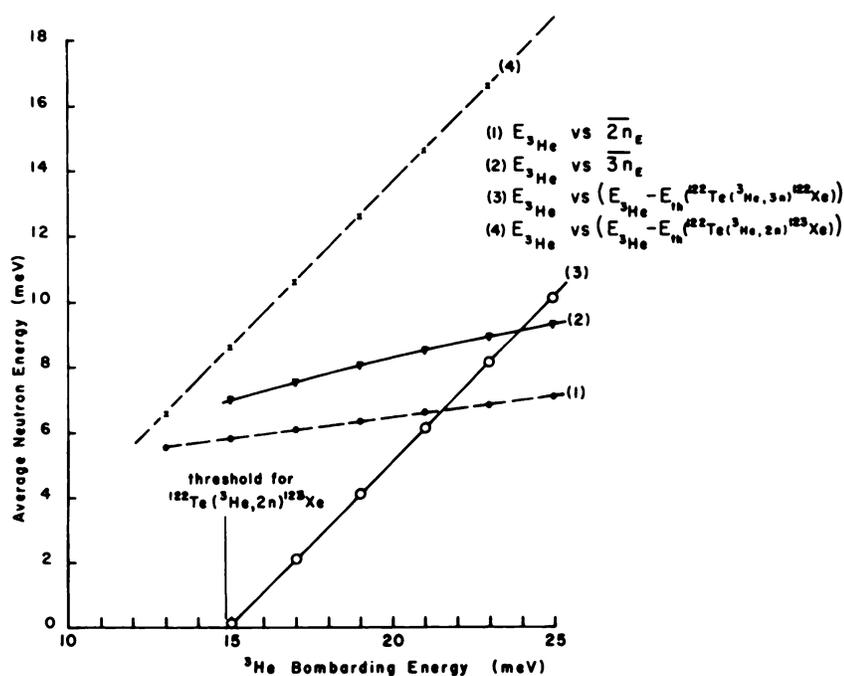

FIG. 5. Plot of average neutron energies carried out of nucleus (calculated from compound statistical model) and actual energy available to neutrons as function of ^{3}He bombarding energy for ^{122}Te(^{3}He,2n; 3n)^{123}Xe; ^{122}Xe reactions.

(1) $E_{^4He}$ vs $\overline{3n}_E$

(2) $E_{^4He}$ vs $\overline{4n}_E$

(3) $E_{^4He}$ vs $\left(E_{^4He} - E_{th}(^{122}Te(^4He,4n)^{122}Xe)\right)$

(4) $E_{^4He}$ vs $\left(E_{^4He} - E_{th}(^{122}Te(^4He,3n)^{122}Xe)\right)$

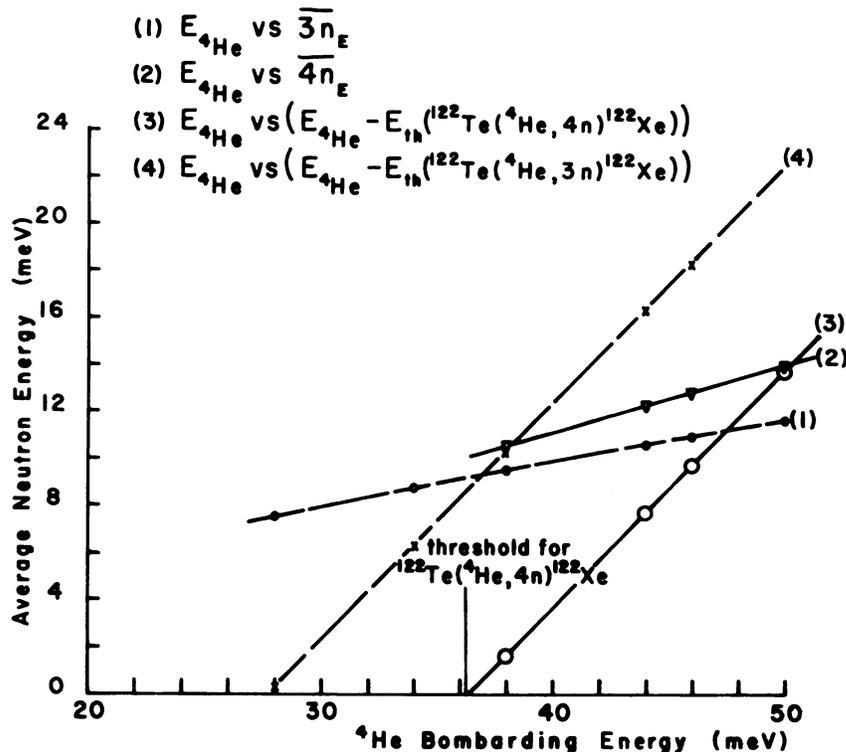threshold for $^{122}Te(^4He,4n)^{122}Xe$

FIG. 6. Plot of average neutron energies carried out of nucleus (calculated from compound statistical model) and actual energy available to neutrons as function of ^{4}He bombarding energy for $^{122}Te(^4He,3n; 4n)^{123}Xe; ^{122}Xe$ reactions.

products at ^{3}He bombarding energies of 16.6, 17.6, and 24 MeV obtained with a Ge(Li) detector, and, as can be seen in Fig. 3, the interference from ^{122}Xe increases with bombarding energy. The $^{123}Xe/(^{122}Xe + ^{123}Xe)$ percentage was calculated at these bombarding energies and found to be >99%, 97.8%, and 75.2%, respectively, which is in very good agreement with the half-life data shown in Fig. 2.

The discussion given here can be applied to any charged-particle compound nuclear reaction and used to estimate the impurities expected. If it is applied to the $^{122}Te(^4He,3n)^{123}Xe$ and $^{122}Te(^4He,4n)^{122}Xe$ reactions (Fig. 6) at an incident energy of ^{4}He of 45 MeV one would expect that a sizable amount of ^{122}Xe will be produced on the target surface, and as the energy of the ^{4}H particle is degraded in a thick target, the radiochemical purity of ^{123}Xe will steadily increase. At an energy of 36.75 MeV the production of ^{122}Xe will cease with ^{123}Xe being produced until the ^{4}He energy drops below 26.7 MeV. Furthermore, if a higher energy ^{4}He beam is used, this will further decrease the $^{123}Xe/(^{122}Xe + ^{123}Xe)$ ratio. An additional factor in the percent ^{123}Xe produced is the collection system since the ratio varies as a function of depth in the target; i.e. if sweep gas flavors the surface reactions, then the radiochemical purity of ^{123}Xe would be worse for flow systems than for target dissolution.

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