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Three New Delayed Alpha Emitters of Low Mass

Luis W. Alvarez

May 31, 1950

Berkeley, California

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Three New Delayed Alpha Emitters of Low Mass

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May 31, 1950

ABSTRACT

Two new positron active isotopes, B^8 and Na^{20}, have been found to decay to excited states of Be^8 and Ne^{20}, which in turn decay "instantaneously" by alpha emission. Their half-lives are 0.65 ± 0.1 sec. and 1/4 sec. respectively. N^{12} is also found to have a low energy positron group which leads to an α-unstable excited state in C^{12}. The masses of B^8 and Na^{20} are 8.027 and 20.015 respectively. B^8 decays by a 13.7 ± 0.3 Mev positron, through the same excited state of Be^8 as does Li^8. Estimates of the energies of the excited state in C^{12} and Ne^{20} are made.

Three New Delayed Alpha Emitters of Low Mass

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Introduction

Until the present time, the only known light, delayed alpha emitter, was Li^8.[1] In the terminology of classical radioactivity, "delayed alpha-particles," such as those from Li^8, are called "long range alpha-particles." They arise from excited states of a daughter nucleus, following a beta-decay, and their real lifetimes are too short to be measured directly. Their apparent lifetimes are those of their parents, with which they are in equilibrium. The expression "delayed neutron emitter," is used for the same reason, to indicate that the observed neutron activity of nuclei such as N^{17},[2] is not a true neutron radioactivity, but rather the "instantaneous" disintegration of an excited beta-decay daughter nucleus. In both neutron and alpha-decays of the delayed variety, it is possible to determine the lifetime of the actual heavy particle reaction, not by time measurements, but indirectly, from the uncertainty principle, using a measurement of the energy spread of the emitted particles.

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(1) Crane, Delsasso, Fowler and Lauritsen, Phys. Rev. 47, 971 (1935)

(2) L. W. Alvarez, Phys. Rev. 75, 1127 (1949)

Li^8 has been investigated by a number of nuclear physicists,[3][4][5][6] and its decay scheme is well understood. The beta transition is first forbidden, and leaves the Be^8 daughter in a broad excited state about 3.1 Mev above the ground state. The width of the state is 0.8 Mev.

Description of Experiments

The present experiments were started in an attempt to observe an example of delayed proton emission. Although this process has not yet been reported, it would be expected from nuclei such as Ne^{17}, O^{13} and C^9. In the case of Ne^{17}, the reactions would be

$$_{10}Ne^{17} \longrightarrow {}_9F^{17*} + e^+$$

$$_9F^{17*} \longrightarrow {}_8O^{16} + {}_1H^1$$

This pair of reactions is similar to the pair describing the delayed neutron activity of N^{17}:

$$_7N^{17} \longrightarrow {}_8O^{17*} + e^-$$

$$_8O^{17*} \longrightarrow {}_8O^{16} + n$$

The 32 Mev proton beam from the Berkeley linear accelerator was used to bombard a proportional counter filled with $B^{10}F_3$. (Protons plus B^{10} could give C^9, and protons plus F^{19} could give Ne^{17}.) The linear accelerator is pulsed 15 times per second, for 300 μ sec., and the proportional counter "cleans up" in a few milliseconds from the huge burst of ions formed during

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(3) D. S. Bayley and H. R. Crane, Phys. Rev. 52, 604 (1937)

(4) Bonner, Evans, Malide and Risser, Phys. Rev. 73, 885 (1948)

(5) F. L. Hereford, Phys. Rev. 73, 574 (1948)

(6) W. F. Hornyak and T. Lauritsen, Phys. Rev. 77, 160 (1950)

the 300 μ sec. pulse. It is therefore very convenient to count delayed heavy particles through a gate circuit which eliminates all pulses during the time the counter is paralyzed. Activities may be followed in this manner, through buildup to equilibrium, and after the accelerator is turned off, through decay. A delayed heavy particle activity was observed in BF_3, with a half-life of about 2/3 sec.

Before giving the reasons for the assignment of this activity to B^8, it will be well to describe other experimental techniques which were used in these investigations. Gaseous targets of CH_4 and Ne were also bombarded, and heavy particle activities were again observed in the manner just described. In the case of BF_3, coincidences between the heavy particles and the positrons were detected. Actually, triple coincidence techniques were used to reduce the background. The positrons were observed with a pair of trays of proportional counters; an absorber was placed between the trays to eliminate coincidences from secondary electrons of gamma-ray origin. The alpha-particles were detected in the bombarded counter. The energy of the B^8 positrons was measured by absorption techniques, using the triple coincidence circuit, and finally comparing the B^8 curve with that of N^{12},[7] in the Feather comparison method. The N^{12} absorption curve was taken in the same geometry, immediately after the B^8 curve.

Excitation curves for all the observed reactions were determined, and the values of the thresholds were measured relative to the $C^{12}(p,n)N^{12}$ threshold, which was investigated very carefully in this laboratory last year.[7] The linear accelerator beam is very mono-energetic at a given time, but the value of the energy may change by a few hundred kilovolts from day

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(7) L. W. Alvarez, Phys. Rev. 75, 1815 (1949)

to day, depending upon the adjustment of the final accelerating gap. It is therefore important to have an easily reproducible energy standard in the region under investigation. The sharp p,n threshold for the production of N^{12} serves this purpose.

Solid targets of Be^9, B^{10} and B^{11} were bombarded on a number of occasions, and both alpha-particles and beta-rays of short half-lives were observed. Alpha-particle range distributions were measured from solid targets, using calibrated mica absorbers and a proportional counter equipped with a thin mica window. Beta radiation from solid targets was detected by a pair of proportional counters in coincidence, and in some experiments, a magnetic field was used to separate the effects of positrons and electrons. This technique was essential in the study of the activities from Be^9, where Li^8 and B^8 are made at the same time. They have similar excitation curves, nearly equal half-lives, almost identical beta-ray upper limits, and identical alpha-particle spectra.

Boron 8

The delayed heavy particles from $B^{10}F_3$ were observed to have a half-life of 0.65 ± 0.1 sec. Since Li^8 has a half-life of 0.88 sec., and since it can be produced by the reaction

$$B^{10}(p,3p)Li^8,$$

it was first necessary to show that the observed activity was not due to Li^8. The energetic threshold for the (p,3p) reaction is 25.8 Mev, but the observed threshold for the delayed heavy particles was found to be 21.2 Mev. If one does not believe that the tri-proton is a stable nucleus with a binding energy of at least 4.6 Mev, these energetic data indicate that the activity cannot be due to Li^8, and must be from some unknown isotope. One

would then tentatively assign the activity either to B^8 or C^9. Assuming the reactions to be

$$B^{10}(p,2n)C^9 \quad \text{or} \quad B^{10}(p,p,2n)B^8$$

one calculates the masses to be

$$C^9 = 9.0270 \quad \text{or} \quad B^8 = 8.0189$$

Barkas'[8] estimate of the mass of C^9 is 9.036. Although his mass estimates have proved to be very reliable, there are no known nuclei with three more protons than neutrons, so there is no way to evaluate the accuracy of his estimates so far from the region of stability. It was hard to believe that the mass of B^8 would be 5.7 Mev lighter than that of Li^8, (8.02502) in view of its greater Coulomb energy. For this reason, it was at first felt that the activity was probably C^9. But if one assumes the reaction to be

$$B^{10}(p,H^3)B^8,$$

the mass of B^8 would be 8.0279. This is a more reasonable value from simple energetic arguments, but it is higher than is allowed by the proton stability of the active nucleus. B^8 must be lighter than the sum of Be^7 plus a proton, which is 8.0273 mass units. The discrepancy between this upper limit and the mass calculated from the threshold energy can be attributed to the barrier effect. In other words, the observed threshold for a (p,H^3) reaction must be greater than the energetic threshold. This effect was observed by Panofsky and Phillips in the $C^{12}(p,d)C^{11}$ reaction, where the masses of all the reacting atoms are known. They found that the observed threshold was about 0.5 Mev higher than the calculated energetic threshold. Since it would take only a 0.6 Mev shift in the experimental threshold for the

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(8) W. Barkas, Phys. Rev. 55, 691 (1939)

$B^{10}(p,T)B^8$ reaction, to make B^8 stable against proton loss, it will be assumed from now on, that the mass of B^8 is almost equal to its maximum stable value of 8.0273. In view of the uncertainties of the actual threshold and the strong evidence that the new activity is B^8, no other view could seriously be taken. (Barkas' estimate of the mass of B^8 is 8.027.)

There are several ways of eliminating C^9 from consideration. In the first place, the maximum possible positron energy it could have (if its mass were 9.027) is about 7 Mev; in the second place, it could not be produced from Be^9, while B^8 could be so produced. Both of these lines of attack were followed, and C^9 was shown to be ruled out by both of them.

The positrons in coincidence with the heavy particles were found by the Feather comparison method to have an upper limit of 13.7 ± .3 Mev, thereby ruling out C^9 with certainty. This upper limit energy checks closely with the value calculated from the mass of B^8, if one assumes that the alpha-particles come from the same state of Be^8 as those following the decay of Li^8. The calculated upper limit is 14.0 Mev. The absorption curve of the alpha-particles, as measured from a solid, thick target of B^{10}, was in agreement with that calculated from the known Li^8 spectrum. This confirms the view at the same excited state of Be^8 is responsible for the delayed alpha-particles from both Li^8 and B^8. The measured energy of the positrons is another indication that the observed heavy particles are alpha-particles with the same energy distribution as those following the Li^8 beta transition.

There is experimental evidence to support the theoretical view that the ground states of pairs of "mirror nuclei" have the same spectroscopic character. Since Li^8 and B^8 have the same forbidden value of ft for their beta decays, it would be very surprising if they did not decay to the same state

in Be^8. The two measurements of the B^8 alpha-particle energy (direct, and by subtraction of the β-energy from the mass difference) support this view.

The first attempts to produce the 0.65 sec. activity by proton bombardment of Be^9 were unsuccessful in that the measured half-life of the observed beta-particles was always close to the 0.88 sec. half-life of Li^8. Since it was felt that the cross sections for producing a p,2n reaction should be much greater than that leading to a p,2p reaction near the threshold, this observation was the best evidence that the new activity was C^9. But later work with magnetic analysis of the beta-rays showed that the shorter lived positrons were present, and with the calculated threshold. The smallness of the cross section is still an unexplained fact, although it might be related to the almost negligible binding energy of the last proton in B^8. Figure 1 shows the yield curves of positrons and electrons from Be^9 bombarded with protons. It is apparent from the curves why the B^8 half-life could not be seen without magnetic analysis. The mass of B^8, as calculated from the Be^9 threshold is 8.0264. This is not so reliable a mass value as that from the B^{10} experiment, since no calibration relative to the N^{12} threshold was made, and no background runs were made. The purpose of the experiment was primarily to see if the 0.65 sec. activity could be made from Be^9 by proton bombardment, thereby ruling out C^9. The threshold value, and its related B^8 mass are merely inaccurate by-products of a qualitative experiment to check the isotope assignment.

An additional method of producing B^8 was found. A proportional counter filled with CH_4 was bombarded with protons, and two delayed alpha emitters were observed. One of these was N^{12}, as will be described in the next section. The other had a lifetime of about 1/2 sec., but the activity was too weak to give an accurate decay curve. (It is an interesting experimental

fact that with the techniques used in this laboratory, a half-life in this range of times is about the most difficult to measure accurately. If it is 10 times shorter, the "moveable gate" technique can be employed, with 15 Mev samples being made every second. Thousands of counts may be taken at each setting of the gate, so an accurate decay curve may be made, as was done in the case of N^{12}. If the lifetime is several seconds, a pen recorder can be used to exhibit the output pulses of a scaler, and the scaling ratio may be changed several times during the course of the decay. But with a half-life of 1/2 sec., the time consumed in changing the scaling factor is appreciable compared to the half-life. The counting rate then changes too rapidly from a value that "jams" the pen recorder to one that is too slow to give more than one count per half-life. This is of course not a fundamental difficulty, but one which depends upon the available techniques.) The evidence that the activity from CH_4 was also B^8 was so strong that it could not have been altered by anything less than a very extensive program of half-life measurement. In addition to the approximate half-life (which could not be off by 50 percent) the threshold for the reaction, which was assumed to be $C^{12}(p,n\alpha)B^8$, agreed closely with that calculated from the previously measured value of B^8. The observed threshold was about 30 Mev. A simple calculation using this threshold shows the mass of B^8 to be 8.029. The increase in apparent threshold due to barrier effect will be greater in this reaction than in the p,T reaction on B^{10}, since an alpha-particle has to come out. In addition it has been found that observed thresholds are higher than true thresholds when the activities are weak. Since the threshold is almost at maximum energy of the linear accelerator, it was impossible to extrapolate the yield curve to zero yield. For those two reasons, it is believed that the observed threshold is of the order of 2 Mev greater than the energetic

threshold. In the following table, the value of 8.029 has been reduced by 2 Mev, so the value listed is ~ 8.027. The energetic arguments for believing that the assignment of the new activity to B^8 is correct are summarized below.

Calculated Mass of	B^8	C^9
From B^{10} + p threshold	8.027	9.027
From Be^9 + p threshold	8.026	Impossible
From C^{12} + p threshold	~8.027	~9.014
From Barkas (theory)	8.027	9.036
From proton stability	8.0273	9.035

Additional energetic arguments can be used to show that no other possible nucleus could have the alpha and positron energies measured. This may be appreciated by noting that if other heavy particles are emitted in the primary reactions, energy is required (of the order of magnitude of binding energy), and this would have to be at the expense of the decay energies, since the threshold is fixed experimentally. The isotope assigment is therefore quite definite, although no chemical identification was possible.

Nitrogen 12

When CH_4 was bombarded by protons, two delayed heavy particle emitters were observed. One of these was shown in the last section to be B^8. The second group of heavily ionizing particles had a measured half-life of 0.013 ± .001 sec. The half-life of the positrons from N^{12}, as measured by the same "delayed gate" equipment, was 0.0125 sec. The threshold for the production of heavy particles was the same as that previously found for the N^{12} positrons. The coincidence of half-life and threshold, makes it quite certain that N^{12} may decay in either of the following two ways:

$$1)\quad N^{12} \longrightarrow C^{12} + e^{+}$$

$$2)\quad N^{12} \longrightarrow C^{12*} + e^{+}$$

$$C^{12*} \longrightarrow Be^{8} + \alpha$$

$$Be^{8} \longrightarrow 2\alpha$$

Shortly after the low energy positron branching of N^{12} was observed, a paper on B^{12} by Hornyak and Lauritsen[9] appeared. These investigators show that a Kurie plot of the B^{12} beta-ray spectrum can be resolved into three components, with end points at 13.4, 6.3 and 2 Mev. Within the accuracy of the Kurie plot resolution, the excited state of C^{12} to which the 6.3 Mev beta-ray leads has the same mass as $Be^{8} + He^{4}$. This state therefore cannot be responsible for the heavily ionizing particles observed in the N^{12} branching, as it must decay by emitting either gamma-rays or very low energy alpha-particles. It is tempting to identify the alpha-particles seen in the N^{12} branching as arising from the decay of the higher excited state of C^{12} postulated by Hornyak and Lauritsen to explain the lowest energy component of the B^{12}. This state should lead to three alpha-particles with a total energy of about 4 Mev. This is consistent with the pulse height observed in the CH_4 filled proportional counters. Alpha-particles from the lower excited state of C^{12} could not have been detected above the gamma-ray background. In the discussion so far, it has been tacitly assumed that the observed heavy particles are alpha-particles. It may easily be shown from the measured mass of N^{12}, that there is insufficient energy available for any other known particle, such as a proton or an He^{3} nucleus to be emitted. It is therefore considered established that the delayed heavy particles are alphas.

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(9) W. F. Hornyak and T. Lauritsen, Phys. Rev. 77, 160 (1950)

It would be interesting to make an accurate determination of the shape of the N^{12} beta spectrum to see if three components exist. (So far only the upper limit of the highest energy group is known.) According to present ideas, the same groups should be observed from B^{12} and N^{12}. Both of the groups which could be detected by the methods employed have shown up, so the results so far are in agreement with the theory that the mirror nuclei are identical in so far as their ground states are concerned.

Lauritsen's table of nuclear energy states[10] shows no excited state of C^{12} at the energy required to explain the lowest energy group of B^{12} beta-rays, and presumably the N^{12} delayed alpha-particles. In fact, the lowest tabulated excited state of C^{12} which leads to α-emission is at 16.1 Mev. This is not too unexpected, since the excited state of Be^8 which gives the delayed α's from Li^8 and B^8 is observed in no other reactions. There must be many highly excited states of the simple nuclei which have not yet been observed, since the density of observed states does not increase markedly with energy, as the Bohr theory requires.

Sodium 20

A proportional counter filled with neon was bombarded with protons, and a new delayed heavy particle activity was found. The half-life is approximately 1/4 sec., and the cross section for the production of the heavily ionizing particles is at least a hundred times greater than that for the two previously described activities. The obvious assignment of this activity is to Na^{20}, since it is a member of the series $Z = 2n + 1$, $A = 4n$. In this series, we have: for $n = 1$, Li^4, which is almost certainly unstable; for $n = 2$, B^8, a delayed α-emitter; for $n = 3$, N^{12}, a delayed α-emitter; for

(10) T. Lauritsen, National Research Council Preliminary Report No. 5 (1949)

n = 4, F^{16}, a nucleus which is almost certainly proton unstable (high energy proton bombardment of O^{16} yields no short-lived, high energy positrons). It is therefore reasonable to assume that the 1/4 sec. activity is Na^{20} (n = 5 in the series), produced in a p,n reaction on Ne^{20}. The threshold for the reaction is 16.9 Mev. The mass of Na^{20}, assuming that the assignment is correct, is 20.0152. Barkas predicts a value of 20.0160, and the highest mass which a Na^{20} atom could have and be stable against proton emission is 20.01593. The predictions of Barkas on the masses of the series Z = 2n + 1, A = 4n are all so close to the limit of proton stability that Barkas could not say whether or not they would be stable. It appears experimentally that B^8, N^{12}, and Na^{20} are just stable, while F^{16} is just unstable.

After noting that Na^{20} is a reasonable assignment of the activity, it is necessary to inquire if any other isotope could equally well fill the bill. Na^{19}, produced in a p,2n reaction with the observed threshold, would be lighter than Ne^{19}, and so it and all lighter isotopes of Na are ruled out. Ne^{19} is known to have a half-life of 23 sec. All lighter Ne isotopes produced at the observed threshold would be lighter than the known isotopes into which they would have to decay, and are therefore ruled out. The same is true for all unknown isotopes of elements with Z less than 10. These energetic arguments complete the identification of the 1/4 sec. activity as Na^{20}.

By the energetic arguments used in the case of N^{12}, it is possible to show that the observed heavy particles from Na^{20} are alpha-particles. It is then possible to set limits on the height of the excited state in Ne^{20}, which decays into an α-particle plus an O^{16} nucleus. The combined mass of He^4 and O^{16} is 20.0039, and the mass of Ne^{20} is 19.99877. The lowest excited state of Ne^{20} which could give rise to an α-particle, is then 4.8 Mev

above the ground state. The highest state of Ne^{20} which could be reached from Na^{20} (by a positron of zero energy), is 14.3 Mev above the ground state. The limits can be pushed together from both directions by the following arguments. The heights of the α-particle pulses indicated that the energy release was greater than 2 Mev. This is qualitative, in view of the fact that the windows of the proportional counter were too thick to allow calibrating α-particles to be introduced. We will therefore raise the lower limit on the excited state to 6.8 Mev. The intensity of the α-particles was so great that it is very improbable that the positron branching ratio to the excited state of Ne^{20} is less than 5 percent. This would make the effective half-life for the transition less than 5 sec., or the positron energy greater than 3.5 Mev. These considerations lower the maximum height of the excited state of Ne^{20} to 10.8 Mev. It seems relatively safe, then, to conclude that the excited state of Ne^{20} is between 6.8 and 10.8 Mev above the ground state.

It should be possible to measure the heights of the excited levels in both C^{12} and Ne^{20} in either of two ways. An absorption curve of the positrons in coincidence with the alpha-particles would give the necessary information, and so would a pulse height analysis of the alpha-particles. The energy distribution of the alpha-particles was observed to be broad, by inspection of the cathode-ray tube display, but no accurate measurements were possible in the absence of a good calibration. A search was made for positrons in coincidence with the N^{12} and Na^{20} alpha-particles. A low intensity of relatively soft positrons was found from N^{12}, but in the case of Na^{20}, no convincing proof of the existence of positrons was found. This shows that most of the Na^{20} positrons to Ne^{20*} were not energetic enough to penetrate the brass walls of the counter, and would indicate that the excited level is somewhat higher than the lower limit of 6.8 Mev quoted in the last paragraph.

It is interesting to note that F^{20}, the mirror image of Ne^{20}, does not

combine in a beta transition with the ground state of Ne^{20}. From the magnitude of the branching ratio in Ne^{20}, it is apparent that the transition between the ground states of Na^{20} and Ne^{20} is also not an allowed one.

The latest isotope chart issued by the General Electric company shows a 3 sec. positron activity assigned to Na^{20}. The classification is "C" (element certain; one of several possible mass numbers). Dr. J. R. Stehn, who prepared the chart, has told the author that the 3 sec. assignment is unwarranted, and that the present identification will replace it in his next edition.

Interpretation of "Hammer Tracks"

Cosmic-ray observers have found many examples of "stars" in photographic emulsions, which have "hammer prongs." A heavily ionizing track is observed to leave the exploding nucleus, and at the end of the track appear a pair of heavy prongs of equal range, but opposite direction. It has always been assumed that such hammer tracks are Li^8 nuclei, and that the heads of the hammers are formed by the resulting pairs of alpha-particles. The lengths of the hammer heads are in agreement with this assumption. But now that it has been found that B^8 gives delayed alpha-particles with the same range distribution, this strict interpretation of hammer tracks can no longer be made. But it is probably true that most of the observed hammer tracks are actually due to Li^8 and not to B^8, since their lower charge would allow them to escape more easily.

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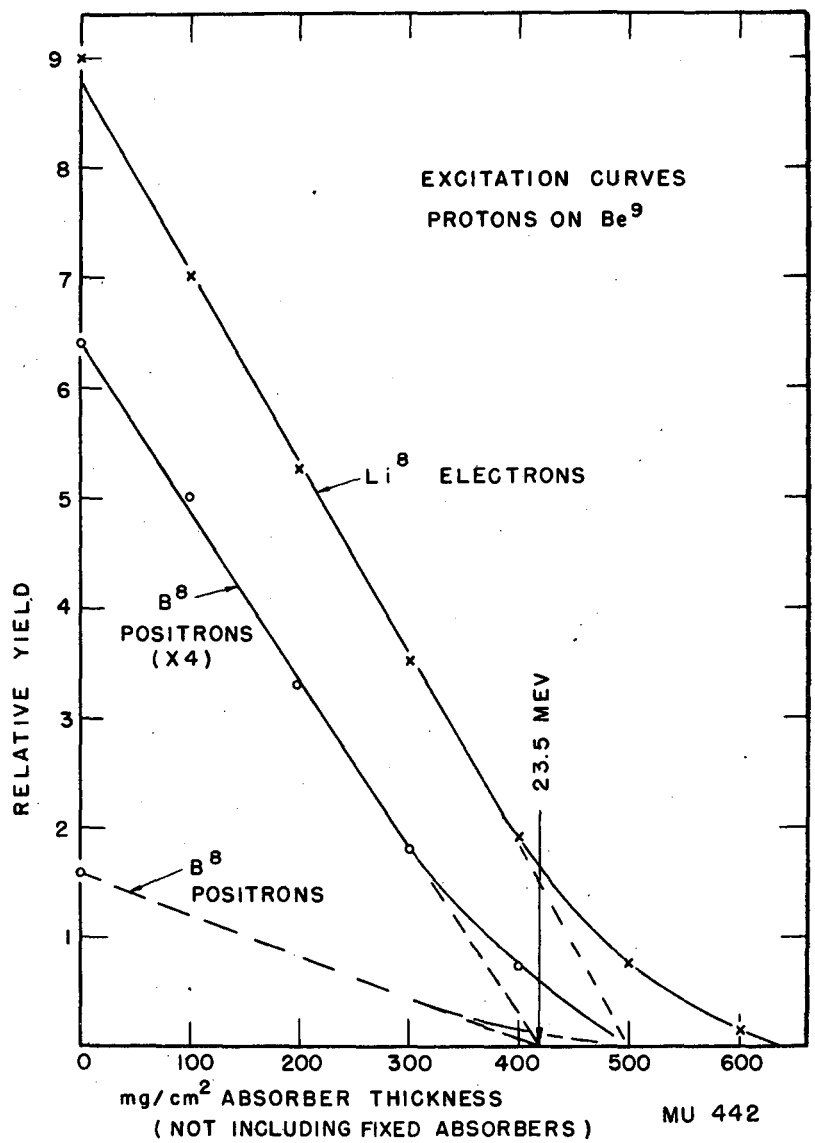
EXCITATION CURVES
PROTONS ON Be9
Li8 ELECTRONS
B8
POSITRONS
(X4)
B8
POSITRONS
23.5 MEV
RELATIVE YIELD
mg/cm2 ABSORBER THICKNESS
(NOT INCLUDING FIXED ABSORBERS)
MU 442
0
100
200
300
400
500
600
1
2
3
4
5
6
7
8
9

FIG. 1

16010-1