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COMPARATIVE YIELDS OF ALKALI ELEMENTS AND THALLIUM FROM URANIUM IRRADIATED WITH GeV PROTONS, ^{3}He AND ^{12}C

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ABSTRACT

Mass-separated ion beams of the alkali elements Na, K, and Fr, and of the element Tl, were produced by bombarding a uranium target with 600 MeV protons, 890 MeV $^{3}He^{2+}$, and 936 MeV $^{12}C^{4+}$. Isotopic production yields are reported. In the case of the ^{12}C beam, these are thick target yields. Absolute cross-sections for the proton beam data were deduced by normalizing the delay-time corrected yield curves to measured cross-sections. For products farthest away from stability, the $^{3}He^{2+}$ beam generally gives the highest yields.

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1. INTRODUCTION

The present work was initiated by the new possibilities of accelerating heavy ions at the CERN Synchro-cyclotron (SC). In addition to the 600 MeV proton and 910 MeV $^{3}He^{2+}$ beams, the SC is also able to accelerate $^{12}C^{4+}$ ions to an energy of 86A MeV. Since neutron emission is usually favoured over the emission of charged particles, the heavy-ion reactions may be suitable for the production of a variety of very neutron-deficient nuclides of Z higher than the target element. The purpose of the present experiment was, however, to investigate the production of nuclei lighter than the target, and to see if the more complex projectiles have higher cross-sections so as to significantly broaden the isotopic yield distributions.

A survey of comparative yield measurements for the elements Na, K, Tl, and Fr, produced by bombardment of 12 g/cm^2 uranium targets, was carried out at ISOLDE [1]. The ^{12}C ions are completely stopped in such thick targets, and therefore these measurements are thick-target yields. Relative to protons and ^{3}He, the effective target thickness is thus reduced considerably. Here we shall discuss some aspects of the utilization of heavy ions for the production of rare nuclear species.

2. EXPERIMENTAL TECHNIQUES

Data for available projectiles at ISOLDE from the CERN 212 cm Synchro-cyclotron are summarized in Table 1. The beam intensities were measured by a secondary emission chamber which was calibrated against the reaction $^{27}Al(X,xnyp)^{24}Na$ [2].

The reaction products separated in the ISOLDE electromagnetic mass separator were brought through a beam-handling system onto a movable aluminized mylar tape [3]. The collected activity was transferred to a thick, 40 mm diameter, 4π plastic scintillator [4] where the β-particles were counted. The detection efficiency, shown in Fig. 1, was measured by means of standard β-sources. The 4π plastic scintillator was also used for the Tl isotopes, which decay by isomeric transition or electron capture. In these cases, the absolute yields are estimated to be uncertain by a factor of 2 to 3 owing to the varying efficiency of the β-detector. The

relative yields for the different projectiles are, however, not affected. For the detection of α-particles, silicon surface barrier detectors [5] were used either placed in the beam behind a carbon collector foil or in combination with a tape-transport system [6]. The neutron-rich nuclides, which are characterized by the emission of β-delayed neutrons, were identified with a 4π neutron counter [7] calibrated with a 48 g sample of uranium. For a few nuclides the gamma-rays were measured with a 17% Ge(Li) standard efficiency detector [7]. The observed counting rates were corrected for decay losses by using the formula presented in ref. [8] in order to obtain the saturation yields. To eliminate the effect or short time variations in the bombarding beam intensity and the separator efficiency, most of the yields were obtained as ratios between two adjacent masses. The saturation yield ratios determined were then normalized to one absolute yield measurement for each element.

3. RESULTS

3.1 Measured yields

The presented production yields from the proton and ^{3}He irradiations are normalized to a beam intensity of 1 pμA and a target thickness of 10 g/cm^2 of uranium, i.e. thin-target yields. Since the ^{12}C beam is completely stopped in the target, its saturation yields are thick-target yields normalized to 1 pμA. The proton beam results are shown in Figs. 2-5. Within the experimental accuracy the isotopic distributions, shown in these figures, reveal no structure due to odd-even effects. The Fr and Tl yields have their maxima at the neutron-deficient side of stability as expected for spallation products [10]. The yields for Na and K are peaked at the neutron-rich side, but closer to the stability line in accordance with the fragmentation model [10]. The yields from the ^{3}He and ^{12}C irradiations are presented in Figs. 7-14 as ratios to the proton-induced yields. The presented distributions will be further discussed in section 4.

3.2 Delay-times and cross-sections

The time distribution for the release of Na from a UC_2-graphite cloth target was determined by measuring the intensity of the ^{25}Na beam, by means of a channeltron

detector, as a function of time after switching off the proton beam. The recorded curve was corrected for decay and general background, and for variable background due to the electrons from the decay of ^{25}Na collected close to the detector. The data are shown in Fig. 6. The parameter μ for the diffusion mechanism(s) in solids, described in ref. [11], was fitted to the experimental points in Fig. 6, giving a value 0.26 s^{-1}. Earlier results from on-line delay measurements, ref. [11], show that the same μ-value is roughly applicable also for K and Fr.

The μ-value determined was used to correct the saturation yields for decay losses in the target in accordance with the formalism in ref. [11]. At temperatures around 2000 °C, mainly nuclides with half-lives shorter than 30 s are affected, because at this temperature a rather fast release of products is normally observed. The 20 ms isotope ^{219}Fr is a good case for testing the relevance of the diffusion correction. The delay-corrected yield of ^{219}Fr, shown as an open circle in Fig. 5, falls within a factor of 2 of the ^{220}Fr yield. This nuclide has about the same formation cross-section but, owing to its long half-life, no delay correction is necessary. To increase the corrected yield of ^{219}Fr to a proper value, the diffusion time in the target-ion source system must be much longer than that found in the present work. For the most short-lived Na isotopes, the saturation yields were increased by a factor of 10-15 because of delay losses, and for the K isotopes the corresponding increase is between 3 and 7. All the Fr isotopes are almost unaffected by decay losses, except for ^{219}Fr for which the saturation yield was increased by a factor of 20. The diffusion time for Tl was not measured for the actual target system, but earlier experiments have shown that it is much longer than for Na, K, and Fr. In this case it was not possible to apply the μ-value obtained for the alkalis, and therefore the Tl yields were not corrected for delay in the target system.

The yields depend very strongly on the performance of the actual target-ion source system and, in order to keep the experimental conditions approximately the same, the presented data for p and ^{3}He were obtained by using the same target unit. Due to temperature differences, the performance of various targets mainly affects

the short-lived nuclides because of their strong sensitivity to decay losses in the target. Occasionally 10-100 times higher yields have been observed for these nuclides. This means that precise cross-section measurements are difficult to perform at an on-line separator such as ISOLDE, but it is very suitable for relative yield measurements. It may still be interesting to estimate approximate cross-sections far away from stability, since it is very difficult to obtain this information from techniques other than on-line measurements. In order to obtain absolute formation cross-sections, the delay-corrected yield have to be related to cross-sections measured, for instance, by radiochemical methods [12,13,14]. The cross-sections given in Figs. 2, 3, and 5 are determined to a precision of a factor of 2 to 3, depending on the uncertainties in the delay-time corrections and the normalization cross-sections.

4. DISCUSSION

For the fragmentation product Na, the yield ratios in Figs. 7 and 8 show that both ^{3}He and ^{12}C give a higher yield at the neutron-deficient side as well as at the neutron-rich side of the distributions. The higher yields are tentatively understood in terms of higher energy deposition in the target nucleus. For production purposes the ^{3}He beam will become even more attractive in the near future because a beam intensity of the same order as the proton beam is within reach at the SC. An unexpected high yield was observed for ^{21}Na with ^{12}C as the projectile. To investigate whether this could be attributed to the reaction ^{12}C on carbon in the target, a separate experiment with ^{12}C on a pure graphite target was performed. The result obtained was in agreement with integration of the 30 to 80 MeV ^{12}C on ^{12}C data [15,16], showing that the ^{21}Na is produced near the end of the range of the beam in the target. This experiment points to the possibility of performing such reactions in thick targets, which may be interesting in order to produce neutron-deficient nuclei in the region $Z < 20$ where suitable high-temperature targets for proton and ^{3}He bombardment are hard to find. In the ^{12}C on ^{12}C experiment a small contribution at masses $A > 24$ was also observed, originating from reactions with the Ta target container.

The ratios for the fragmentation product potassium, K, are shown in Figs. 9 and 10. The trend of higher yields from the ^{3}He and ^{12}C irradiations is not as pronounced as for Na, but still there is a gain in yield at both the neutron-deficient side and the neutron-rich side of the distributions. The argument used above for the Na ratios is also applicable in this case to explain the higher yields produced in the ^{3}He and ^{12}C irradiations.

For the deep spallation product Tl, higher yields are expected when using ^{3}He or ^{12}C as projectiles instead of protons, because the higher total energy transferred to the system favours the evaporation of many particles. The effect is shown in Figs. 11 and 12. The experimental ratio illustrated in Fig. 11 shows that the yields for $A < 187$ are 10 to 100 times higher from ^{3}He than from protons. When using ^{12}C instead of protons as projectiles the effect is not so pronounced, but still there is a gain in yield, as shown in Fig. 12. The measurements of the Tl isotopes were not extended to the neutron-rich side of stability because of contamination from the isobaric Fr isotopes.

In the case of the close spallation product Fr (Figs. 13 and 14), the higher bombarding energy of the ^{3}He and ^{12}C beams does not favour the yields. This is analogous to the case where the proton energy is increased from medium to high energy, where the cross-section decreases for close spallation products but increases for deep spallation and fragmentation products [10]. In the fission region the cross-section is hardly affected by the higher bombarding energy, in agreement with observations for Cs and Rb isotopes produced in 910 MeV ^{3}He irradiations [8]. For the most neutron-deficient nuclides, shown in Fig. 13, there is a gain by using ^{3}He as projectiles, while nuclides closer to stability are not favoured. Because of contamination from neighbouring masses in the separator, this gives cleaner conditions when studying nuclear properties in the very light Fr isotopes. The low ratio shown in Fig. 14 is not only a consequence of the reaction mechanism but also an effect of the small effective target thickness for ^{12}C, while the proton and ^{3}He yields are thin-target yields.

During the ^{12}C experiment an attempt was made to produce elements heavier than the target. The aim was to produce Am isotopes by irradiating UC_2-graphite cloth by ^{12}C, which involves a transfer of three protons to the target nucleus. In this experiment a thermal ion source was used. The detection system was optimized to measure both alpha and fission-fragment energies. The most typical characteristic of the Am isotopes would be spontaneous fission, but no such events were observed. It is possible that relatively high-temperature stable Am compounds were formed, thus preventing the release of Am from the target. However, some weak alpha peaks were observed in the expected region of energy for Am. Assigning these alpha peaks to Am, an estimated upper limit of 30 μb for the production of ^{237}Am could be made, which roughly agrees with the data obtained by a low-energy ^{12}C beam [17].

The present work shows that the results obtained with the ^{3}He beam are very encouraging and higher production yields are established, especially for the deep spallation and fragmentation products. For production purposes the ^{3}He beam will become even more attractive with the planned higher beam intensity. The ^{12}C beam does not seem to offer any advantage for production of elements lighter than the target. The higher energy available does not compensate for its low intensity and shorter range as compared to ^{3}He and protons.

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Table 1

Summary of beam data at the ISOLDE target

Projectile	Beam intensity (pμA)*)	Incident beam energy (MeV)	Energy loss in target (MeV)
p	3	600	30
$^{3}He^{2+}$	0.5	890	200
$^{12}C^{4+}$	0.1	936	936

*) 1 pμA = 1 particle microampere = 6.24×10^{12} particles/s.

Figure captions

Fig. 1 : The efficiency of the 4π plastic detector obtained by using standard beta-sources.

Fig. 2 : Production yields of Na isotopes. Filled circles are normalized saturation yields (see text). The P_n values used are normalized to the new P_n value for ^{9}Li of (50 ± 4)% [7]. The cross-section scale on the right-hand axis is normalized to 0.19 mb measured for ^{24}Na [12]. This scale applies to the decay-corrected yields (open circles) according to the text. The points at masses 33 and 34 are within parentheses, because the correction for the daughter activities is not taken into account.

Fig. 3 : Production yields of K isotopes. See caption of Fig. 2. The P_n values used are taken from ref. [9]. The cross-section scale is normalized to 0.35 mb for ^{44}K, assuming the same cross-section at the maximum of the yield distribution as that measured for Sc [13].

Fig. 4 : Production yields of Tl isotopes. See caption of Fig. 2. No decay correction is applied to the points (see text). The letters m and g indicate the metastable and the ground state, respectively. The same detection efficiency as for beta-particles was used.

Fig. 5 : Production yields of Fr isotopes. See caption of Fig. 2. The cross-section scale is normalized to 0.20 mb measured for ^{212}Fr [14]. The error bars are from the uncertainties in the μ-value and the normal temperature variations of the target.

Fig. 6 : Fractional yield F(t), defined as the activity per sample after beam-off relative to the equilibrium activity measured while beam-on, for Na from UC_2-graphite cloth at about 2000 °C. The curve is a fit to the experimental points (see text).

Fig. 7 : Ratios of the ^{3}He to the proton-induced saturation yields of Na isotopes normalized to the same beam intensity.

Fig. 8 : Ratios of the ^{12}C to the proton-induced saturation yields of Na isotopes normalized to the same beam intensity.

Fig. 9 : Ratios of the K isotopes. See caption of Fig. 7.

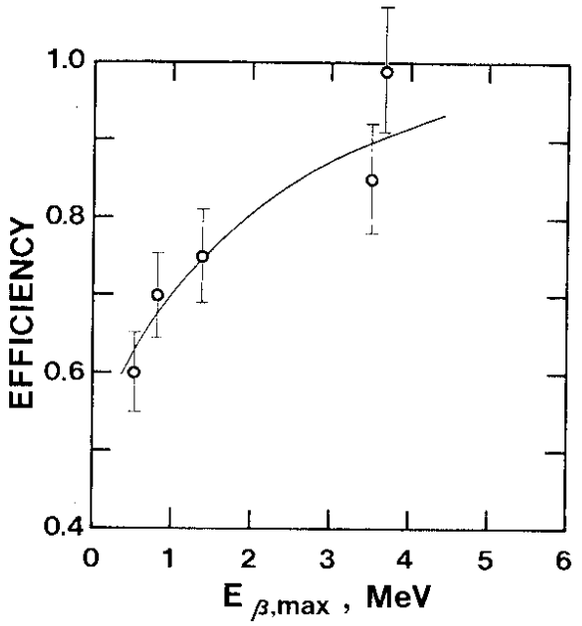
Fig. 10 : Ratios of the K isotopes. See caption of Fig. 8.

Fig. 11 : Ratios of the Tl isotopes. See caption of Fig. 7. For the letters m and g see caption of Fig. 4.

Fig. 12 : Ratios of the Tl isotopes. See caption of Fig. 8. For the letters m and g see caption of Fig. 4.

Fig. 13 : Ratios of the Fr isotopes. See caption of Fig. 7.

Fig. 14 : Ratios of the Fr isotopes. See caption of Fig. 8.



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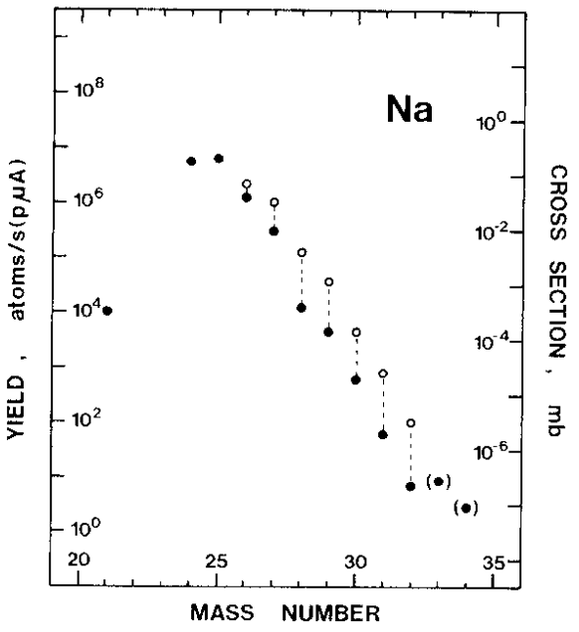

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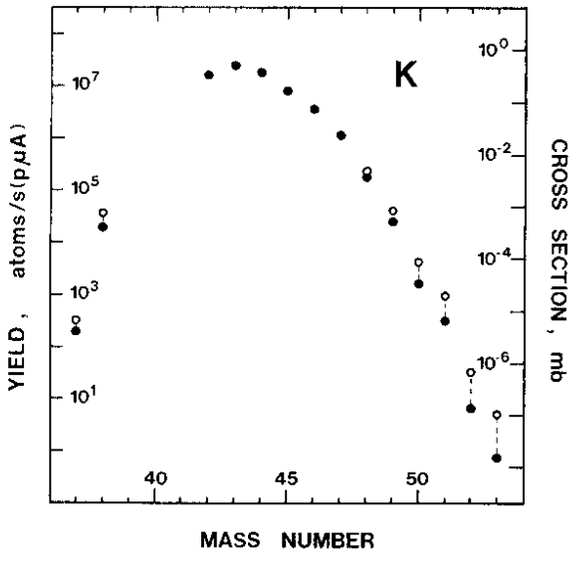

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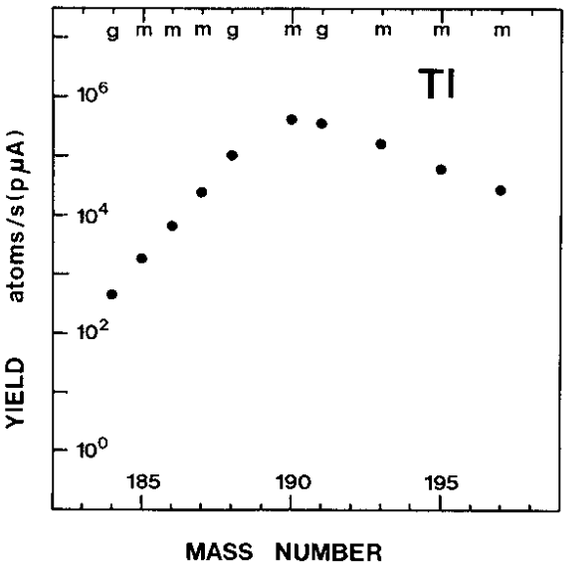

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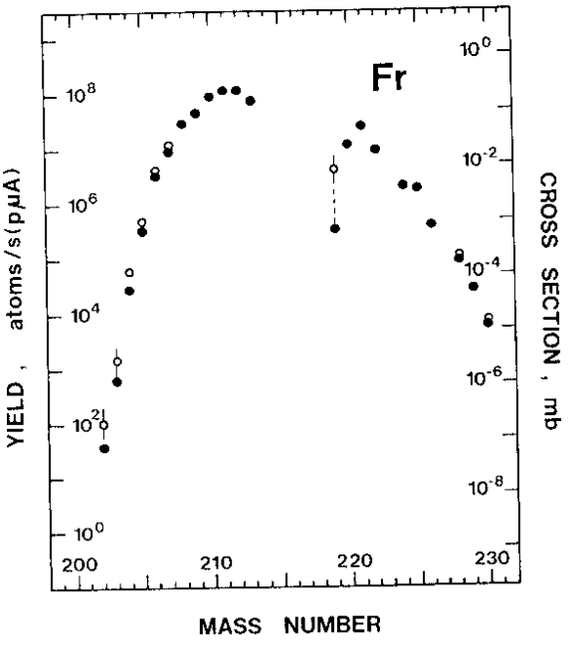

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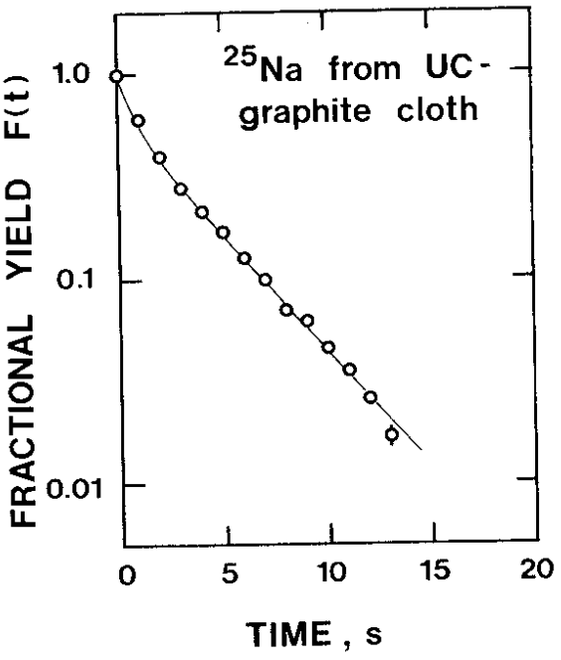

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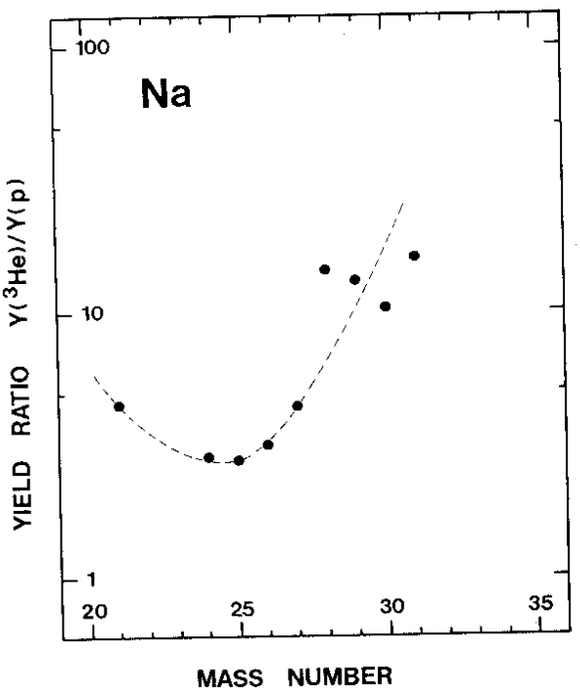
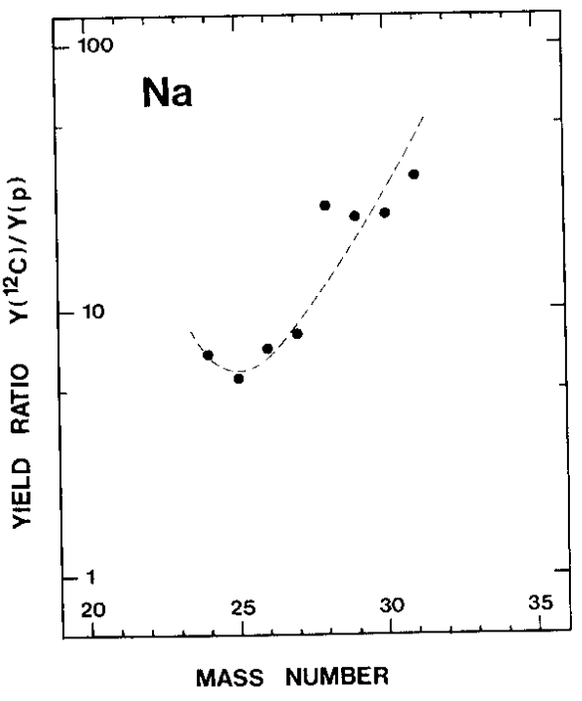


Fig. 7



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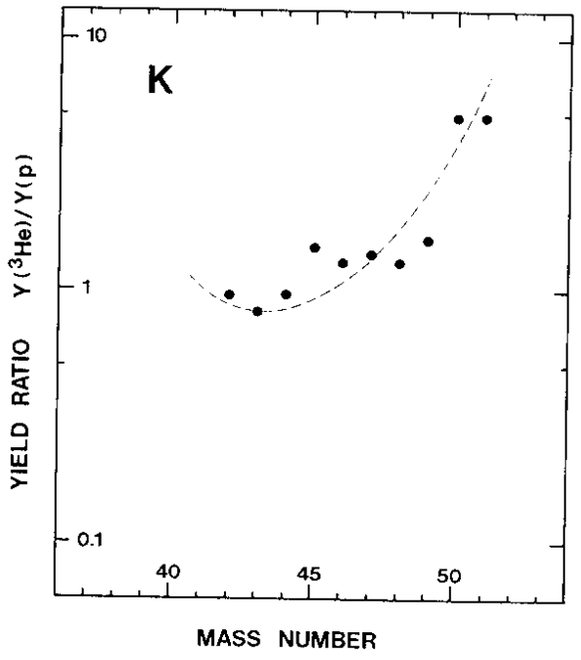

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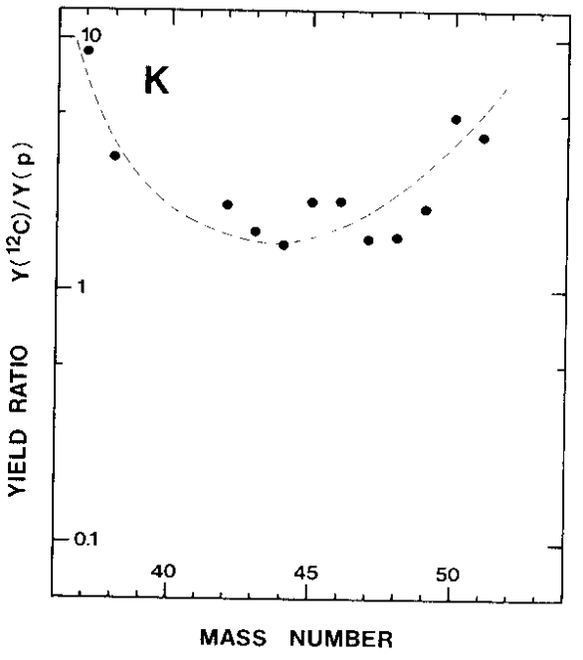

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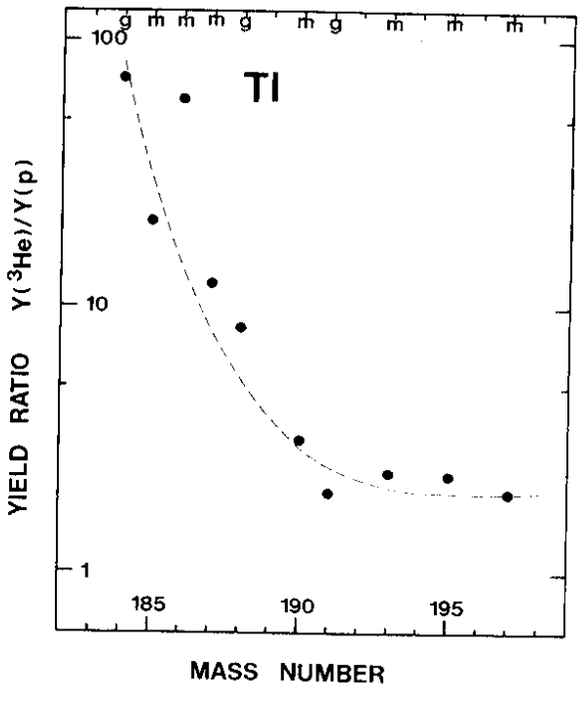
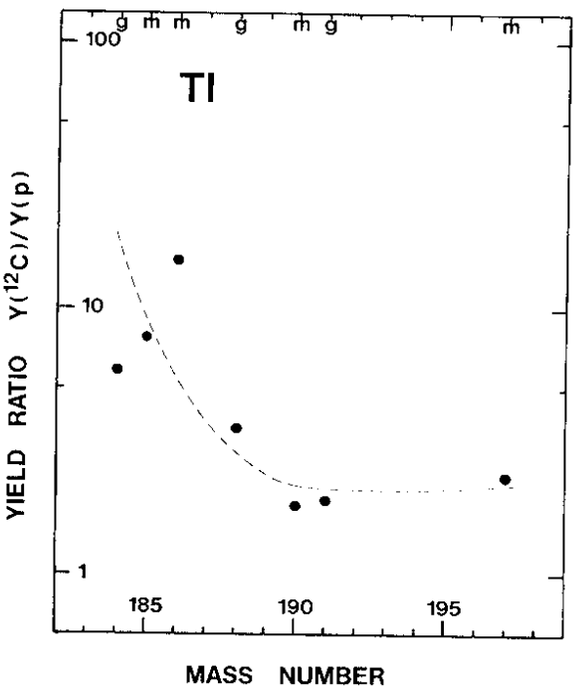


Fig. 11



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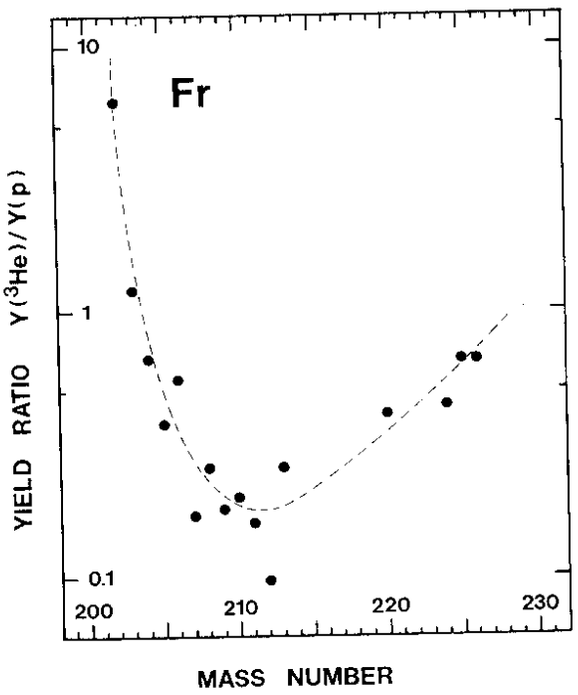
Fr
10
1
0.1
200
210
220
230
YIELD RATIO Y(3He)/Y(p)
MASS NUMBER

Fig. 13

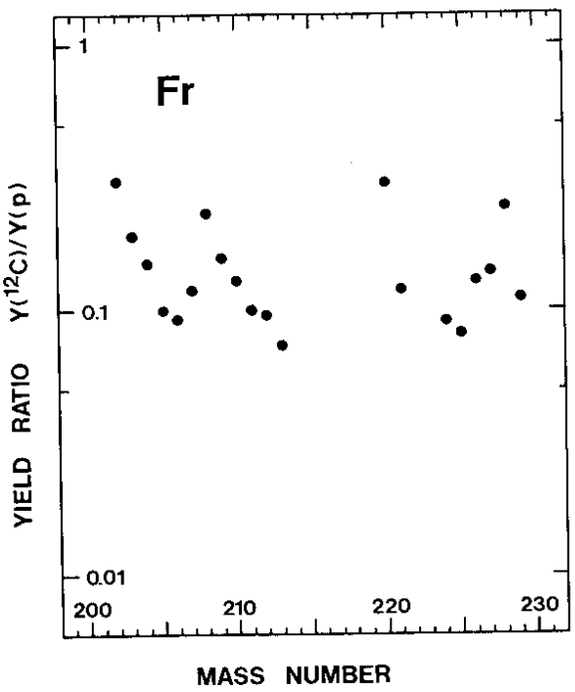
Fr
1
0.1
0.01
200
210
220
230
YIELD RATIO Y(12C)/Y(p)
MASS NUMBER

Fig. 14