PRODUCTION CROSS-SECTIONS OF TRITIUM
AND RARE GASES IN VARIOUS TARGET ELEMENTS

by

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PRODUCTION CROSS-SECTIONS OF TRITIUM
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SUMMARY

Tritium and rare gas production cross-sections for high-energy protons were measured in different materials, including complex materials such as olivine and stone meteorites, in order to estimate production rates and ratios of these isotopes in meteorites. To find the general behaviour of the excitation functions, measurements were made between energies of 0.6 and 25 GeV. The influence on the cross-sections due to irradiation technique, monitoring cross-sections, and gas separation from the target was studied. Values are given for the cross-sections with absolute errors of 15%, and in some cases below 10%. Production ratios were determined from the cross-section measurements, and the final cross-section results have been compared with expected values from Monte Carlo calculations and from Rudstam's spallation formulae. In general, agreement was satisfactory, although the parameters chosen for the stable isotopes in Rudstam's spallation formulae were slightly different from the values adopted for the radioactive isotopes. The results obtained also show in general good agreement with the predictions of the evaporation theory.
I. INTRODUCTION

The present work deals with the cross-section determination of gaseous spallation products such as tritium, helium, neon, and argon, produced by high-energy protons in several elements from manganese to lead. These products were chosen because they are extensively studied in meteorites where they are induced by high-energy cosmic radiation. Target elements from manganese to copper have the same or similar atomic numbers as the most abundant elements in meteorites. However, heavier elements were also used as target materials for a systematic study of cross-sections, and to cover a large range of energies for cosmic radiation production the irradiation was carried out at 0.6, 2.2, and 25 GeV.

Rare gas isotopes can be separated relatively easily from meteoric materials, and the yields are sufficient for measurements of stable isotopes\(^1\). Until now, many yield measurements of radioactive spallation products have been made at all available energies, to study spallation systematics under different radiation conditions, and semi-empirical formulae have been applied to predict these cross-sections\(^2\). However, the parameters used in these formulae have only been fitted to radioactive isotopes. Since the stable spallation products often have the highest yields, the validity of the predictions of the spallation formulae were investigated for the stable products. The sum of all spallation cross-sections of one target element must equal the inelastic cross-section\(^3\), so that the error in summing all measured and interpolated cross-sections is mainly determined by the uncertainty in the estimation of the yield of the stable isotopes. These formulae were checked using measured cross-sections of argon and neon isotopes in iron, copper, and nickel. The dependence of the production of tritium, helium-3 and helium-4 on the atomic weight of the target elements was also investigated.
To determine the age of meteorites from cosmic-ray exposures, the $^3\text{He}/^4\text{He}$ production ratio must be known, because helium-3 is partly produced by the radioactive decay of tritium in the meteorite. To obtain direct information on the production of tritium and of rare gas isotopes in meteorites, olivine and a sample of the meteorite "Ramsdorf" were also irradiated. Tritium can be measured accurately in recently fallen meteorites, so if in addition the helium-3 content and the production ratio of these isotopes are known, then the "exposure time" of the meteorites to cosmic radiation can be calculated. From the absolute tritium cross-sections, and from its specific activity, information about the cosmic-ray intensity outside the earth may be obtained.

Many publications deal with the determination of tritium and rare gas cross-sections, but most of the measurements have so far been made with special target materials or with single energy radiation. The results show considerable scatter as the experimental methods vary. So it seemed to be of some importance to make quantitative determinations of spallation products in different target materials over a wide range of energies, using standard methods for the proton flux measurements. The measurements of the stable, rare gas isotopes were carried out at the Max-Planck Institut für Kernphysik, Heidelberg, using sensitive mass spectrometers. As the initial concentration of the rare gases can be kept very low by degassing the target material before the irradiation, the isotopes considered are, in practice, the most important of the few stable spallation products for which cross-sections can be determined accurately with available proton fluxes. The measurements of the radioactive gaseous isotopes were made at CERN using GM counters. Argon-37 was measured in both laboratories to compare and check the two methods for systematic errors.

Techniques for the tritium determination have already been applied for cross-section measurements in iron with proton energies between 50 and 170 MeV. Very similar mass spectrometric measurements for the rare gases were made at 0.16, 1, 3 and 6 GeV. The results for 25 GeV already published are revised here with respect to recently measured monitor cross-sections with better corrections for secondary particles.
II. EXPERIMENTAL PROCEDURE

1. PROTON IRRADIATION

1) Internal synchro-cyclotron irradiation

The target arrangement on the "probe target" is shown in Fig. 1. The circulating protons hit the target first at the edge facing the machine centre. Figure 2 shows the distribution of the sodium-24 activity produced in an internal target. The abscissa gives the radial distance from the centre of the SC. From this distribution, it is clear that the individual target sheets and aluminium foils for monitoring the proton beam must be aligned very carefully to better than 5° mm. Under this condition, the proton flux between the individual sheets is the same to within 5%. In most of the targets, the aluminium monitor foils were placed at different target depths. This enables an estimate to be made of the error due to target misalignment. The general target arrangement is similar to the one used earlier in the irradiation with the synchro-cyclotron at the University of Uppsala.1)

The proton energy for the irradiation was deduced from the radius of the target position, and most of the irradiations were done between 580 and 590 MeV. The proton current in the machine varied considerably during the time of these experiments (from 0.05 - 0.8 A), but the irradiations for the production of the stable rare gas isotopes were made at high internal proton currents. Under high intensive bombardments, the exposed material can heat up. However, recent tests have shown that tritium and argon losses under such a bombardment due to diffusion are relatively low.12) During the bombardment the target temperature was measured by thermo-sensitive colours, but as some of the target materials used have a relatively low heat conductivity, local higher temperatures on the target edges could have existed. The irradiation lasted between 5 and 250 min, and after the target was removed from the machine it was left for one or two hours to allow the short-lived isotopes to decay before the measurement of the monitoring foil was started.
ii) Internal irradiations in the
"Saturne" proton synchrotron at Saclay

An internal irradiation at 2.2 GeV proton energy was carried out with the proton synchrotron at Saclay. Figure 3 shows the target arrangement for this irradiation. The irradiation lasted 30 hours because the machine had to stop for some time due to technical difficulties. The total number of circulating protons was estimated to be about $5 \times 10^{12}$ protons. From the monitor reaction used in this particular irradiation, an estimate of about eight multiple reversals per proton through the target was made. The target used at this proton energy was considerably thicker than those used at 600 MeV or 25 GeV.

iii) Internal irradiations at the CERN Proton Synchrotron

The irradiations carried out at the CERN PS have already been described. However, for the sake of completeness, the results are recalculated in the present work, as new cross-sections for the monitor reactions have to be applied. Figure 4 shows the arrangements for these irradiations, as well as the activity distribution in the monitor foil. Here again, the activity is highest at the cutting edge of the target. The target was flipped into the beam at a proton energy of 25 GeV. The energy value was obtained from the magnetic field-energy relation. About $5 \times 10^{16}$ protons circulated in the machine. With the target thickness used, a single proton traversed the target about 17 times.

iv) External proton irradiations at 600 MeV

The external proton beam of the CERN SC was used to determine tritium production cross-sections under very different conditions in order to eliminate systematic errors. The intensity of this beam can be measured independently from other monitoring reactions by using a secondary emission chamber which is compared with a calibrated ionization chamber. The secondary emission chamber was constructed to
measure particle fluxes of up to $10^{11}$ protons/sec/cm². The comparison with an ionization chamber was made at reduced intensities ($10^8$ protons/sec/cm²). The ionization chamber was calibrated using a proton beam with an intensity of $10^2$ to $10^4$ particles/sec, in which each individual proton was registered by a counter telescope). Figure 5 shows the experimental layout for the external target exposures. The target thickness for the external target was about the same as for the internal one; the proton energy was determined by the proton momentum as 592 MeV. The targets were placed in the focus of the beam in the experimental hall. The beam cross-sectional area was about 10 cm².

2. MONITORING OF THE PROTON BEAM

In the internal irradiation at 600 MeV and at 25 GeV proton energy, and in the external irradiation at 600 MeV, the reaction $^{27}$Al($p$,3pn)$^{24}$Mg was used for measuring the proton flux. The cross-section values for the monitor reactions used at these energies have been carefully measured in the external proton beams of the two machines. For 592 MeV proton energy, a cross-section of $11.0 \pm 0.5$ mb was used, and at 25 GeV the values obtained for 19 GeV proton energy were adapted. The values measured at CERN have been checked for systematic errors. They agree quite well with measurements made in other laboratories for the same or similar proton energies (Fig.6).

In using these cross-sections, one is, however, confronted with the problem that the same end product, sodium-24, is produced by a (n,α) reaction. Therefore, the values for these reactions were compared with the simultaneously measured cross-sections for the reaction $^{27}$Al($p$,5p5n)$^{30}$F which is not sensitive to low-energy secondaries. In external proton beam exposures both sodium-24 and fluorine-18 were counted, whereas in the internal irradiation it was preferable to count only sodium-24 because its absolute activity can be easily measured (see below).
The production ratio $\text{Na}^{24}/\text{F}^{18}$ from aluminium is known for thin targets (for reference see captions to Fig.7). From the ratio $\text{Na}^{24}/\text{F}^{18}$ found for medium and thick targets, the fraction of sodium-24 produced by low-energy secondaries was calculated. Figures 7 and 8 give the $\text{Na}^{24}/\text{F}^{18}$ ratio for thin targets and the $(n,\alpha)$ cross-section for the production of sodium-24. For different target thicknesses and material, the ratio $\text{Na}^{24}/\text{F}^{18}$ was measured in the external proton beam of the SC. The particle flux was determined separately by the method described above, using the secondary emission chamber.

The corrections found for the sodium-24 and fluorine-18 productions due to fast and slow secondaries are given in Table 1. In Fig.9 the excess of proton flux measured by sodium-24 and fluorine-18 is plotted versus effective target thickness. For fluorine-18 excess, this effective thickness is equal to the target thickness in g/cm², but for sodium-24 the target thickness has been multiplied by a factor (see captions to Fig.9) approximately proportional to $A^{1/2}$ of the target material to account for the different number of secondary neutrons per proton and per g/cm².

Similar estimations for secondaries have been made in other publications by comparing the reactions $\text{Au}(\text{spall.})\text{Tb}^{149}$ and $\text{Al}^{27}-\text{Na}^{24}$. In front of a 9 in. steel target, 26% of the sodium-24 was found to be produced by secondaries at 1 GeV proton energy. A 50% correction for the sodium-24 was found in the rear of a 36 g/cm² steel target at 2.2 GeV². At 0.24 g/cm² target thickness, the production of sodium-24 by neutrons was less than 2% at 3 GeV and less than 3% at 30 GeV. No influence of secondary particles was found at a target thickness below 1 g/cm² and only about 3% effect was found at 2.05 GeV with a target thickness of about 2 - 3 g/cm²². Here the average energy of the neutrons was estimated to be about 7 MeV. All these values are in accordance with the data given in Fig.9 and Table 1. Table 2 shows the actual proton flux through the targets calculated for the different exposures after being corrected with the values of Fig.9.
For the Saclay target irradiated at 2.2 GeV proton energy, the reaction Au(spl.\text{spall.})^{149}\text{Tb}\text{ gave an estimate of the proton flux. However, the low proton current and the long exposure did not provide good final results with this method as the corrections were rather big. Therefore the most recent argon-37 and argon-39 cross-sections\textsuperscript{19} were used for iron and copper, and the absolute cross-sections are given relative to these values. Tritium produced in aluminium could be used in addition as a monitoring reaction, but the cross-section for aluminium at this energy would have to be taken from older values in the literature. A value of 50 mb for the tritium production in aluminium\textsuperscript{20} seems to be a lower limit. Compared with Davis et al.\textsuperscript{19}, the cross-section of tritium in aluminium at 2.1 GeV would turn out to be 75 mb. This value is still within the limits of systematic errors of the older determinations.

Tritium production in aluminium was used as a monitoring reaction for three thick targets exposed to a high total flux. The cross-section of this reaction was precisely measured in early experiments of this work.

For the external irradiations at the SC, proton fluxes were measured with the secondary emission chamber which was compared with a calibrated ionization chamber\textsuperscript{13,14}. No secondary particles affected the flux measurement by the secondary emission chamber, as the proton beam was pure and the chamber very thin (0.2 g/cm\textsuperscript{2}).

In all cases where sodium-24 was used for the proton flux measurement, the decay rate of this isotope was determined by a $\beta-\gamma$ coincidence method. The decay of the monitoring foils was normally followed over 3 - 5 half-lives. The counting statistics of the sodium-24 measurement were good, so that the most important error for the monitoring was due to the correction factor for secondaries, discussed above. An over-all error of 10% is
allowed for the proton flux measurement, of which 5% is allocated to
errors in absolute counting and foil thickness measurements, etc.,
and 7 - 8% is due to uncertainty in the corrections for secondaries.
The error for the monitoring with argon and tritium production in
iron and aluminium is somewhat higher, as the absolute cross-section
is known only to within about 10%, and production by secondaries in
the relatively thick target at 2.2 GeV depends strongly on the
position of the monitor foils in the target.  The monitoring with
tritium production in aluminium at 600 GeV (target I-10, I-11, I-12)
is subject to an error less than 10%, as the cross-section is known to
within 5% and counting efficiency corrections were less than about 5%.
Secondaries of energies above 100 MeV have a similar effect on the
monitoring as they do in the production of tritium, helium and argon
isotopes measured in the exposures, therefore no corrections for
them were applied.

3. APPARATUS FOR TRITIUM AND ARGON-37
DETERMINATION, AND MEASURING PROCEDURE

i) Generalities

The apparatus used for the extraction of radioactive gases
is shown schematically in Fig. 10.

The sample is introduced into the evacuated apparatus through
a mercury lift (L) without breaking the vacuum.  The small air bubbles
which normally adhere to the sample may, of course, cause a slight
pressure rise in the apparatus, but this in no way affects the working
conditions.  The sample is then brought into the crucible (C) by
manipulation with a magnet from the outside.  (Non-magnetic materials
are wrapped in a small piece of iron wire; granulated or powdery
materials are put in a short length of thin-walled tubes of magnetic
material whose ends are squeezed and folded tightly).  The alumina
crucible is placed inside a cylindrical molybden vessel which is then
heated by induction from a high-frequency coil placed outside around
the quartz furnace tube (F).

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The crucible arrangement was modified several times in order to get optimal gas extraction and to check for systematic errors in the degassing procedure. Two of the arrangements most often used are shown in Fig. 11.

The gases liberated were removed from the furnace by a mercury diffusion pump (1) and pumped into the separation part of the apparatus. (It may be noted here that in order to avoid contamination, all stopcocks were of the mercury U-tube type. Where normal greased stopcocks were inevitable as at the pump-outs, these were separated from the apparatus itself by a mercury U-tube.) In the separation part, the gases were led through a beryllium oven (Be) at about 1000°C to decompose water vapour, and over a hot rhodium spiral for cracking carbohydrates. The hydrogen was then separated from the other gases through a palladium thimble (Pd) and transferred by the mercury diffusion pump (P3) to the automatic Topler pump (TP1) which pumped it into the Geiger-Müller counter (2), where the activity was continuously measured to check for completeness.

If, even after prolonged extraction, the counting rate of the tritium counter showed no further increase, the residual gases were transferred into the rare-gases handling part via the stopcock (S4) and the transfer pump (P4). The initial pressure in this part could be measured by the McLeod gauge (G2). The gas was brought into contact with copper oxide (CuO) and calcium (Ca) to remove all non-rare gases. When the pressure had reached its lowest value the rare gases were pumped by the automatic Topler pump (TP2) into the counter (3) where the argon activity was measured immediately.

Normally, the calcium and the copper oxide were predegassed at 600°C and 400°C respectively, before the residual gas from the separation part was admitted. After the admission of the rare gas, temperatures were lowered to 300°C and 200°C respectively, and were maintained at these values.
ii) **Special features**

a) **Crucible arrangement**

In order to obtain optimal results, the crucible arrangement was improved and modified several times. The most important stages are shown in Fig. 11. Heat conduction by the support was kept as low as possible. It was found that, especially with samples of easily evaporating materials, it is useful to put a lid over the crucible to avoid evaporation on the cooler parts of the apparatus (mainly the outer quartz tube which is cooled by an air blower) because these thin evaporated metal films may have getter gases. For the same reason, a glow discharge in the furnace had to be avoided, because in such a discharge gas molecules could be bombarded into the evaporated film or even into the quartz wall. A lid over the crucible does not impede evaporation but most of the evaporated material will condense on the lid, which is somewhat cooler than the crucible itself but nevertheless still hot enough to keep the condensed material in the liquid state, and from the lid the material may drop back into the crucible. In this way the material is in a constant distillation circuit, which facilitates the complete degassing.

b) **Gas-flow equipment**

Another feature of the apparatus is the gas-flow equipment. At the top of the furnace section, a needle valve permits a controlled admission of pure hydrogen or argon during the heating procedure. From this needle valve, a small tube goes down as far as is feasible to the crucible to make sure that the gas is not pumped away by the transfer pump (T1) before it comes near the crucible. The surest way would be, of course, to bring the gas directly into the crucible, but on account of the difficulties involved this was not done. (It would be almost impossible to remove the lid from the crucible sufficiently to introduce the sample.)
c) **Gas volumeter**

If two pumps are connected in series, the intermediate pressure depends on the flow rate and on the ratio of the pumping speeds of the two pumps. This fact was used in connecting two pumps \( P_1 \) and \( P_2 \) together. A thermocouple gauge head (TG) allows the intermediate pressure to be read and to be recorded continuously on a chart recorder.

The device was calibrated in a manner normally used in determining pumping speeds: a leak-rate capillary was connected to the needle valve, and the time \( t \) needed for the displacement of the liquid level from \( a \) to \( b \) (\( t \)) was measured and the flow rate calculated as \( A = \frac{qP_{atm}}{t} \text{ [torr} \, \text{sec]} \) where \( q \) = cross-sectional area, and \( P_{atm} = 720 \) torr (actual barometric pressure at 400 m altitude). The result of this calibration is shown in Fig. 13.

Flow rates from \( 10^{-2} \) torr \( \text{sec} \) down to about \( 2 \times 10^{-4} \) torr \( \text{sec} \) may be read directly from the continuous record. For lower flow rates the following method is possible: if the stopcock \( S_1 \) (in Fig. 10) is closed, the gas will cause a pressure rise in the confined fore vacuum volume of pump \( P_1 \). The time needed for a given pressure rise depends on the volume and on the flow rate of the incoming gas. By intermittently closing and opening the stopcock \( S_1 \), a sequence of peaks is traced on the recorder chart, from which the flow rate can be evaluated. There are two possibilities: either the stopcock is closed for a certain preset time, then the peaks are of constant width and their heights vary directly with the flow rate; or, the stopcock stays closed until a certain pressure is reached, then the peaks are of constant height and their width varies inversely with the flow rate. The second way is more sensitive, because even very small leak rates in the apparatus itself could be measured. In our apparatus the volume of the fore vacuum line was about 140 cm\(^3\) and the pressure rise was \( 5 \times 10^{-2} \) torr, and so for a rise-time of one minute the flow was about \( 1.2 \times 10^{-4} \) torr \( \text{sec} \) (see Fig. 12).
iii) Degassing procedure

A certain "routine" procedure was developed for the degassing of the samples. After the sample was dropped into the crucible, which was then covered with the lid (operated magnetically from outside), the high-frequency generator was switched on. The coupling between the generator and the induction coil was then adjusted so that the crucible was heated to about 1200°C. At intervals of about 10 minutes, the generator current was raised in steps such that the maximum current was reached in about 30 to 40 minutes. During routine measurements the actual temperature inside the crucible could not be determined, but the outside temperature of the lid was found to be about 1350°C. With this outside temperature, the internal temperature of the crucible was found, in a separate experiment, to be at least 1600°C. The temperature of the sample was maintained at this value for one hour. Then the generator was switched off and hydrogen was admitted to the furnace up to a pressure of 3 to 5 torr after the transfer pump had stopped. Then the sample was heated again for an hour under hydrogen, the temperature being somewhat lower because of the high heat conductivity of hydrogen. After one hour the transfer pump was switched on and the gas was pumped into the apparatus. Heating of the sample continued for one-half to three-quarters of an hour. If the amount of tritium extracted during the second heating period was small compared with that extracted during the first run, degassing was considered complete. (If, for example, the second run contributed 10% of the amount extracted in the first run, a third one did not contribute more than 10% of the second or 1% of the first run.) If a higher percentage was obtained in the second run, the procedure was repeated until the increase in activity gained by the run could be neglected.

To prevent a measurable fraction of tritium from being retained in the palladium thimble, the thimble was "rinsed" after each run with some hydrogen introduced into the apparatus by means of the gas flow equipment.
iv) Tritium diffusion through palladium

A hydrogen and tritium mixture diffusing through a hot palladium wall may undergo an isotopic separation by enrichment of tritium on one side of the palladium. Extensive tests have therefore been made to study the diffusion of such hydrogen-tritium mixtures. The result will be reported elsewhere, but the main features are briefly summarized in Fig. 14 which shows the hydrogen and tritium diffusion through palladium at different temperatures. At the temperature selected for the hydrogen separation, 600°C, a complete diffusion of tritium occurs. It should be mentioned that the diffusion measurements were done under exactly the same conditions as the target experiments. The palladium tube and the method of temperature determination was the same in both cases, as was the specific activity of the gas mixture. Only when the palladium was poisoned by silicium or carbon hydrites, did the diffusion of both hydrogen and tritium through palladium slow down. These cases, however, were easily excluded, so that under normal conditions no measurable tritium losses occurred.

v) Counting procedure

a) Tritium and argon-37 activity measurements

Tritium and argon-37 were measured in Geiger-Müller counters. The radioactive gases were added to the standard counting gas (50 torr ethylene and 60 torr argon). The plateau of the counters was checked before each measurement and was accepted when the slope was less than 3%/100 V.

Tritium was normally added together with a considerable amount of inactive hydrogen from the sample, and therefore the pressure in the GM counter increased in some cases up to 200 to 300 torr. During the filling procedure, the plateau of the counter was checked and the admission of hydrogen was stopped by cooling down the palladium thimble when the plateau became worse than 5-7%/100 V. In a few cases, the GM counter had then to be refilled with fresh counting gas and the procedure continued. The final counting rates were normally measured
at different times after the filling of the counter had been completed. If possible, measurements were taken at periods up to 12 hours afterwards. In this way it was ensured that the gas was completely mixed, and spurious counts occurring in freshly filled counters had disappeared. After waiting some time, the counting rate is in general some few percent lower than immediately after the filling. A reproducible counting rate was obtained when the hydrogen of a counter filling was diffused through palladium and transferred into a second counter. It was found that, even after several days, the tritium added to the counting mixture did not exchange with hydrogen of the ethylene. The counting efficiency of the GM counters was measured using a liquid tritium standard. In addition, a special apparatus (described below was used to prepare small tritium substandards from a large tritium sample. This sample was compared with the liquid standard and was, as a gaseous sample, easier to use in routine procedures.

The argon counting did not present any particular problem as no inactive argon was released from the sample. Because of this, the amount of carrier could be controlled during the degassing procedure, and so the plateau of the argon counter remained the same before and after the addition of active argon.

The counting rates for tritium as well as for argon-37 were very high for all targets, with the exception of one, so that the statistical error could be completely disregarded compared with other experimental uncertainties.

b) Determination of dead-time losses

With irradiated targets, tritium counting rates were generally rather high; values of about $10^4$ cpm were quite normal, but in some cases counting rates up to $10^5$ cpm occurred. In this range, correction of the counting rate for dead-time losses becomes extremely important as the correction may easily be 50%, or even more.
The determination of dead-time $\tau$ by the method of paired sources involves a small difference of two large numbers, and so the possible error is high. It is therefore dangerous to use dead-times measured this way at counting rates which are sensibly higher than those used for determining $\tau$. So for counting rates of the order of $10^5$ cpm the following method was used.

If the counter gas expands into a large volume, the decay rate per unit volume diminishes proportionally to the pressure in the counter.

With an observed counting rate $n_1$ at the pressure $p_1$, and correspondingly $n_2$ at the pressure $p_2$, then:

$$\frac{n_2}{p_2(1-n_2\tau)} = \frac{n_1}{p_1(1-n_1\tau)}$$

and

$$\tau = \frac{n_2p_1 - n_1p_2}{n_1n_2(p_1 - p_2)}$$

As the activities were counted directly on the extraction apparatus, the expansion was easily effected by lifting the steel ball, which served as a non-return valve on top of the Toepler pump, and controlled by lowering the mercury level in the Toepler volume. The pressure was obtained from the difference of the two mercury levels. As an example, a series of measurements made at different pressures are given:

<table>
<thead>
<tr>
<th>No.</th>
<th>Pressure (torr)</th>
<th>Counting rate (cpm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>186</td>
<td>123816</td>
</tr>
<tr>
<td>2</td>
<td>148</td>
<td>106416</td>
</tr>
<tr>
<td>3</td>
<td>120</td>
<td>91624</td>
</tr>
<tr>
<td>4</td>
<td>93</td>
<td>75886</td>
</tr>
</tbody>
</table>
If $\tau_{ik}$ is the dead-time calculated from the values obtained from the measurements $i$ and $k$, then

\[
\begin{align*}
\tau_{12} &= 2.932 \times 10^{-6} \text{ min} \\
\tau_{23} &= 2.893 \times 10^{-6} \\
\tau_{34} &= 3.117 \times 10^{-6} \\
\tau_{14} &= 2.975 \times 10^{-6} \\
\tau_{24} &= 3.003 \times 10^{-6} \\
\tau_{13} &= 2.917 \times 10^{-6}.
\end{align*}
\]

The mean value is then $(2.973 \pm 0.033) \times 10^{-6}$ min. The advantages of this method is that the highest counting rate observed is used for the calculation, and therefore an extrapolation error is avoided.

c) **Calibration of GM counters**

In order to determine the actual counting efficiency of counters, an apparatus was developed which allows well-defined tritium activities to be brought into a counter. This apparatus, schematically shown in Fig. 13, consists of a reservoir $R$ of about 7 litres in which a gas mixture with a tritium content of about 200 dpm/torr $T$ is stored at a pressure of some 400 torr. This reservoir is separated from the "dosimeter" part (which is formed by a McLeod gauge $G$ with a range up to some 10 torr, and a special Toepler pump $T$ with a working volume of 2.75 ml) by a ball stopcock. The total volume of the "dosimeter" part was calibrated by expanding a quantity of gas trapped in the capillary of the McLeod, and measuring the pressure afterwards $^c$). The volume of the dosimeter part was $228.7 \pm 0.8$ ml. The quantity of gas removed from the dosimeter part by one stroke of the Toepler pump was 1.2% of the total gas content and thus a sufficiently fine dosing of the counter with active gas was possible.

$^c$) Note: For the reading of the pressure with the McLeod, the gas pressure on the mercury of the "open" capillary must be taken into account as it is not negligible. The exact formula for the pressure then becomes

\[
p = \frac{T\Delta h}{V - q}.
\]

where $l$ is the length of the gas-filled part of the capillary, $\Delta h$ is the difference of the mercury levels in both capillaries, $V$ the volume of the McLeod bulb (including the capillary), and $q$ is the cross-section of the capillary.
The counting efficiency was measured in the following way: after the counter, previously filled with counting gas, had been connected to the apparatus by the ground-tapered joint C, the volume between the counter stopcock and the Toeppler pump was evacuated. Then the v-stopcock in the pumping line was closed. By carefully lowering the mercury level in the ball stopcock S, a small amount of active gas was introduced into the dosimeter part and the pressure in it was measured with the McLeod. The desired quantity of active gas was then withdrawn from the dosimeter part by repeated strokes of the Toeppler pump and brought into the counter by raising the mercury up to the counter stopcock. Finally, the remaining pressure in the dosimeter part was measured again, and from the pressure difference the exact amount of gas withdrawn was calculated.

To determine the actual activity of the stored gas, a counter was used, the effective length of which had been determined beforehand24). Repeated checks with this counter proved that the activity of the stored gas was 198 cpmp/torr J. From the ratio of the observed counting rate to the activity introduced into the counter, the efficiency of it was thus determined and a correction factor for each type of counter was given which included end-effects as well as dead volume.

4. MASS SPECTROMETRIC DETERMINATION OF THE RARE GASES

The target materials used for rare-gas measurements were degassed prior to the irradiation in an ultra-high vacuum system in order to keep the atmospheric contamination low. Some of the material was commercially available.

For the extraction of the rare gases, the targets were melted in an ultra-high vacuum system, which was connected with metal valves to a high-sensitivity mass spectrometer. The ultra-high vacuum was achieved by baking both systems to 400°C. In order to keep the helium diffusion through glass very low, the extraction system, including the extraction furnace, is built of Supremax glass.

The targets were melted in a molybdenum crucible by high-frequency heating. For the iron, cobalt, nickel and copper targets, an Al₂O₃ or quartz insert was used. The schematic arrangement is shown in Fig. 15. During the melting, the argon was trapped in liquid air-cooled charcoal to avoid absorption and gettering. The rare gases were removed in calcium and copper-oxide furnaces at 600°C and 400°C, respectively. These furnaces consist of covar tubings, sealed directly to the glass-line.
The mass spectrometer used is a 60° magnetic type and made from molybdenum-glass and metal. For ion detection, a system of the Dumont 6269 multiplier is used. The resistors of 1 MΩ (Pyrofilm) are inside the tube and directly connected to the multiplier system. The multiplier is intensity-dependent and had to be calibrated. For the present measurements, where the amount of gases were small, the correction applied was below 5%.

The spectrometer was operated under static conditions. First the helium and neon were introduced, while the argon was trapped on charcoal in order to avoid an argon\(^{40}\) background on mass 20. Argon was then introduced over a U-tube cooled with liquid air. Absorption in the trap was checked by removing the liquid air.

For calibrations, comparable amounts of rare gases were introduced in the same way through the extraction system and were added to the sample. The amount of each isotope in the sample was determined from the increase in the intensity.

In order to introduce a known amount of a rare gas into the apparatus, a pipette system was used. This has already been described in an earlier work\(^{22}\). It is based on volume dilution only. From a 5-litre reservoir of helium-3, helium-4, atmospheric neon, and argon, about 100 mm\(^3\) are used for the calibration. This corresponds to about 10\(^{-7}\)–10\(^{-6}\) cc at NTP of each gas. A design of the pipette is given in the lower part of Fig. 15, where a glass capillary and mercury cut-offs are used. Metal tubes with ultra-high vacuum valves are now in operation.
III. RESULTS

1. TRITIUM SPALLATION CROSS-SECTIONS AT 600 MeV

Table 3 is arranged to show results of particular exposures. It gives an impression of the reproducibility of the tritium measurements, but it must be remembered that the experiments referred to, took place over a long period of time and that during this time changes based on experience gained in the degassing and counting procedure occurred. It is especially interesting to compare the results of internal and external exposures, and the earlier results obtained at low proton currents (~ 0.1 µA) with more recent ones obtained with improved techniques and higher internal proton currents (~ 1.0 µA). The variations in the tritium cross-sections obtained from iron target measurements and the rather low values obtained from the silicon target measurements indicate that from some extractions the tritium separations were not always complete. For this reason most of the degassing problems were studied on these materials.

The results for the tritium cross-sections in vacuum-melted iron were lower than the cross-sections measured in commercial steel. To avoid the degassing difficulties, pure iron, which was produced by reduction under hydrogen, was also irradiated. Such material was therefore saturated with hydrogen before the exposure, and the tritium was completely separated from the hydrogen-loaded iron after heating for 20 - 30 minutes, instead of the several hours needed for the vacuum-melted material.

The tritium values given in Table 4 are mean values from different experiments. In all cases, with the exception of the 2.2 GeV target, at least two targets were exposed.

The argon-37 was counted over at least a period of three months (three half-lives), and the counting rates given in the table are those at the end of the proton irradiation. Normally the separation of argon-37 from the target material did not cause any
difficulties, and the accuracy of this measurement is high as the counting rates are between $10^3$ and $10^6$ cpm. Some errors are due to the relatively large corrections for the decay of the isotope as some of the measurements were made only eight to ten half-lives after the exposure. The value of the half-life taken for the evaluation of the data was 34.3 days.

The argon-37 was stored from individual targets in Geiger-Müller counters for more than eighteen months and its decay was followed for as long as possible. Only in some cases could values be produced for argon-39, but the errors are much higher. The final results for argon-39 were obtained by counting the individual samples in an anticoincidence shield where the GM counters had a background between 2 and 3.5 cpm. Figure 16 shows typical decay curves for the argon measured with GM counters.

In Table 4, columns 5 and 7, results obtained by mass spectrometric analysis are given for comparison. These values have been taken from Table 5 and show that even with very different methods the results are in agreement within the limit of experimental errors.

The results from Table 4 are given in Figs. 17a and 17b, together with earlier results and values from other references. For carbon, aluminium, iron, nickel, silver, copper and lead the values are plotted over an energy range from threshold to 25 GeV. The results are also presented as a function of atomic weight at 600 MeV (Fig.18).

2. STABLE RARE GAS ISOTOPES

Table 5 contains all measured production cross-sections of helium, neon, and argon isotopes. As in Table 4 the results are mean values. The errors are not stated in the table. They will be discussed in the following section. Values in brackets are more uncertain than others. Some of these results are also presented in Figs. 18 and 19, together with some of the most reliable, newer values from other laboratories.
3. TRITIUM AND RARE GAS PRODUCTION CROSS-SECTIONS FOR COMPLEX TARGET MATERIAL

In order to obtain spallation cross-sections for tritium and rare gases in meteorites, complex material was also irradiated. Olivine and material of the meteorite "Ramsdorf" (chondrite) were exposed to 600 MeV protons. The results for all the isotopes studied with this complex target material are given in Table 6. For argon-37, the results with GM counters and mass spectrometers are given separately.

IV. DISCUSSION

In the previous chapters, the difficulties which may arise in the determination of tritium and rare gases as spallation products in different target materials have been described. It is interesting to note that some of the difficulties which occurred in the present experiment have also been met by other people. A complete extraction of hydrogen from metals has been studied in connection with metallurgy problems\(^2\)) and it is well known that under certain conditions steel retains hydrogen up to very high temperatures. Different phases of metals and alloys of metals have very different solubilities for hydrogen. The initial concentration of hydrogen in the metal, especially in iron, could be shown to be of great importance for the completeness of the tritium extraction. Iron degassed in vacuum before the proton exposure retains tritium much more than ordinary iron. As the hydrogen concentration in the degassed material is very low and the solubility of hydrogen in iron rises with temperature, the radiation-produced hydrogen isotopes are very strongly bound in this material. From the gas-flow rate chart it seems to be evident that degassing is more complete when there is a rapid rise in temperature in the crucible, but, on the other hand, iron with a high initial hydrogen content (iron reduced in hydrogen) gives off tritium very quickly. Recycling of the degassing procedure with this type of iron showed that the first degassing was already complete. The tritium cross-section for this hydrogen-rich iron is therefore taken as the true value. Special tests were made with simultaneous irradiation of vacuum-melted and hydrogen-rich iron (target 20), which showed that a quick temperature rise in the first degassing and recycling with a large quantity of carrier hydrogen finally give the same result for both materials.
1. **Accuracy of Measurements**

Possible errors may occur for the tritium measurements in the separation procedure as the tritium may not completely diffuse through the palladium wall as discussed before. A thorough investigation was made of the diffusion properties of such gas mixtures under the conditions used through palladium, showing that no tritium is lost with this procedure.

Some errors have to be allowed for in the absolute counting of tritium and argon-37; the former have a low $\beta$ energy and the latter decays by electron capture. Both have therefore to be brought into the counter. In some cases where too much hydrogen was used, the good counting properties of the GM counters were spoilt, but for the results given in the tables, errors due to such sources are less than 10%. Also taking into account the errors made in monitoring the proton flux, the final cross-sections presented in this paper have relative errors less than 10%, and absolute errors less than 15%.

2. **Comparison with Other Results**

Tritium cross-section results from other laboratories are shown in Figs. 17a and 17b together with values obtained in this work.

At Brookhaven laboratory, Libby and his group\(^1\)) measured tritium cross-sections at 0.45 and 2.05 GeV in different target materials from carbon to uranium. The values obtained in single measurements scatter widely, though the method used for the tritium separation is similar to the one described here. Absolute errors are estimated to be 30 - 50%.

Another series of experiments were done at Dubna. Mokhodov and collaborators have measured tritium cross-sections for many target elements, most of them above iron. The degassing was different from the method used in this work as they heated the samples in a resistance furnace. These results compare with our values for iron, lead and aluminium. Absolute errors are not stated\(^6\).
Currie\textsuperscript{20} gives values for "Bevatron-produced tritium" for target elements from carbon to lead. The cross-sections obtained do not compare very well with the values reported here. His result shows a decrease of the tritium cross-section from 2 to 6 GeV for iron and lead which was not found between 2.2 and 25 GeV.

At Orsay, Lefort et al.\textsuperscript{24} measured tritium cross-sections at 150 MeV proton energy. The tritium cross-section is approximately proportional to $A^{1/3}$. A similar result was found in this work at 600 MeV, but their absolute values for the production tritium cross-section at 150 MeV are nearly twice as high as those described in this experiment. These values were confirmed by an independent measurement at Brookhaven by Fireman and Zähringer. Higher preliminary values for tritium cross-sections were recently reported by the Orsay group for 600 MeV. The reason for this discrepancy is not yet understood.

The rare gas cross-sections of this work are in good agreement with the results of the 160 and 450 MeV targets by Schaeffer and Zähringer\textsuperscript{25}. These results were the first absolute cross-section measurements of stable spallation products. However, their helium data of the 2 GeV irradiation are too high by a factor of two. The proton flux for the target was quite low and consequently so also were the amounts of spallation products. Therefore an error in the quantitative gas analysis is possible. Since techniques have improved in the meantime, the new results are believed to be more accurate. They give lower $\text{He}^3/\text{H}^3$ ratios and fit better in the general picture of spallation calculations.

Agreement with the recent work of Bieri and Rutsch\textsuperscript{26} also exists in general, if one corrects their data for the decay during storage of the target. Their irradiation took place on 13 March 1959 and the measurements were made in spring 1962, so that tritium and sodium-22 had already decayed. From their helium-3 and neon-22 values 14% and 33% respectively have to be subtracted.
No other stable rare gas cross-sections have been measured. Values for radioactive argon-37 and argon-41 in iron exist and for purposes of comparison the \( \frac{A^7}{A^8} \) ratio from our 600 MeV irradiation on iron was taken and the argon-38 cross-sections were calculated, since the \( \frac{A^7}{A^8} \) ratio is not very energy dependent. All known values are plotted in Fig. 19 and though quite different techniques have been used, the agreement is satisfactory. Figure 20 represents production ratios calculated from the results shown in Figs. 18 and 19.

The main errors in the rare gas analysis are caused by incomplete degassing, contaminations, and errors in the calibration systems. Incomplete degassing usually only affects the argon when discharges occur. These were avoided and the degassing was checked by repeated melting at increasing temperatures. In all the experiments the error is believed to below 5%.

The contamination of the mass spectrometer only affected the 2.2 GeV targets, because the amount of spallation-produced gases was low. Mass 3 is affected by hydrogen compounds, such as HD\(^+\) and H\(_3^+\). Mass 4 has a contribution from diffusion of atmospheric helium through the glass walls. Masses 20 and 22 have a background from argon\(^{18}\) and carbon-dioxide\(^{14}\). The most reliable neon isotope is mass 21. The argon isotopes are all more or less affected by hydrocarbon lines and masses 40 and 36 by atmospheric argon contaminations. The intensity on mass 40 was considered to be air-argon, so masses 36 and 38 were accordingly corrected. The error for all targets of this irradiation is higher and is estimated for all isotopes to be about 15%.

The total error in the gas analysis is estimated to be 10% for the 0.6 and 2.5 GeV irradiations and to be 15 - 20% for the 2.2 GeV bombardment.
Comparison of the production ratios in stainless steel and copper targets by Bieni\textsuperscript{27} and Signer and Mier\textsuperscript{28} is also possible and the results are given in the following table:

<table>
<thead>
<tr>
<th>Target element</th>
<th>He\textsuperscript{3}/Ne\textsuperscript{4}</th>
<th>He\textsuperscript{3}/A\textsuperscript{38}</th>
<th>He\textsuperscript{3}/Ne\textsuperscript{21}</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Copper</td>
<td>0.171</td>
<td>13</td>
<td>24</td>
<td>Bierni (1958)</td>
</tr>
<tr>
<td>2 GeV</td>
<td>0.13</td>
<td>8.2</td>
<td>29</td>
<td>present paper</td>
</tr>
<tr>
<td>25 GeV</td>
<td>0.17</td>
<td>12</td>
<td>18</td>
<td></td>
</tr>
<tr>
<td>Stainless steel</td>
<td>0.17</td>
<td>10</td>
<td>19</td>
<td>Signer Mier (1960)</td>
</tr>
<tr>
<td>Iron</td>
<td>0.1</td>
<td>5</td>
<td>26</td>
<td>present paper</td>
</tr>
<tr>
<td>25 GeV</td>
<td>0.18</td>
<td>88.1</td>
<td>19</td>
<td></td>
</tr>
</tbody>
</table>

3. COMPARISON WITH CROSS-SECTION CALCULATIONS

In theoretical treatments, the inelastic nuclear events are described according to the following model: when a highly energetic particle is entering a nucleus, a nucleon-nucleon collision may occur. This is normally taken as a collision between free partners. Any nucleon structure or property of nuclear matter is disregarded, but as the nuclear matter is very dense, both collision partners have a certain chance of interacting again with other nucleons. This part of the interaction is known as a "nuclear cascade" and depending on the size of the nucleus, quite a number of particles can be involved in such a process. This process comes to an end either when all cascade particles have left the nucleus or when they are absorbed. A more or less arbitrary threshold is given for the maximum kinetic energy of a nucleon which can be absorbed by the nucleus. This threshold is thought to be near the energies up to which compound nucleus formation is possible (~ 20 MeV). The absorbed energy will be distributed between all degrees of freedom of the whole nucleus so that after the cascade has passed, the nucleus has a smaller number of nucleons than
initially and is left in an excited state. The model describes the part of the interaction following the cascade as "nuclear evaporation". Accidentally, the kinetic energy of a nucleon or a cluster of nucleons may be high enough to allow these particles to leave the nucleus. Between the evaporation steps a rearrangement and new distribution of the excitation energy takes place. Finally, when the energy is not sufficient to evaporate a particle, the energy is emitted as $\beta$ or $\gamma$ radiation.

This model roughly describes the typical interactions that were studied during the present work. However, in the cascade process complex particles and fragments of a nucleus can also be emitted. In many experiments it has been found\textsuperscript{29} that especially for deuterons, alphas, and tritons, etc., the energy distribution of the emitted particles differs from the one expected by the evaporation theory: the forward direction is heavily pronounced in the particle emission and the kinetic energy spectrum has a long tail towards higher energies.

On the other hand, a theoretical treatment for a direct emission of complex particles or nuclear fragments is quite complicated. For very fast deuterons found in nuclear emulsions, a treatment was given by Blokhinstev\textsuperscript{29} who explained the "deuterons" interacting with the incoming protons (660 MeV) as density variations in the Fermi gas-like nucleus. According to the same calculations, fast triton emission is very unlikely.

In the following comparison, we regard triton, helium-3 and helium-4 as evaporated particles whereas neon and argon isotopes are considered as residual nuclei.

1) **Evaporated tritons, helium-3 and alpha particles**

Table 7 shows calculated evaporation cross-sections and measured values. The calculations have been done for an incident proton energy of 600 MeV. The difficulty of predicting the number of evaporated particles comes from the fact that neither the nuclear temperature nor
the excitation energy of the nucleus, after the cascade has passed it, is well known. In our tables we used figures obtained from Monte Carlo calculations by Metropolis et al. The excitation energy has been plotted versus the atomic number at different energies (Fig. 21). Dostrovsky et al. have used the Monte Carlo method to calculate the number and the energy distribution of the evaporated particles (Fig. 22). The number of evaporated particles of any type is roughly proportional to the excitation energy but the curves flatten off at high excitation. The amount of energy carried away by an evaporated nucleon rises with the nuclear temperature. The agreement of the measured values (Table 5) and the prediction of the Monte Carlo calculation is fairly good if, for the tritium calculation, the parameters $A/10 V_0$ are used; for helium-3 values between $A/10 V_0$ and $A/10 V_1$; and for helium-4, $A/10 V_1$ and $A/20 V_0$, where the parameters used are those of Dostrovsky et al. The spectrum of evaporated tritons, helium-3 and helium-4 from bombardment of copper is shown in Fig. 16. The kinetic energy of these particles is low so that losses from the targets can be excluded. The average kinetic energy of the calculated spectrum is 11 MeV for tritons, 13 MeV for helium-3 and 14.5 MeV for $\alpha$ particles. Measurements have been made indicating that the tritium losses are very unlikely in a target thickness more than 0.1 g/cm$^2$.

ii) Argon and neon isotopes as residual nuclei

The distribution of the residual nuclei can be calculated, assuming an exponential decrease of the cross-sections to smaller masses and a Gaussian distribution for the different isotopes of the same element. This is expressed in the following formula first given by Rudstam:

$$(Z,A) \text{ mb} = \exp[PA - Q - R(Z - SA)^2].$$ (1)
This formula has recently\textsuperscript{33}) been modified to allow for the higher experimental yields for large \((Z - S A)\):

\[ (Z, A) \text{mb} = K \exp[PA - R | Z - SA + TA^2 |^{3/2}] \quad (2) \]

From determinations of other spallation products at different energies\textsuperscript{33}) the parameters \(P, Q, R\) and \(S\) can be chosen with a least-squares fit. For 600 MeV, Table 8 contains the values expected according to this formula which can be compared with the experimental results (Fig. 23) also shown.

Agreement within a factor of two for argon is quite satisfactory. Figure 19 shows how the cross-sections of argon and neon depend on energy. Argon production cross-sections in target materials with atomic numbers near to argon remain constant between some 600 MeV up to 25 GeV. The neon isotopes, however, have cross-sections rising with increasing energy. Neon may be produced to a large extent as a fragmentation product; this may also be seen from the neon excitation curve.

The results of this work are not discussed with regard to meteorite application. It is planned to give such a treatment together with more and newer meteorite data elsewhere\textsuperscript{3}).

\*

\*[It is important to note that the Gaussian distribution [formula (1)] represents the measured values quite well for iron and copper with the following parameters:

\[ P = 1.7, \quad R = 2.1, \quad S = 0.476. \]

For nickel, however, \(S\) has to be modified to 0.477 to allow for the fact that the stable nickel isotopes do not lie near the general line for stable nuclei in the energy valley. If an average of the parameters used at this proton energy\textsuperscript{33}) given in other publications is made, the following parameters would appear to be more likely:

\[ P = 1.8 \text{ and } S = 0.473. \]

The production cross-sections obtained in this work do not indicate clearly which of the two formulae predict more accurately experimental values. However the argon-42 result, measured in copper, seems to favour formula (2), since the experimental value is double that calculated from formula (1).]
ACKNOWLEDGEMENTS

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## Table 1

**Corrections for internal proton fluxes**

<table>
<thead>
<tr>
<th>Target No.</th>
<th>&quot;Weighted&quot; thickness g/cm²</th>
<th>Proton flux corrections when measured by</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>a) Al²⁷ → Na²⁴</td>
</tr>
<tr>
<td>I₁₂</td>
<td>0.2</td>
<td>-</td>
</tr>
<tr>
<td>I₁₅</td>
<td>1.2</td>
<td>- 4%</td>
</tr>
<tr>
<td>I₁₁</td>
<td>1.9</td>
<td>- 6%</td>
</tr>
<tr>
<td>I₁₀</td>
<td>2.5</td>
<td>-10%</td>
</tr>
<tr>
<td>I₂</td>
<td>3.3</td>
<td>-12%</td>
</tr>
<tr>
<td>I₂₀</td>
<td>6.0</td>
<td>-22%</td>
</tr>
<tr>
<td>I₂₄</td>
<td>7.1</td>
<td>-26%</td>
</tr>
<tr>
<td>I₁₂</td>
<td>7.7</td>
<td>-29%</td>
</tr>
<tr>
<td>I₅</td>
<td>11.9</td>
<td>-25%</td>
</tr>
<tr>
<td>I₆</td>
<td>15.4</td>
<td>-56%</td>
</tr>
<tr>
<td>(FS 1)</td>
<td>2.7</td>
<td>-10%</td>
</tr>
<tr>
<td>(FS 2)</td>
<td>3.5</td>
<td>-12%</td>
</tr>
<tr>
<td>I₁₆</td>
<td>1.6</td>
<td>- 5%</td>
</tr>
<tr>
<td>I₂₁</td>
<td>1.8</td>
<td>- 6%</td>
</tr>
</tbody>
</table>

Col. 2) gives only approximate figures as the corrections depend on the position of the material in the target.
<table>
<thead>
<tr>
<th>Target No.</th>
<th>End of Exposure date</th>
<th>Exposure time hours. min.</th>
<th>Proton energy MeV</th>
<th>Monitor reaction mb</th>
<th>dpm</th>
<th>Monitor foil thickness atoms/cm²</th>
<th>Protons uncorrected</th>
<th>Corrections %</th>
<th>Total number of protons on target</th>
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</thead>
<tbody>
<tr>
<td>I 11</td>
<td>5. 7.60</td>
<td>10.42</td>
<td>239</td>
<td>600</td>
<td>T in Al 32</td>
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<tr>
<td>I 12</td>
<td>5. 7.60</td>
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<td>240</td>
<td>600</td>
<td>T in Al 32</td>
<td>$2.14 \times 10^5$</td>
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<td>5.35 x 10^{16}</td>
<td>4.95 x 10^{16}</td>
</tr>
<tr>
<td>I 10</td>
<td>5. 7.60</td>
<td>06.12</td>
<td>180</td>
<td>600</td>
<td>T in Al 32</td>
<td>$2.22 \times 10^5$</td>
<td>$13.10 \times 10^{20}$</td>
<td>5.00 x 10^{16}</td>
<td>4.90 x 10^{16}</td>
</tr>
<tr>
<td>I 15</td>
<td>27. 3.61</td>
<td>02.29</td>
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<td>600</td>
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<tr>
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<td>3.10 x 10^{16}</td>
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<td>12. 2.62</td>
<td>01.14</td>
<td>60</td>
<td>600</td>
<td>Al^{27} $\rightarrow$ Na^{24} 11.0</td>
<td>$3.20 \times 10^7$</td>
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<td>1.70 x 10^{16}</td>
<td></td>
</tr>
<tr>
<td>I 2</td>
<td>23. 9.58</td>
<td>10.59</td>
<td>20</td>
<td>600</td>
<td>Al^{27} $\rightarrow$ Na^{24} 11.0</td>
<td>$3.80 \times 10^6$</td>
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<td>9</td>
<td>600</td>
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<td>$1.70 \times 10^6$</td>
<td>$2.25 \times 10^{20}$</td>
<td>8.95 x 10^{14}</td>
<td></td>
</tr>
<tr>
<td>I 6</td>
<td>9. 9.59</td>
<td>03.25</td>
<td>5</td>
<td>600</td>
<td>Al^{27} $\rightarrow$ Na^{24} 11.0</td>
<td>$1.16 \times 10^6$</td>
<td>$2.25 \times 10^{20}$</td>
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<td></td>
</tr>
<tr>
<td>I 21</td>
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<td>04.26</td>
<td>150</td>
<td>600</td>
<td>Al^{27} $\rightarrow$ Na^{24} 11.0</td>
<td>$9.20 \times 10^7$</td>
<td>$2.25 \times 10^{20}$</td>
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<td>2200</td>
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<td>Argon-37 cross-section</td>
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<td>$1.22 \times 10^{4}$</td>
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<tr>
<td></td>
<td>Mg</td>
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<tr>
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<tr>
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<tr>
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<td>Al</td>
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<td>$0.83 \times 10^{4}$</td>
<td>36, $32 \pm 3$</td>
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<tr>
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<td>Tritium cross-section (mb)</td>
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<td>Argon-57 cross-section (mb)</td>
<td>Fertility</td>
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<td>45</td>
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<td>2.90 ± 0.7</td>
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<tr>
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<td>45</td>
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<tr>
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<td>99.99%</td>
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<td></td>
</tr>
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<td>-</td>
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<td></td>
</tr>
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<td>37</td>
<td>0.99 x 10^6</td>
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<td>99.99%</td>
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<td>37</td>
<td>0.99 x 10^6</td>
<td>-</td>
<td>99.99%</td>
<td></td>
<td></td>
</tr>
<tr>
<td>E 4</td>
<td>Fe</td>
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<td>4.36 x 10^4</td>
<td>37</td>
<td>0.99 x 10^6</td>
<td>-</td>
<td>99.99%</td>
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Table 3 (cont.)

Fertility: 99.9% pure iron, 99.9% reduced in H₂; 99.99% pure iron, 99.99% reduced in H₂.
Table 3 (cont.)

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<tr>
<th>Target No.</th>
<th>Target material</th>
<th>Sheet No.</th>
<th>Purity</th>
<th>Thickness of sheet atoms/cm²</th>
<th>No. of protons in target</th>
<th>Tritium activity dpm</th>
<th>Tritium cross-section mean value mb</th>
<th>Argon-37 activity dpm</th>
<th>Argon-37 cross-section</th>
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<td>Cu</td>
<td>(2)</td>
<td>electrolyte</td>
<td>$4.23 \times 10^{21}$</td>
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<td>$1.07 \times 10^{4}$</td>
<td>$55$</td>
<td>$3.63 \times 10^{5}$</td>
<td>1.76</td>
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<tr>
<td>I 15</td>
<td>Cu</td>
<td>(3)</td>
<td>99.5%</td>
<td>$4.71 \times 10^{20}$</td>
<td>$3.10 \times 10^{16}$</td>
<td>$8.10 \times 10^{4}$</td>
<td>$52$</td>
<td>$3.33 \times 10^{5}$</td>
<td>1.74, 1.75 ± 0.5</td>
</tr>
<tr>
<td></td>
<td>Cu</td>
<td>(5b)</td>
<td>99.999%</td>
<td>$1.67 \times 10^{21}$</td>
<td>$3.10 \times 10^{16}$</td>
<td>$2.93 \times 10^{5}$</td>
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<td>$12.80 \times 10^{5}$</td>
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<tr>
<td>1</td>
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<td>pure</td>
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<td>$4.28 \times 10^{14}$</td>
<td>$1.15 \times 10^{4}$</td>
<td>$86 \pm 10$</td>
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</tr>
<tr>
<td>I 5</td>
<td>Pb</td>
<td>(13)</td>
<td>ordinary</td>
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<td>$2.95 \times 10^{4}$</td>
<td>$122$</td>
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<tr>
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<td>Pb</td>
<td>(14)</td>
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<td>$131$</td>
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<tr>
<td></td>
<td>Bi</td>
<td>pure</td>
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<td>$4.13 \times 10^{21}$</td>
<td>$7.50 \times 10^{14}$</td>
<td>$3.23 \times 10^{4}$</td>
<td>$98 \pm 15$</td>
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Note: Target No. in chronological order
I internal target
E external target
Table 4

T and $^{A_{37}}$ production cross-sections at 0.6, 2.2 and 25 GeV

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<th>Target</th>
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<th>$^{A_{37}}$</th>
<th>$^{A_{39}}$</th>
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<td>GeV</td>
<td>mb</td>
<td>GM counter</td>
<td>Mass spectrometer for comparison</td>
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<td>15 ± 4</td>
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</tr>
<tr>
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<td>0.6</td>
<td>22 ± 2</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2.2</td>
<td>75</td>
<td></td>
<td></td>
</tr>
<tr>
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<td>25</td>
<td>71</td>
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</tr>
<tr>
<td>Si</td>
<td>0.6</td>
<td>18 ± 3</td>
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</tr>
<tr>
<td>V</td>
<td>0.6</td>
<td>52 ± 8</td>
<td></td>
<td>4.2 ± 0.5</td>
</tr>
<tr>
<td>Cr</td>
<td>0.6</td>
<td>49 ± 8</td>
<td></td>
<td>3.8 ± 0.7</td>
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<td>Mn</td>
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<td>60 ± 10</td>
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</tr>
<tr>
<td>Fe</td>
<td>0.6</td>
<td>48 ± 5</td>
<td>66 ± 10*</td>
<td>6.1 ± 0.3</td>
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<td>104 ± 10</td>
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<td>10.0 ± 1</td>
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<td>135 ± 9</td>
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<tr>
<td>Ni</td>
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<td>41 ± 15</td>
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<td>1.75 ± 0.5</td>
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<td>132 ± 8</td>
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<td>1.75 ± 0.5</td>
<td>2.5</td>
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<td>132 ± 8</td>
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<td>86 ± 10</td>
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*) Not including a systematic error in monitoring and degassing.

**) " " " " " " " monitoring.
### Table 5

Mass spectrometric measurements for rare gas isotope production cross-sections

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<th>$\text{A}^{37}$ (mb)</th>
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<td>(17)</td>
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<td>-</td>
<td>2.4</td>
<td>(1.2)</td>
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<td>12.6</td>
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<td>785</td>
<td>-</td>
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<td>(0.07)</td>
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Table 6

Spallation yields in complex targets

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<th>Target</th>
<th>Proton energy GeV</th>
<th>Proton flux</th>
<th>T</th>
<th>He$^3$</th>
<th>He$^4$</th>
<th>Ne$^{20}$</th>
<th>Ne$^{21}$</th>
<th>Ne$^{22}$</th>
<th>A$^{36}$</th>
<th>A$^{37}$</th>
<th>A$^{38}$</th>
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<th>A$^{42}$</th>
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<td>9.3</td>
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<td>(0.09)</td>
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<td>Ramsdorfer</td>
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<td>GMC</td>
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Rare gas isotopes and T produced in olivine and in the chondrite Ramsdorfer.

(The chemical composition of olivine is: 29% Mg, 7.2% Fe and 64% SiO$_2$.

Ramsdorfer consists of 21% Si, 13.3% Mg, 1.8% Al, 1.8% Ca, 4.3% O$_2$, 18.3% Fe and 1.3% Ni.)

MS = Mass Spectrometer; GMC = Geiger-Müller Counter.
They are given for 0.45 GeV < getRequest proton energy.

Calculated values for proton were obtained by Professor Postnovsky (personal communication).

<table>
<thead>
<tr>
<th>Energy (GeV)</th>
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<th>Experimental Wert</th>
<th>Theoretical Wert</th>
<th>Experimental Wert</th>
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Table 7
Cross-sections of rare gas isotopes calculated by spallation formulae

\[ \sigma_{X}(t, A) = \exp[\frac{P_{A} - P_{X}}{a_{A} + 1}] \]

where \( k = e^{-q} \)

| Target | \( \sigma_{mb} \) | Producing Isotope | \( ^{20} \text{Ne} \) | \( ^{21} \text{Ne} \) | \( ^{22} \text{Ne} \) | \( ^{23} \text{Ne} \) | \( ^{24} \text{Ne} \) | \( ^{25} \text{Ne} \) | \( ^{26} \text{Ne} \) | \( ^{27} \text{Ar} \) | \( ^{28} \text{Ar} \) | \( ^{29} \text{Ar} \) | \( ^{30} \text{Ar} \) |
|--------|----------------|-----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|
| \( ^{4} \text{He} \) | \( ^{36} \text{Ar} \) | 1.13 | 1.44 | 2.07 | 0.79 | 0.76 | 0.9 | 0.027 | - | 0.068 | 0.71 | 0.44 | 0.29 | 0.54 |
| \( ^{4} \text{He} \) | \( ^{37} \text{Ar} \) | 1.57 | 4.65 | 3.96 | 1.86 | 0.76 | 0.76 | 0.058 | - | 0.19 | 0.88 | 0.29 | 0.13 | - |
| \( ^{4} \text{He} \) | \( ^{38} \text{Ar} \) | 2.06 | 9.8 | 9.51 | 5.17 | 0.76 | 0.76 | 0.07 | - | 0.19 | 4.89 | 0.12 | - | - |
| \( ^{4} \text{He} \) | \( ^{39} \text{Ar} \) | 2.06 | 5.58 | 3.40 | 2.82 | 0.76 | 0.76 | 0.07 | - | 0.17 | 4.82 | 0.12 | - | - |
| \( ^{4} \text{He} \) | \( ^{40} \text{Ar} \) | 2.06 | 9.84 | 9.51 | 5.17 | 0.76 | 0.76 | 0.07 | - | 0.17 | 4.82 | 0.12 | - | - |
| \( ^{4} \text{He} \) | \( ^{41} \text{Ar} \) | 2.06 | 5.58 | 3.40 | 2.82 | 0.76 | 0.76 | 0.07 | - | 0.17 | 4.82 | 0.12 | - | - |
| \( ^{4} \text{He} \) | \( ^{42} \text{Ar} \) | 2.06 | 9.84 | 9.51 | 5.17 | 0.76 | 0.76 | 0.07 | - | 0.17 | 4.82 | 0.12 | - | - |

Table 8
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* * *

8150/p/mn
FIGURE CAPTIONS

Fig. 1: Target Arrangements for the Internal SC Irradiations

Three aluminium monitor foils were placed near the front, behind the target. The target was mounted on a probe target which permits the selection of the energy for the irradiation by varying the radius.

Fig. 2: Activity of an aluminium monitor foil in the radial direction in the target. The unbroken line gives the radial decrease of the activity in the target. It can be seen that the activity drops off very quickly towards higher radii.

Fig. 3: Target position in the internal "Saturpe" (Saclay) irradiations and composition of the target.

Fig. 4: Internal FS irradiations

Left) Flip target with its position in the accelerating tube.
Right) Percentage distribution of the activity in one of the monitoring foils.

V.C. = vacuum chamber; M.C. = machine centre;
shaded area = approx. beam size.

Fig. 5: Lay-out for the external target exposures at the SC

SEC : secondary emission chamber for the flux determinations during exposure.
ICH : ion chamber.
BM : bending magnet for the proton beam transport.

Monitor foils are arranged to give the percentage distribution of the total beam in the target.
Q : quadrupole lenses.
Fig. 6: Cross-sections of $\text{Al}^{27}(p,2pn)\text{Ne}^{24}$ versus proton energy. Above 100 MeV, the cross-section is taken as nearly constant.

Fig. 7: The production ratio of $\text{Na}^{24}/F^{18}$ by high-energy protons in aluminium.

Fig. 8: Cross-sections of the reaction $\text{Al}^{27}(n,\alpha)\text{Ne}^{24}$ versus neutron energy.

Fig. 9: Corrections for the proton flux measurements by aluminimum monitor foils.

The ordinate on the left side gives the additional production of $\text{Na}^{24}$ by secondary neutrons; the ordinate right side, indicates the additional production of $F^{18}$ by fast secondaries. For the corrections according to secondary neutrons, the abscissa values giving the target thickness in $g/cm^2$ were obtained as follows:

the target thickness was multiplied by a factor $f$ which depends on the atomic number of the target material. The values used are the following:

\[
\begin{align*}
  f(\text{Al}) f(\text{Mg}) f(\text{Si}) f(\text{Olivine}) & = 1 \\
  f(\text{Fe}) f(\text{Ni}) f(\text{Co}) f(\text{Cu}) & = 1.4 \\
  f(\text{Ag}) & = 1.7 \\
  f(\text{Pb}) f(\text{Bi}) & = 1.8 
\end{align*}
\]

Fig. 10: Gas extraction apparatus

- Be : Beryllium oven
- Ca : Calcium oven
- CuO : Copper monoxide oven
- McL : McLeod gauges
- DP : Transfer pumps
- Pd : Palladium thimble
- $S_1, S_2$ : Mercury stopcocks
- TG : Thermocouple gauge head
- TP1, TP2 : Toepler pumps
- Hg : Hydrogen reservoir
- A : Argon reservoir
- L : Sample lift.
Fig. 11: Different crucible arrangements:

I Lid and crucible support in Mo.

II Lid and crucible support in Al₂O₃.

The Al₂O₃ lid does not heat up too much, so that the vapour condenses on it.

Fig. 12: Gas extraction from the target

The diagram represents a typical degassing curve (target material Ramsdorf meteorite). The ordinate on the left side (top) gives the flow-rate with open stopcock S₁ (Fig. 10). If stopcock S₁ is closed, the gas flow-rate is inversely proportional to the time elapsed, during which the pressure has risen from p₀ to p₁ (bottom, right). At 13.45 when the generator was switched on (↑) strong degassing occurred. At the moment when the sample starts to melt, degassing decreases but rises again afterwards (m). At the point h the temperature was increased in steps. When discharges occurred (d), gas escaped from the degassing vessel. In most cases, the generator was switched off immediately. (↓) The flow-rate is given by

\[
\frac{2 \times 10^{-5}}{t₁} = \text{torr} \frac{t}{\text{sec}}.
\]

Fig. 13: Apparatus for calibrating radioactive gas samples

B : barometric height
C : ground tapered joint for connection of counter
G : McLeod gauge
R : gas reservoir
S : double ball stopcock
T : Toepler pump.
Fig. 14: $H^3/H$ diffusion ratio through palladium.

The quantity $\tau = (\Delta H^3/\Delta H)(H_2/H_3^3)$ is plotted versus palladium temperature. At temperatures higher than $450^\circ C$ the ratio $\Delta H^3/\Delta H$ which diffuses in a certain time through the palladium is equal to the initial ratio of the two isotopes $H_2^3/H_2$.

Fig. 15: Degassing apparatus and mass spectrometer

$F = $ furnaces for bake out  
$Q = $ quartz shield  
$HF = $ high frequency coil from induction heater (10 kw, 600 kc)  
$MoC = $ Molybdenum crucible  
$Ca = $ Calcium furnace heated to $600^\circ C$  
$Cu-CuO = $ Copper oxide furnace heated to $400^\circ C$.  
$ChC = $ Charcoal, cooled with liquid air  
$Ct = $ Cool trap  
$MV = $ metal valve, bakeable to $400^\circ C$.  
$AG = $ Alpert gauge  
$IS = $ Ion source  
$IM = $ Ion multiplier (system of Dumont 6269)  
$Al_2O_3 = $ Porous alumina getter  
$P = $ Pipettes system (see underneath figure)

Systems for preparing calibrated amounts of about $10^{-7}$ to $10^{-6}$ cc of rare gases. To the left is a mercury pipette. A capillary is filled by mercury cut-offs to a 5 litre reservoir. The rectangular arms are ground glass seals and can be turned through $180^\circ$. To the right a metal valve pipette, where the mercury cut-offs are replaced by bakeable metal valves with membranes. The capillary consists of a metal tubing (1 mm diameter).
Fig. 16: Decay curve for the radioactive argon

The slope on the logarithmic graph corresponds exactly to the argon-37 decay constant; the constant activity is due to argon-39.

Fig. 17: Tritium production in different target elements

a) H³ excitation curves for C, Ni, Cu and Ag
b) for Al, Fe, Pb.

The letters near the points in the graph refer to other publications. The points given with error bars are results from the present work.

Fig. 18: H³, He³, He⁴, A³⁸ Ne²⁴ production cross-section versus atomic number at 600 MeV.

Fig. 19: The production cross-section of rare gases and tritium in iron versus proton energy.

Fig. 20: Production ratios in iron versus energy. All ratios found in iron meteorites lie within the shaded regions.

Fig. 21: Excitation energy of atomic nucleus A after proton bombardment of 0.5 and 2.2 GeV. All values are taken from graphs by J. Dostrovsky, P. Rabinowitz and R. Rivins. The straight line gives the excitation energy in the region of AgBr and Ru¹⁰⁶ (See Ref. 33).

Fig. 22: Energy distribution of evaporated tritium, helium-3 and helium-4 in iron according to Monte Carlo calculations (also by Dostrovsky et al. and personal communication).
Fig. 23: Calculated and measured argon cross-sections in Fe, Cu and Ni. The calculations are made with the Rudstam spallation formula:

\[ \sigma(Z,A) = \exp \left[ PA - Q - R (ZSA)^2 \right] \]

The parameters used for these calculations were:

<table>
<thead>
<tr>
<th></th>
<th>P</th>
<th>Q</th>
<th>R</th>
<th>S</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe</td>
<td>0.17</td>
<td>4.2</td>
<td>2.1</td>
<td>0.476</td>
</tr>
<tr>
<td>Cu</td>
<td>0.17</td>
<td>4.8</td>
<td>2.1</td>
<td>0.476</td>
</tr>
<tr>
<td>Ni</td>
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<td>4.45</td>
<td>2.1</td>
<td>0.477</td>
</tr>
</tbody>
</table>

* * *

8150/p/mm
Na$^{24}$ Distribution in Al - Monitor Foils

\[ \text{FIG. 2} \]
Al$^{27}$(p,3pn)Na$^{24}$ REACTION CROSS SECTION

△ MARQEZ 1952 (34)
○ TURKEVICH 1954 (40)
▲ FRIEDLANDER, HUDIS, WOLFGANG 1955 (41)
■ CURRIE, LIBBY, WOLFGANG 1956 (18)
▽ CRANDALL, MILBURN, PYLE, BIRNBAUM 1956 (35)
● HICKS, STEVENSON, NERVIK 1956 (36)
○ JONES 1956 (37)
▼ PROSKOSHIN, TIAKIN 1957 (38)
◆ BARR 1957 (42)
□ CUMMING, FRIEDLANDER, SWARTZ 1958 (43)
◇ BENIOFF 1960 (44)
★ GOEBEL, SCHULTES et al. 1960 (13)
▼ YULE, TURKEVICH 1960 (39)
十 CUMMING, FRIEDLANDER, HUDIS, POSKANZER 1962 (45)
$F^{18}/\text{Na}^{24}$ RATIO IN ALUMINIUM

**FIG. 7**

- ○ Hintz, Ramsey 1952 (46)
- ◆ Chackell, Reasbeck, Symonds, Warren 1956 (47)
- □ Parikh 1960 (48)
- + Friedlander, Hudis, Wolfgang 1955 (41)
- ● Cumming, Poskanzer, Kaufman 1962 (49)
- × Goebel, Schultes 1960 (13)
FIG. 8

Al$^{27}(n,\alpha)$ Na$^{24}$

- Mani 1960 (50)
- Grundl 1958 (51)
- Kern 1959 (52)
- Gabbard, Kern 1962 (53)
- Tewes (54)
- Schmidt, Halperin 1961 (55)
- Bayhurst, Prestwood 1961 (56)
- Upper limit: Knox 1949 (57)
- Marquez 1952 (58)
- Mean value (59)

$Q = -3.11$
Flux Corrections for Internal Proton Irradiations

Excess of proton flux measured by Al\textsuperscript{27} No\textsuperscript{27}.
GAS EXTRACTION APPARATUS

FIG. 10
SYSTEM II

NS 60

Al₂O₃
Quartz
Al₂O₃
Mo
Al₂O₃
Quartz support

SYSTEM I

NS 60

Mo Lid
Al₂O₃
Mo
Mo strips
Quartz
Fig. 15 Degassing apparatus and mass spectrometer.
Fig. 18 Cross sections of $H^3$, $He^3$, $He^4$, $Ne^{21}$, $A^{38}$ in dependence of atomic weight of elements at 600 MeV proton energy.
Fig. 19 The dependence of tritium and rare gas cross sections on energy for iron.
Fig. 20 PRODUCTION RATIOS AS A FUNCTION OF ENERGY
Fig. 23 ARGON PRODUCTION CROSS SECTIONS AT 600 MeV PROTON ENERGY