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"STUDIES ON THE RADIOACTIVE DATING OF THE LUNAR SURFACE"

by

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I. EXPERIMENTAL AND TECHNICAL STUDIES

I. 1 Introduction

The extraction and analysis of rare gases from silicate materials, such as terrestrial rocks and stone meteorites, is routinely performed in many laboratories. From the measured concentrations and isotopic compositions, K/Ar, U, Th/He, I^{129}/Xe^{129} ages, radiation ages and other data can be derived. The gases are released by heating and melting the sample in specially designed extraction systems at temperatures of up to 1800°C. Induction or internal resistance heaters are used and the power consumption is in the order of several kwh. A variety of getters cleans the evolved gases. After several purification steps the rare gases are analyzed in suitable mass spectrometers. The analysis of a single sample requires a large number of individual operations, especially during the extraction and purification stage.

In the framework of our studies on radioactive dating of the lunar surface, the feasibility of remote-controlled rare gas analysis had to be investigated. The main problem was to show that a simple, lightweight, rugged rare gas extraction and purification system with negligible external power consumption could be developed. No straightforward adaptation of currently available extraction systems seemed possible due to their high power consumption. No serious difficulties are expected for the remote-controlled mass spectrometric analyses of the extracted rare gases. Mass spectrometers similar to those used in upper atmosphere research (cf. Nier et al., 1964) could be employed.

I. 2 Development of a Rare Gas Extraction System Suitable for Remote-Controlled Operation

To liberate rare gases quantitatively from silicate materials, temperatures of 1500°C and higher are necessary (Geiss and Hess, 1958; Reynolds, 1963). The required temperature can be lowered considerably by using a flux such as NaOH at 600° (Wasserburg and Hayden, 1954) or $Na_2B_4O_7$ at 1100-1200° (Geiss and Hess, 1958). However, flux has several drawbacks: relatively long reaction times, evolution of large quantities of gases such as H_2 in the case of NaOH, difficulties in removing atmospheric contamination, and foaming.

Wet chemical techniques could also be used for the extraction of rare gases. Paneth and coworkers dissolved iron meteorites in sulfuric acid saturated with potassium persulfate and were able to recover He and Ne quantitatively (Chackett, Reasbeck and Wilson, 1953). However, hydrofluoric acid would be required for dissolving silicates and an elaborate purification system would be necessary to remove water vapors, etc. The recovery of Ar and the heavier rare gases from the solution might also be difficult.

We therefore decided to develop a high-temperature extraction system with a chemical reaction as heat source. Thermite, a mixture of iron oxide and aluminum, seemed a suitable energy source. Temperatures higher than 1500°C are reached and the heat of solidification of the molten iron would prolong the period of time the sample is at high temperature. Figure 1 shows a cross section of the final version of the complete extraction and purification system. The sample is contained in a Mo-extraction crucible. The walls of the extraction crucible are lined with Ti-sponge, held in place by a Mo-insert. The Ti-sponge acts as getter for the cleanup of the evolved gases. A circular opening, ~7 mm in diameter, allows the introduction of the sample after the whole extraction system has been assembled. All parts which are in contact with the vacuum system are thoroughly pre-degassed. Brazing is done in high vacuum. The thermite

mixture is held in a graphite crucible. A graphite liner prevents the contact of the molten iron with the Mo-crucible. A Mo-radiation shield acts also as supporting structure of the graphite crucible. The whole assembly is enclosed in a sealed can, preventing the escape of any gas from the reaction. The can is filled with CO_2 at 1/3 atmosphere to facilitate the ignition of the thermite (ignition in vacuum proved to be very difficult). The total weight of the system ready to operate is below 500 g, the diameter 7 cm, and the length 13 cm.

Several complete systems were built and tested, some with slightly larger overall dimensions. A large number of tests were also made with the outer can replaced by a flanged vacuum container.

The same basic principle of design can also be used to heat several Mo-crucibles simultaneously, thus permitting the simultaneous extraction of several samples. Such a four-sample extraction system was developed and tested. Some of the results reported here were obtained with four-sample thermite extraction systems.

I. 3 Performance of the Extraction System

The temperature of the bottom of the Mo-crucible was measured with a W-W, Re thermocouple. The exact location of the thermocouple junction is indicated in Figure 1. From melting point determinations it was concluded that the measured temperature agrees within 20 to 30°C with the temperature reached by the sample. Figure 2 shows a typical temperature curve. A maximum temperature higher than 1600°C was reached. The maximum temperature depends on several design parameters, as can be seen from Figures 3 and 4.

The overall extraction and pruification performance of the system was tested with 0.5 to 2 g samples of stone meteorites. The temperatures reached were sufficient to completely melt the meteorite sample into a

glassy bead. The total pressure during the extraction was measured with a Pirani gauge. Figure 5 shows a typical pressure-time curve. Shortly after the ignition, the pressure raises to approximately 100 mm Hg, but within 20 minutes the Ti-sponge getter reduces it by 5 orders of magnitude. The clean-up is sufficient to allow a direct mass spectrometric analysis of the extracted rare gases. Figure 6 shows the mass spectrum in the mass range of 2 to 44 obtained from the extraction of approximately 2 grams of the stone meteorite Finney. The thermite extraction system had been connected to a statically operated UHV mass spectrometer. After the extraction system had cooled down (approximately 20 min.), the gases were admitted to the mass spectrometer and the spectrum given in Figure 6 was obtained. Interference from background was well below 5 percent for Ar^{40}, $He^{3,4}$ and $Ne^{21,22}$. Some background corrections had to be applied for $Ar^{36,38}$. The presence of Ar^{++} prevented the measurement of Ne^{20}. The background could be further reduced if a second (cold) getter would be added.

In Table 1 the He, Ne and Ar isotopic compositions obtained in this extraction are given and compared with values from standard laboratory extraction systems (Eberhardt et al., 1966). The agreement is within the assigned errors. The total amounts of He and Ne released were estimated from ion beam intensities and agree with the values obtained with the isotopic dilution method (Eberhardt et al., 1966).

The extraction yield for Ar was tested in several extractions of the same meteorite. The results are given in Table 2. All amounts have been estimated from the ion beam intensities in the mass spectrometer and may be subject to errors of up to 10 percent. The average concentration for the thermite extractions agrees within 5 percent with the result of the conventional method. A re-extraction of the argon from the glassy melt of sample remaining in the extraction crucible gave less than one percent of the total Ar^{40}. These results indicate that He, Ne and Ar are quantitatively released, despite the relatively short heating time.

II. THEORETICAL STUDIES

Under this grant, a general theoretical study on age determinations and other applications of rare gas measurements on lunar surface material was carried out and published by P. Eberhardt (1964). This work has shown the great importance which should be attached to rare gas measurements in lunar surface material. Such measurements could be applied for the determination of the ages of surface features of the moon. The history of the high energy particle radiation field in the solar system can be investigated by measuring spallation products in lunar surface material. From this, results superior to those obtained from measurements on meteorites and on terrestrial samples can be expected. Spallation products and neutron produced isotopes can be used for studying erosion rates at the surface of the moon. All these methods are outlined and discussed in detail in the paper by P. Eberhardt (1964).

In a letter to the Journal of Geophysical Research we have suggested, in collaboration with P. Signer from the University of Minnesota, an experiment for the determination of the composition of the solar wind (P. Signer, P. Eberhardt, and J. Geiss, 1965). The method outlined in this publication was in part conceived as a result of the work performed under this grant.

III. INVENTIONS

No inventions required to be reported have been made in the performance of work under the grant.

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Signer P., P. Eberhardt, and J. Geiss; Possible determination of the solar wind composition; J. Geophys. Res. 70 (9), 2243-2244, 1965.

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Extraction method	$\frac{He^3}{He^4}$	$\frac{Ne^{22}}{Ne^{21}}$	$\frac{Ar^{36}}{Ar^{38}}$	$\frac{Ar^{40}}{Ar^{36}}$	He^4	Ne^{21}
4-sample thermite extraction system	27 ±3	1.25 ±0.1	1.80 ±0.35	3000 ±500	700 ±100	4.2 ±0.6
Conventional (Eberhardt, Eugster Geiss and Marti, 1966)	24.1 ±0.6	1.18 ±0.01	1.67 ±0.05	3620 ± 70	708 ± 15	3.92 ±0.10

Table 1 Isotopic composition and absolute amounts of rare gases extracted from the meteorite Finney.

Extraction method	Sample size, g	Ar^{40} x 10^5 cc STP/g
Thermite	0.50	4.6
Thermite	1.24	4.6
Thermite	0.55	5.0
Thermite	0.50	5.7
Thermite	0.50	5.0
Average thermite (standard deviation)		5.0 ± 0.4
Conventional		5.2

Table 2 Argon-40 yield of 5 different thermite extractions on samples of a stone meteorite. For a detailed description of the conventional extraction system see Eberhardt et al., 1966. All rare gas concentrations were estimated from the ion beam intensity in the mass spectrometer and may be subject to errors of up to 10 percent.

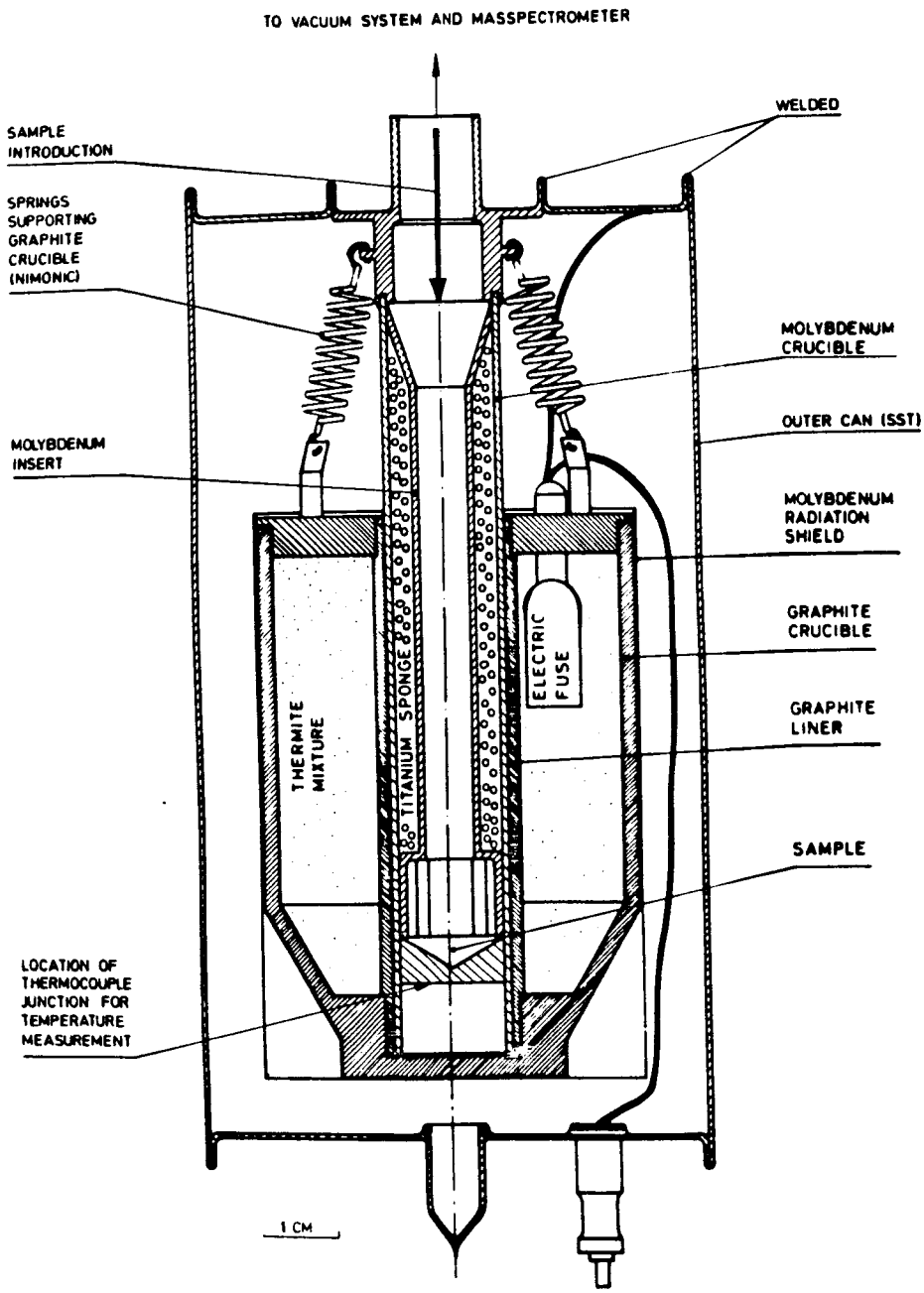

FIG 1 ONE SAMPLE EXTRACTION AND PURIFICATION SYSTEM WITH THERMITE HEATING (MODEL 63)

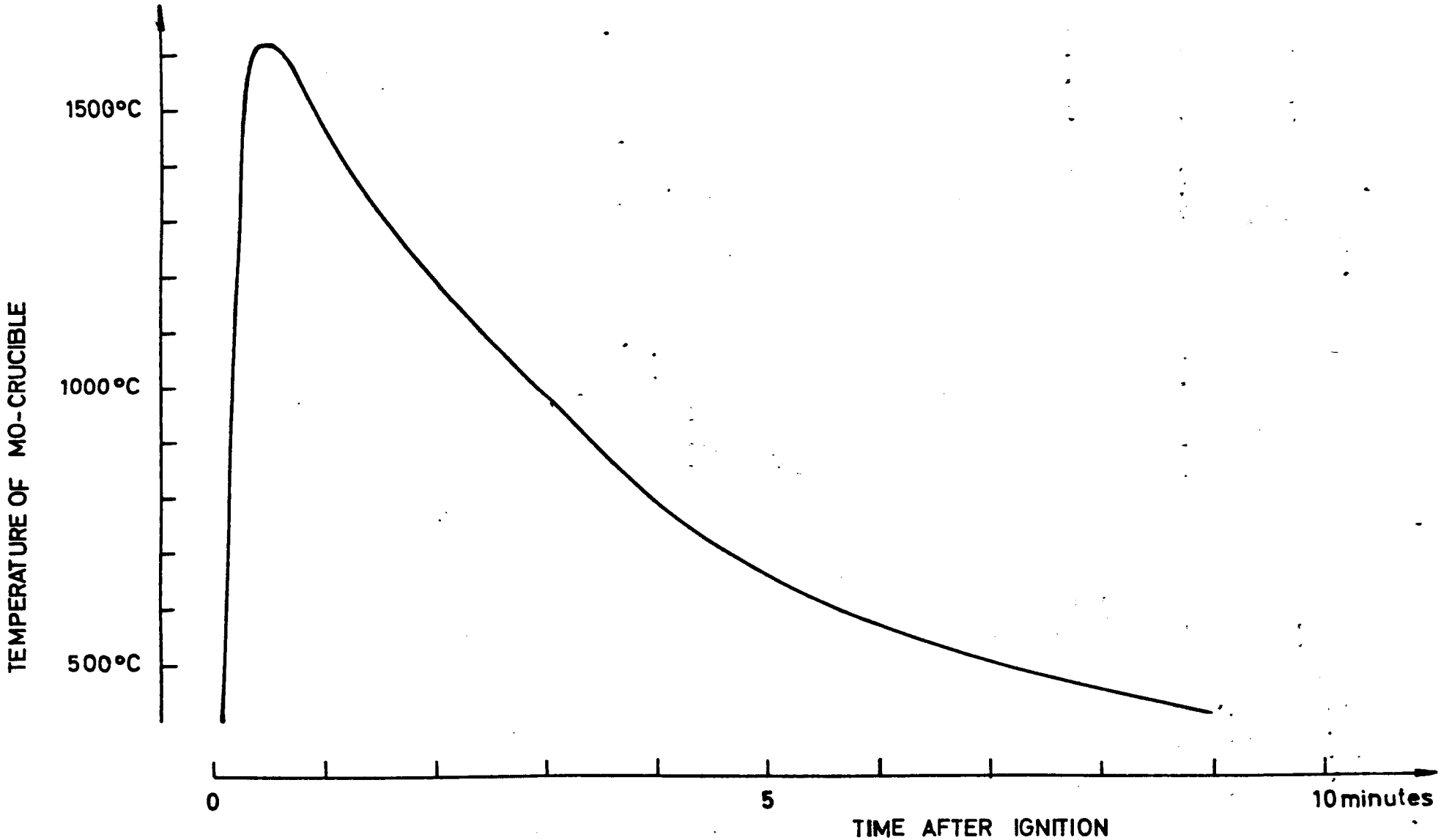

Figure 2 Temperature of the sample during extraction.

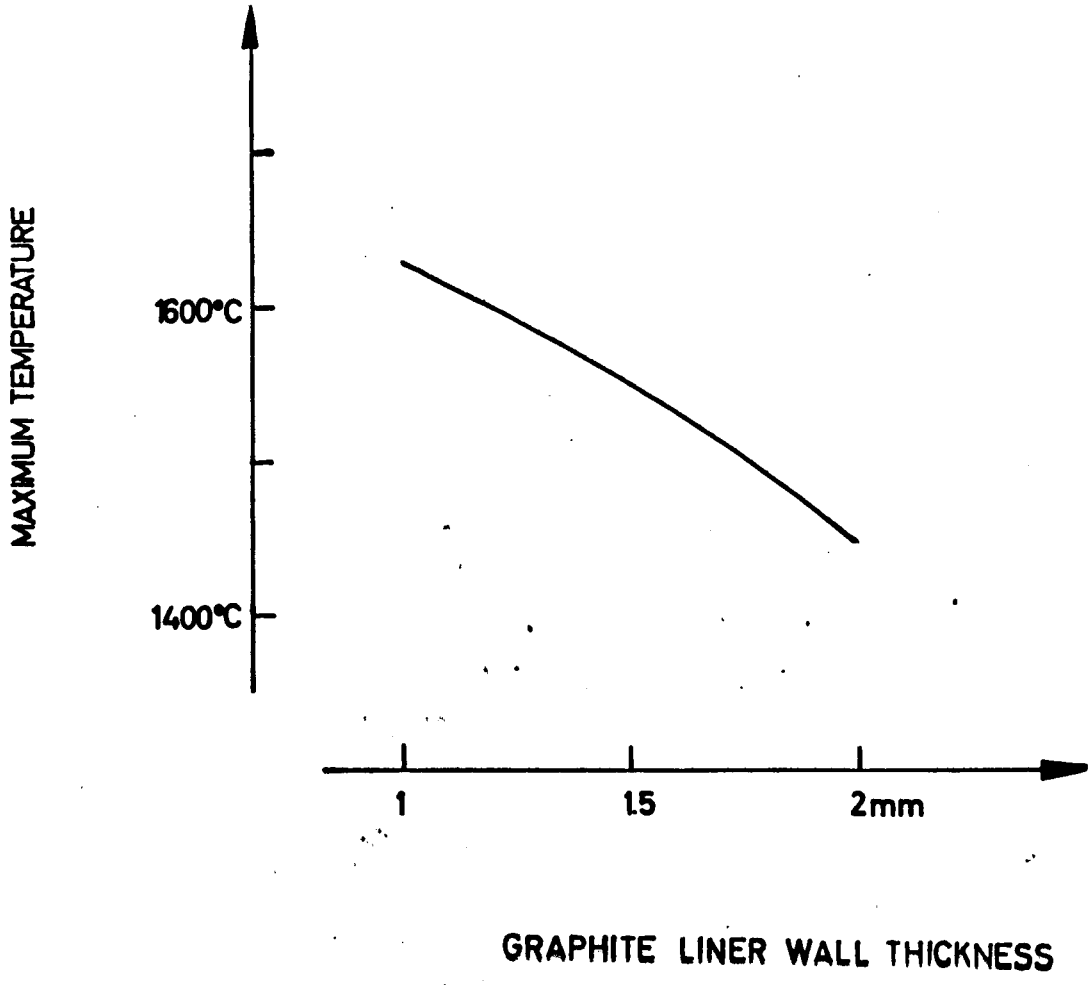

Figure 3 Dependence of the maximum temperature reached by the sample on the wall thickness of the graphite liner.

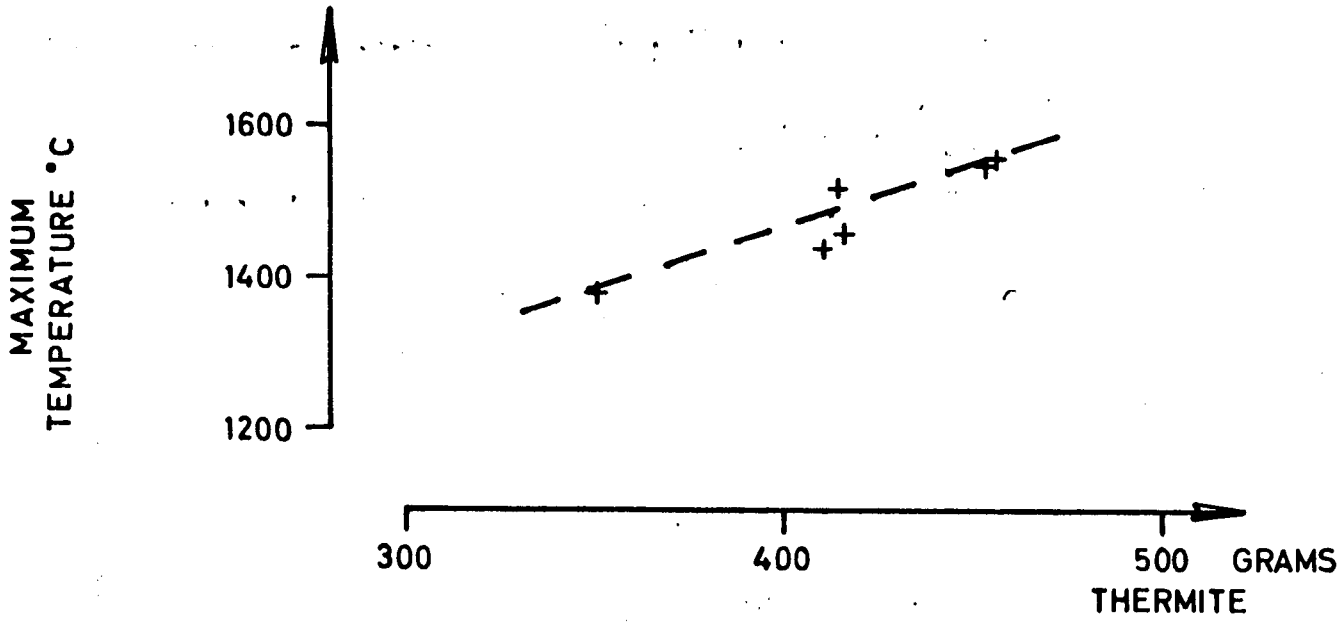

Figure 4 Maximum temperature reached by the sample in a four-sample extraction system (Type 64-1) for different amounts of thermite mixture.

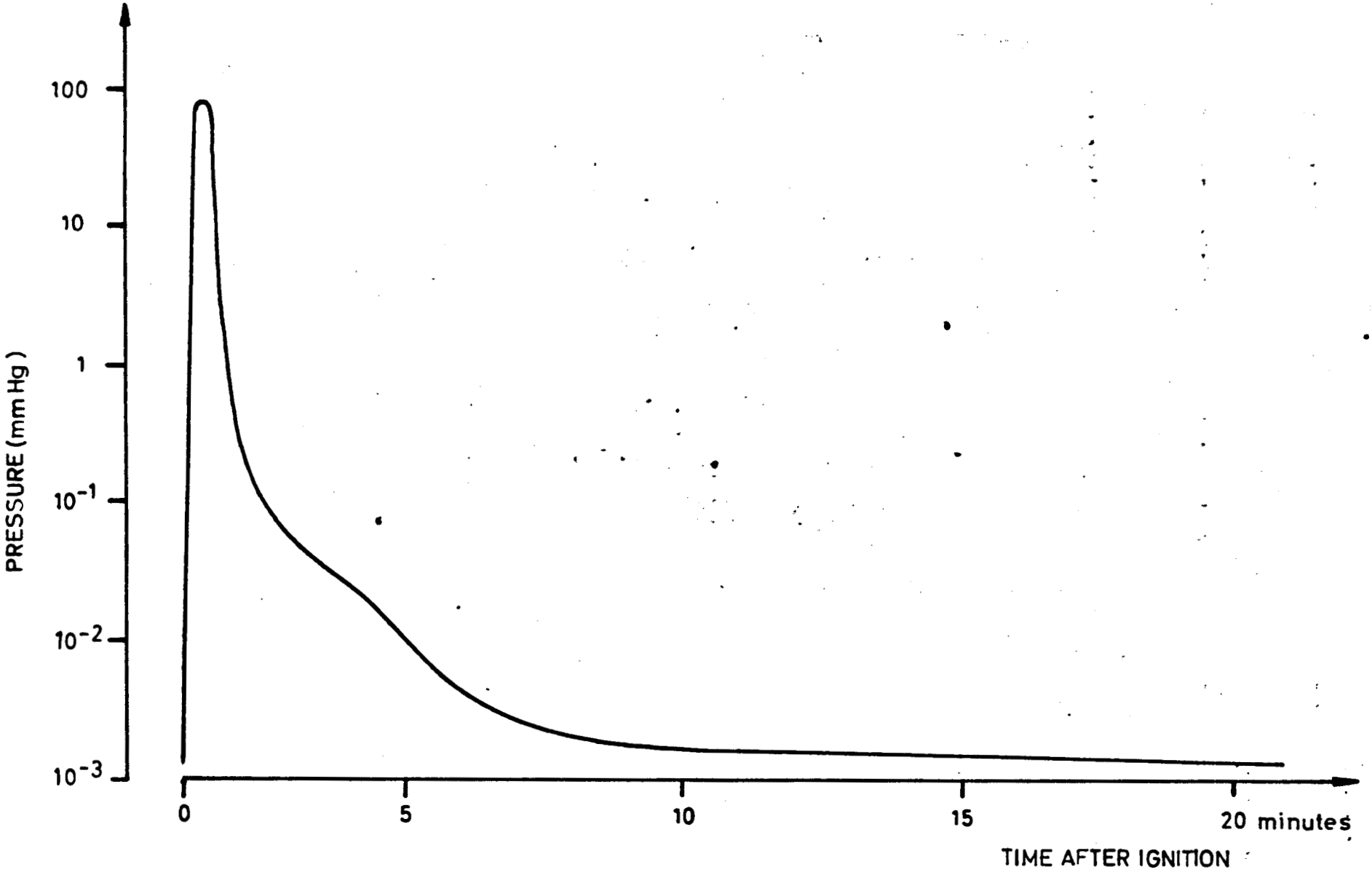

Figure 5 Total gas pressure during an extraction of a stone meteorite sample. The pressure is measured with a pirani type gauge and the equivalent air pressure is given. The pressure drop by 5 orders of magnitude is due to the gettering action of the internal Ti-sponge.

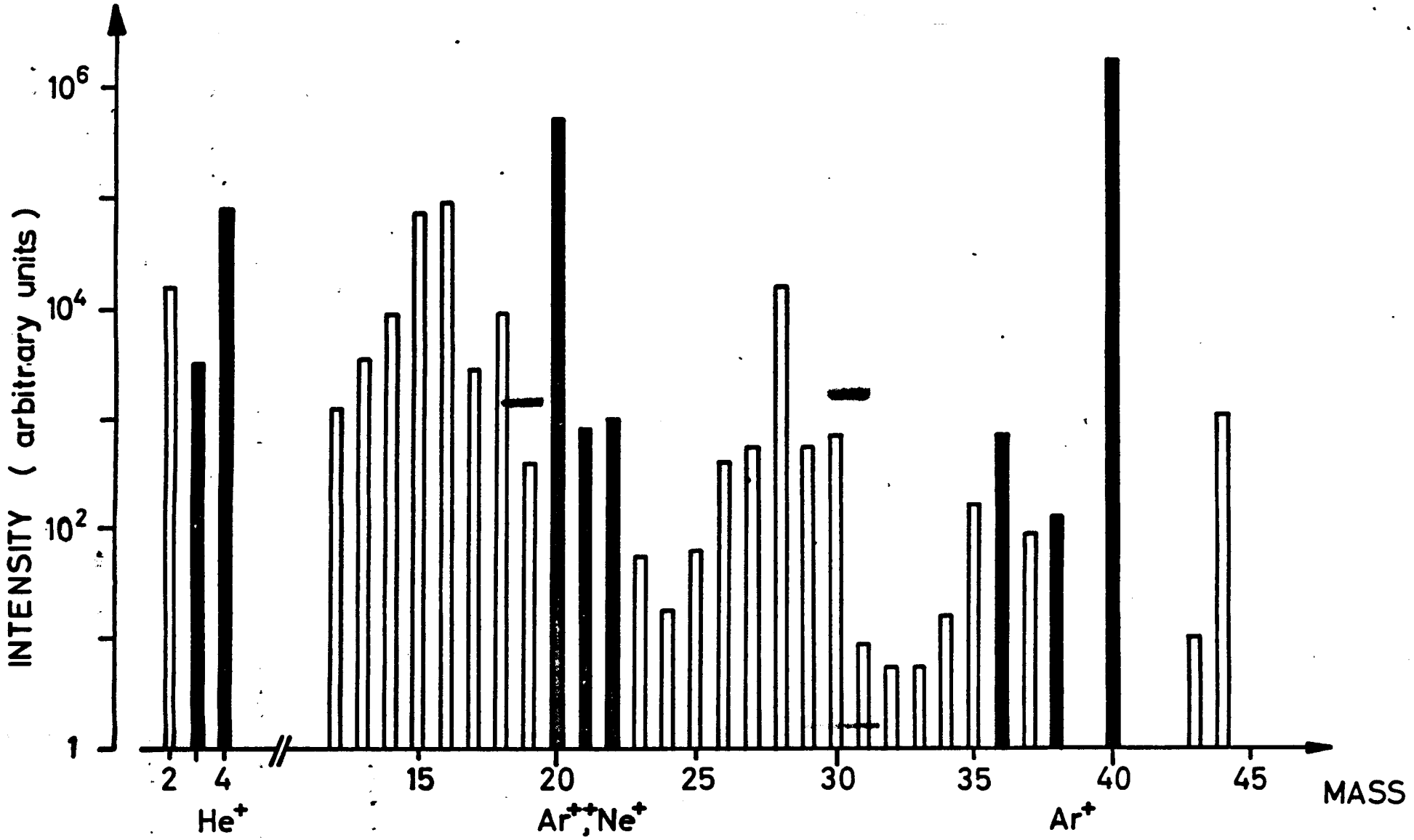

Figure 6 Mass spectra of gases extracted with four-sample thermite extraction system from 2 g of stone meteorite **Finney**. The peaks corresponding to rare gas isotopes are shaded. No additional purification was applied. Approximate amounts: He^4: 1.4×10^{-5} cc STP; Ne^{21}: 8×10^{-8} cc STP; Ar^{40}: 8×10^{-5} cc STP.