



**HAL**  
open science

**Measurement of cross sections for  $^{22}\text{Na}$ ,  $^{20-22}\text{Ne}$  and  $^{36-42}\text{Ar}$  in the spallation of Mg, Al, Si, Ca and Fe.  
Production ratios of some cosmogenic nuclides in  
meteorites**

F. Baros, S. Regnier

► **To cite this version:**

F. Baros, S. Regnier. Measurement of cross sections for  $^{22}\text{Na}$ ,  $^{20-22}\text{Ne}$  and  $^{36-42}\text{Ar}$  in the spallation of Mg, Al, Si, Ca and Fe. Production ratios of some cosmogenic nuclides in meteorites. *Journal de Physique*, 1984, 45 (5), pp.855-861. 10.1051/jphys:01984004505085500 . jpa-00209817

**HAL Id: jpa-00209817**

**<https://hal.science/jpa-00209817v1>**

Submitted on 4 Feb 2008

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Classification

Physics Abstracts

25.40 — 27.30 — 27.40 — 94.40

## Measurement of cross sections for $^{22}\text{Na}$ , $^{20-22}\text{Ne}$ and $^{36-42}\text{Ar}$ in the spallation of Mg, Al, Si, Ca and Fe. Production ratios of some cosmogenic nuclides in meteorites

F. Baros and S. Regnier

Centre d'Etudes Nucléaires de Bordeaux-Gradignan,  
Laboratoire de Chimie Nucléaire (\*), 33170 Gradignan, France

(Reçu le 14 octobre 1983, révisé le 18 janvier 1984, accepté le 26 janvier 1984)

**Résumé.** — Nous avons mesuré, par spectrométrie de masse des gaz rares et par comptage  $\gamma$ , les sections efficaces  $\sigma$  des réactions nucléaires conduisant à  $^{22}\text{Na}$ ,  $^{20-22}\text{Ne}$  et  $^{36-42}\text{Ar}$  dans des cibles de Mg, Al, Si, Ca et Fe bombardées par des protons de 0,059 à 24 GeV. Ces nouvelles mesures et celles existant dans la littérature nous ont permis d'évaluer les rapports des sections efficaces  $\sigma_R/\sigma_S$  en fonction de l'énergie de quatre paires d'isotopes, l'un stable  $S$  et l'autre radioactif  $R$  :  $^{22}\text{Na}/^{22}\text{Ne}$ ,  $^{26}\text{Al}/^{21}\text{Ne}$ ,  $^{39}\text{Ar}/^{38}\text{Ar}$  et  $^{36}\text{Cl}/^{36}\text{Ar}$ . Ces paires d'isotopes permettent de calculer l'âge d'exposition des météorites, si on connaît le rapport de leur taux de production  $P_R/P_S$ . Notre travail établit des limites aux rapports  $P_R/P_S$  dans les météorites de fer à partir de la seule considération des rapports  $\sigma_R/\sigma_S$ . Dans les météorites de pierre on peut seulement montrer, par cette approche, l'importance des effets de composition chimique et de blindage.

**Abstract.** — We measured the cross sections of  $^{22}\text{Na}$ ,  $^{20,21,22}\text{Ne}$  and  $^{36,38,39,42}\text{Ar}$  in nuclear reactions induced by the proton bombardment of Mg, Al, Si, Ca and Fe at several energies between 0.059 and 24 GeV using high-sensitivity mass-spectrometry for noble gases and  $\gamma$ -counting for  $^{22}\text{Na}$ . We then evaluated the cross section ratio versus energy for the  $^{22}\text{Na}/^{22}\text{Ne}$ ,  $^{26}\text{Al}/^{21}\text{Ne}$ ,  $^{39}\text{Ar}/^{38}\text{Ar}$  and  $^{36}\text{Cl}/^{36}\text{Ar}$  radioactive-stable pairs of isotopes, which are useful for measuring the exposure age of meteorites. From the results alone, we can set some limits to the corresponding production ratios in iron meteorites, but not in stony objects because a knowledge of chemical abundances of Na, Mg, Al, Si, S, K, Ca, Fe, and shielding conditions are required.

### 1. Introduction.

Cosmic-ray-produced nuclides provide sensitive tools for studying exposure history of extra-terrestrial materials such as meteorites and lunar rocks. These nuclides are produced through spallation reactions induced by primary and secondary cosmic-ray particles in the near surface (up to a few meters) of bodies in the solar system that are not radiation-shielded by an atmosphere or strong magnetic fields. A detailed study of the exposure history of a meteoritic or lunar sample should comprise several chapters dealing with : the duration of exposure (or exposure age  $T$ ); the flux of galactic and solar cosmic rays and its variations in time and space; the shielding history of the sample, involving possibly multiple breakups; the space erosion by micrometeorites. The measurement of as

many cosmogenic nuclides as possible is required to decipher the relative importance of so many variables.

In the present paper we investigate a group of methods for measuring  $T$ , which involve a pair of one stable  $S$  and one radioactive  $R$  cosmogenic nuclides. The production rate  $P$  of any cosmogenic nuclide may be defined as the number of atoms produced per time unit through nuclear reactions. It depends on the intensity and energy spectrum of cosmic radiations and also on the chemical composition and the shielding depth of the sample. The growth curve for a radioactive nuclide is such as  $R = P_R(1 - e^{-\lambda T})$ . After several half-lives the activity  $R$  is in secular equilibrium and  $R = P_R$ . So a given isotope  $R$  integrates the cosmic ray flux over several half-lives. The concentration of a stable nuclide is given by  $S = S_0 + P_S T$  where  $S_0$  is the contribution of sources other than spallation reactions. In favorable cases (mainly isotopes of noble gases),  $S_0$  can be neglected or can be subtracted.  $P_S$  is generally unknown. Assuming cons-

(\*) E.R.A. n° 144 du C.N.R.S.

tancy for  $P_R$  and  $P_S$ , a simple expression for exposure age is thus :

$$T = \frac{P_R}{P_S} \frac{S}{R} \quad (1)$$

if secular equilibrium has been reached for the radioisotope and if the meteorite didn't have a complex history. Convenient  $R$ - $S$  pairs involve cosmogenic nuclides that are produced by similar nuclear reactions and will generally have very similar atomic masses. The key parameter in such a method is the production ratio  $P_R/P_S$ , because  $S$  and  $R$  are measured quantities in the sample. At first [1-5], many production ratios were calibrated against some adopted exposure age. This approach does not provide an independent method and ignores likely effects from target chemistry and shielding. Also, absolute and independent calibrations are necessary to compare ages from different methods and obtain additional information about the exposure history of a sample or about the temporal variations in the fluxes of cosmic-ray particles. However some production ratios have been tentatively inferred from cross section data [6-9]. Independent calibrations were used for the  $^{81}\text{Kr}$ - $^{83}\text{Kr}$  method [10] and for the  $^{40}\text{K}$ - $^{41}\text{K}$  method which applies only to iron meteorites [11]. We will focus here on four  $R$ - $S$  pairs :  $^{22}\text{Na}$ - $^{22}\text{Ne}$ ,  $^{26}\text{Al}$ - $^{21}\text{Ne}$ ,  $^{39}\text{Ar}$ - $^{38}\text{Ar}$  and  $^{36}\text{Cl}$ - $^{36}\text{Ar}$  and determine the limits of their production ratios  $P_R/P_S$  starting from cross sections ratios  $\sigma_R/\sigma_S$ .

We measured new cross sections for  $^{22}\text{Na}$ ,  $^{20-22}\text{Ne}$  and  $^{36-42}\text{Ar}$  in Mg, Al, Si, Ca and Fe targets bombarded with 59 MeV to 24 GeV protons. From these new measurements and from literature data, we deduced excitation functions  $\sigma(E)$  for  $^{22}\text{Na}$ ,  $^{22}\text{Ne}$ ,  $^{26}\text{Al}$ ,  $^{21}\text{Ne}$ ,  $^{39}\text{Ar}$ ,  $^{38}\text{Ar}$ ,  $^{36}\text{Cl}$  and  $^{36}\text{Ar}$  in targets that are abundant in meteorites. Neutron-induced reactions were mostly estimated. We then examined the cross section ratios  $\sigma_R/\sigma_S$  and tried to deduce limits on the corresponding  $P_R/P_S$  ratios and their possible variations with chemistry and shielding.

## 2. Experimental.

Pure target foils of Mg (99.9 % or 3N), Al (5N) and Fe (5N), and also crystals of Si (4N) and  $\text{CaF}_2$  (4N) were stacked, 2 or 3 at a time, with monitor and guard Al (5N) foils. Exact thickness was determined by weighing and was (20 to 50)  $10^{-3}$  mm for foils, and 0.6 to 1 mm for crystals. The targets were irradiated between 1969 and 1980 in the accelerators of Louvain-La-Neuve (0.059 and 0.075 GeV), Orsay (0.150, 0.168 and 0.200 GeV), CERN-SC (0.600 GeV), Saturne (1.0, 1.05 and 2.5 GeV) and CERN-PS (24 GeV). Both internal and external proton beams were used at 1 and 24 GeV, internal beam only at 0.150 and 0.600 GeV, and external beam only at 0.059, 0.075, 0.168, 0.200 and 2.5 GeV. Special care was taken to minimize secondary particle and recoil effects.

Proton fluences were measured according to the  $^{27}\text{Al}(p, X)^{22}\text{Na}$  monitor reaction whose cross sections are 28.1, 23.6, 17, 16.7, 16.1, 15.3, 11.7 and 10.0 mb, respectively at 0.059, 0.075, 0.150, 0.168, 0.200, 1, 2.5 and 24 GeV [12]. Between  $2 \times 10^{15}$  and  $4 \times 10^{17}$  protons struck the targets. By comparing results from either internal and external irradiation, or young and old targets, no diffusion loss for gaseous products neon and argon was observed. More details about targets and irradiations are given in references [13] and [14].

Some Mg, Si and Ca targets were counted for the 1.27-MeV  $\gamma$  peak of  $^{22}\text{Na}$  in a 4-keV resolution Ge(Li) gamma ray spectrometer, calibrated with  $^{22}\text{Na}$  standards.

Noble gases Ne and Ar were analysed by means of a Micromass 60° sector, 12 cm radius mass-spectrometer equipped with a gas extraction and purification line, two activated charcoals and two calibration systems. After at least two years following the irradiation (2 to 12 years) targets and blank foils were stored, up to 10 at a time, in the extraction-purification line in which an ultrahigh vacuum (around  $10^{-9}$  torr) was progressively attained by pumping and baking. Targets were baked two days between 60 and 80 °C. We checked that this procedure did not outgas any significant amount of reaction-produced neon or argon. Each foil was then melted inside a molybdenum crucible heated under vacuum by an electron-bombardment oven, between 700 and 1 600 °C depending on the metal or crystal. Attempts to separate trapped gases of atmospheric isotopic composition and nuclear reaction-produced gases by step-wise heating were unsuccessful. The noble gases were purified in several steps, using Ti, CuO-Pd and Al-Zr getters between 200 and 800° for eliminating chemically active species. Argon was adsorbed on a first activated charcoal trap at  $-196$  °C. About half of the neon was introduced into the mass spectrometer. During isotopic analysis, most memory argon was adsorbed on a second charcoal trap at  $-196$  °C. Interferences at  $m/e = 20$  from  $\text{H}_2^{18}\text{O}^+$  or  $^{40}\text{Ar}^{2+}$  ions, and at  $m/e = 22$  from  $\text{CO}_2^+$  ions were monitored by measuring also  $m/e = 18$ , 40 and 44 peaks. No interference was detected at  $m/e = 21$ . In most cases, the second half of neon was used for isotopic dilution and was measured independently after mixing with a known amount of atmospheric neon. In Ca and Fe targets, argon was also analysed. Almost all  $^{40}\text{Ar}$  was atmospheric, except a small part (always less than 1 %) deduced in an iterative procedure from the charge-dispersion curves of products. Corresponding corrections were applied to  $^{36}\text{Ar}$  and  $^{38}\text{Ar}$ . Background peaks due to hydrocarbon ions had to be corrected at  $m/e = 39$  and  $m/e = 42$ . In this work, flat top peaks were obtained for a resolution of 250 (1 % valley). Calibration was made by frequently introducing air standards from a pipette systems or from an independent system composed of several known volumes and measured pressure using a capacitance manometer. The isotope

ratios were corrected for memory or pumping effects, isotopic discrimination, blank, hydrocarbons (for  $^{38,39,42}\text{Ar}$ ) and interfering ions. Complete outgassing of melted targets was checked in reextraction runs, and a small correction (0 to 1 % in most cases) was applied. Depending on cross sections, target thicknesses and fraction of gases that was analysed,  $10^7$  to  $10^{11}$  atoms of neon 20, 21, 22 or argon 36, 38, 39, 40 and 42 were measured.

**3. Results.**

The cross sections measured for  $^{22}\text{Na}$  in the targets Mg, Si and Ca, of  $^{20-22}\text{Ne}$  in Mg, Al, Si, Ca and Fe, and of  $^{36-42}\text{Ar}$  in Ca are reported in tables I, II and III.

The results are averages of  $N$  independent measurements weighed by individual uncertainties. The root mean square errors included the various error sources for each measurement : target preparation and impurities ; monitoring and alignment ; secondary particles ; blanks, interfering ions or hydrocarbons (the associated error was 50 % of the correction); zero time corrections to mass-spectrometer analysis; re-extractions; calibration. Recoil corrections for spallation (0 to 3 % in most cases) were calculated according to Sauvageon [15]. Correction for radioactive decay was made for  $^{39}\text{Ar}$  and  $^{42}\text{Ar}$ . Also  $^{22}\text{Ne}$  was corrected for decay of  $^{22}\text{Na}$  in table II. Many errors cancel out for the isotope ratios.

**3.1  $^{22}\text{Na}$ .** — Our measurements in Mg (Table I) agree within errors with those of Raisbeck and Yiou, 1975 [16]. Additional cross sections at other energies can be found in Korteling and Caretto, 1970 [17] and the compilation of de Lassus and Tobailem, 1972 [18]. Excitation function  $\sigma(E)$  in Si was already well documented from Raisbeck and Yiou [16] and Korteling and Caretto [17], and also from the compilation of Tobailem, 1981 [19]. In Ca, there was only one

Table I. — Cross sections  $\sigma$  (in mb) for  $^{22}\text{Na}$  in Mg, Si and Ca, as functions of the incident energy  $E_p$  (in GeV).  $N$  is the number of independent measurements.

Target	$E_p$ (GeV)	N	$\sigma$ (mb)
Mg	1.0	1	$33.1 \pm 3.3$
	2.5	1	$22.2 \pm 2.2$
	24.0	1	$21.7 \pm 2.2$
Si	0.150	3	$18.5 \pm 1.9$
	24.0	2	$15.2 \pm 1.5$
Ca	0.168	1	$0.44 \pm 0.04$
	1.0	1	$6.7 \pm 0.7$
	2.5	1	$7.1 \pm 0.7$

measurement at 400 MeV from Korteling and Caretto, 1967 [20].

**3.2  $^{20,21,22}\text{Ne}$ .** — Excitation functions for stable Ne isotopes were quite well documented in Mg, Al and Si targets in the low energy part  $E_p < 70$  MeV (see Ref. [21, 22]), and our results notably extend our knowledge to higher energies. No neon cross section in Ca was measured until now. Our measurements (Table II) disagree in a systematic way with those of Goebel *et al.*, 1964 [8] as is evident in figure 1 where we plotted excitation functions for  $^{20-22}\text{Ne}$  in Fe. However we agree with Bieri and Rutsch, 1962 [23].

**3.3  $^{36-42}\text{Ar}$ .** — Ar measurements in Ca are reported (Table III) at 0.168, 1.0 and 2.5 GeV.  $^{39}\text{Ar}$  cross sections at 1.0 and 2.5 GeV are mostly upper limits, due to strong hydrocarbon interference at  $m/e = 39$ . Previous results were available at 600 MeV (Stoener *et al.*, 1970 [24]) and 12 to 45 MeV (Walton *et al.*, 1976 [25]). We also measured argon isotopes in iron targets, and our results were in agreement with those of Regnier, 1979 [14]. This agreement shows that there is no significant loss of spallation neon or argon in « old » iron targets.

Table II. — Cross sections  $\sigma$  (in mb) for neon isotopes in Mg, Al, Si, Ca and Fe, as functions of the incident energy  $E_p$  (in GeV). Values for  $^{22}\text{Ne}$  are either corrected for  $^{22}\text{Na}$ , or cumulative ( $^{22}\text{Ne} + ^{22}\text{Na}$ ). The  $^{22}\text{Na}$  cross section adopted for correction took into account all published values and not only that of table I. The cumulative cross sections in the last column are more reliable because they imply much less correction to the measured  $^{22}\text{Ne}$ .  $N$  is the number of independent measurements.

Target	$E_p$ (GeV)	N	$^{20}\text{Ne}$ $\sigma$ (mb)	$^{21}\text{Ne}$ $\sigma$ (mb)	$^{22}\text{Ne}$ $\sigma$ (mb)	$^{22}\text{Ne} + ^{22}\text{Na}$ $\sigma$ (mb)
Mg	0.059	2	$43.3 \pm 5.2$	$40.9 \pm 4.9$	$15.2 \pm 2.0$	$77.2 \pm 10.2$
	0.075	2	$45.2 \pm 5.3$	$43.5 \pm 5.1$	$18.3 \pm 2.4$	$69.3 \pm 9.1$
	0.168	1	$25.4 \pm 3.3$	$27.1 \pm 3.6$	$10.6 \pm 1.6$	$41.6 \pm 6.3$
	0.200	2	$28.2 \pm 3.4$	$30.0 \pm 3.7$	$12.6 \pm 1.7$	$42.6 \pm 5.7$
	1.0	1	$35.8 \pm 5.9$	$33.6 \pm 4.9$	$22.2 \pm 3.7$	$51.8 \pm 8.0$
	2.5	1	$14.6 \pm 4.3$	$18.4 \pm 2.8$	$8.2 \pm 1.4$	$31.7 \pm 5.4$
	24.0	1	$20.6 \pm 2.7$	$20.9 \pm 2.8$	$9.0 \pm 1.3$	$30.7 \pm 4.4$
Al	0.059	2	$14.3 \pm 1.8$	$32.1 \pm 4.1$	$9.9 \pm 1.5$	$38.0 \pm 5.8$
	0.075	2	$26.1 \pm 3.1$	$32.9 \pm 3.9$	$11.5 \pm 1.5$	$35.1 \pm 4.6$
	0.150	2	$22.6 \pm 2.5$	$24.3 \pm 2.7$	$10.4 \pm 1.2$	$27.4 \pm 3.2$
	0.168	2	$22.0 \pm 2.5$	$23.6 \pm 2.7$	$10.1 \pm 1.2$	$26.8 \pm 3.2$
	0.200	2	$26.3 \pm 2.9$	$27.7 \pm 3.1$	$12.9 \pm 1.5$	$29.0 \pm 3.4$
	1.0	3	$22.2 \pm 2.8$	$22.4 \pm 2.8$	$11.3 \pm 1.6$	$26.6 \pm 3.8$
	1.05	2	$28.5 \pm 3.4$	$28.4 \pm 3.4$	$16.5 \pm 2.0$	$31.8 \pm 3.9$
	2.5	3	$21.2 \pm 2.4$	$21.9 \pm 2.5$	$12.0 \pm 1.3$	$23.7 \pm 2.6$
	24.0	2	$16.1 \pm 2.2$	$18.8 \pm 2.4$	$10.4 \pm 1.7$	$20.4 \pm 3.3$
Si	0.150	3	$17.0 \pm 1.8$	$15.8 \pm 1.7$	$4.1 \pm 0.5$	$21.3 \pm 2.6$
	24.0	2	$17.8 \pm 2.0$	$15.9 \pm 1.8$	$4.9 \pm 0.6$	$18.6 \pm 2.3$
Ca	0.168	1	$0.83 \pm 0.09$	$0.53 \pm 0.06$	$0.21 \pm 0.04$	$0.65 \pm 0.12$
	1.0	1	$11.9 \pm 1.4$	$11.3 \pm 1.3$	$5.3 \pm 0.9$	$12.0 \pm 2.0$
	2.5	1	$12.4 \pm 1.4$	$11.4 \pm 1.3$	$5.1 \pm 0.8$	$12.2 \pm 1.9$
Fe	0.600	2	$1.04 \pm 0.20$	$1.04 \pm 0.20$	$0.80 \pm 0.15$	$1.07 \pm 0.20$
	2.5	1	$6.5 \pm 0.8$	$6.5 \pm 0.8$	$4.3 \pm 0.6$	$6.5 \pm 0.9$
	24.0	2	$8.0 \pm 1.2$	$7.8 \pm 0.9$	$5.6 \pm 0.9$	$7.9 \pm 1.3$

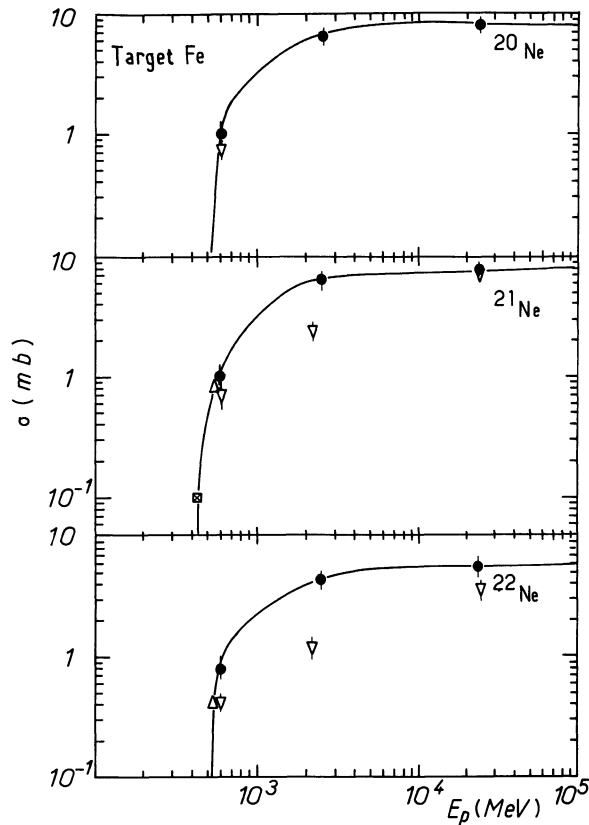


Fig. 1. — Measured cross sections  $\sigma$  in mb for the production of  $^{20}\text{Ne}$ ,  $^{21}\text{Ne}$  and  $^{22}\text{Ne}$  in iron bombarded with protons of energy  $E$  in MeV. Filled symbols : this work ; others :  $\nabla$  ref. [8],  $\Delta$  ref. [23],  $\boxtimes$  ref. [40]. Solid lines are adopted excitation functions (see text).

Table III. — Cross sections  $\sigma$  (in mb) for  $^{36-42}\text{Ar}$  in Ca, versus energy  $E_p$  (in GeV). Large error bars for  $^{39}\text{Ar}$  are due to hydrocarbon interference at 1.0 and 2.5 GeV.

$E_p$ (GeV)	N	$\sigma_{36}$ (mb)	$\sigma_{38}$ (mb)	$\sigma_{39}$ (mb)	$\sigma_{42}$ (mb)
0.168	1	$39.9 \pm 4.1$	$82.6 \pm 8.3$	$1.12 \pm 0.12$	$0.032 \pm 0.008$
1.0	1	$29.0 \pm 3.0$	$58.4 \pm 5.9$	$1.1 \pm 0.9$	
2.5	1	$20.6 \pm 2.1$	$42.1 \pm 4.2$	$0.9 \pm 0.7$	

#### 4. Discussion.

From our results and others found in literature, we examined the  $\sigma_R/\sigma_S$  cross section ratios, versus incident energy, of some radioactive-stable pairs of isotopes that are in common use for calculating the exposure age  $T$  of meteorites. Our general approach was to obtain excitation functions for all appropriate nuclear reactions, either from experimental cross sections or from systematics. We largely used the Silberberg and Tsao [26] formula for spallation of Mg to Fe targets at more than 100 MeV. When possible we calibrated the formula using experimental cross sections. In the lower energy part we used nuclear

systematics from analog reactions in neighbouring targets, applying appropriate corrections for different thresholds. We also used the Keller *et al.* [27] method as well as  $N/Z$  ratio corrections. Secondary neutrons are very important in meteorites, especially when size and geometry conditions favour their growth. At energies lower than 100 MeV, they are responsible for most inelastic collisions. More details are discussed in Baros [13]. From the excitation functions, we computed the cross section ratios versus energy curves.

4.1  $^{22}\text{Na}/^{22}\text{Ne}$ . — In figure 2 we plotted the cross section ratio  $\sigma(^{22}\text{Na})/\sigma(^{22}\text{Ne})$  versus energy for incident protons (solid lines) or neutrons (broken lines) in several target elements. In stony meteorites such as chondrites (about 90 % of observed falls), the main target elements for both  $^{22}\text{Na}$  and  $^{22}\text{Ne}$  are Mg and Si; then there is some contribution from Al, S, Ca, Fe and other elements. Iron meteorites are composed of Fe-Ni alloys with microscopic to centimeter-sized inclusions. In chondrites there is a clear difference between the cross section ratio inferred from : i) proton-induced reactions, mostly from measured values, giving  $\sigma(^{22}\text{Na})/\sigma(^{22}\text{Ne})$  ratios between 0.5 and 0.8, ii) neutron induced reactions in Mg, corresponding to  $\Delta A \lesssim 4$ . Neutron channels are mostly evaluated from systematics. The  $\text{Mg}(n, x)^{22}\text{Ne}$  excitation function was taken from Reedy *et al.*, 1979 [28], for energies below 20 MeV. The cross-section ratio  $\sigma(^{22}\text{Na})/\sigma(^{22}\text{Ne})$  was evaluated by Nishiizumi *et al.*, 1980 [7] to be between 0.50 and 0.65. We agree with their upper limit but the present results tend to slightly decrease the lower limit in cases where low energy neutrons are important. The situation is simpler in iron meteorites where mostly particles with energy higher than 500 MeV are effective, specifically for samples reasonably free of sulfur or phosphorus-rich inclusions. From cross sections only, the production ratio  $P(^{22}\text{Na})/P(^{22}\text{Ne})$  is  $0.27 \pm 0.03$  in iron meteorites.

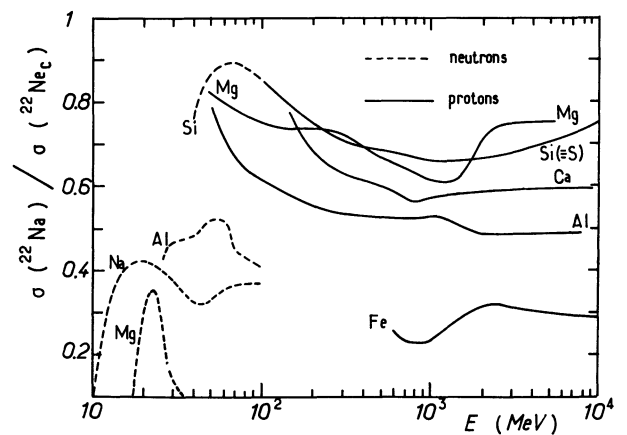


Fig. 2. — Cross section ratio  $\sigma(^{22}\text{Na})/(\sigma(^{22}\text{Na}) + \sigma(^{22}\text{Ne}))$  versus incident energy  $E$  for protons (solid lines) and neutrons (dashed lines) in several targets. Low energy neutrons favour  $^{22}\text{Ne}$  over  $^{22}\text{Na}$  in the very important magnesium target.

4.2  $^{26}\text{Al}/^{21}\text{Ne}$ . — *A priori* this pair of radioactive-stable isotopes is not a good candidate for calculating exposure ages, because  $^{26}\text{Al}$  and  $^{21}\text{Ne}$  have quite different atomic masses. Thus they are expected to arise from different reactions in targets such as Al or Si, and of course in Mg where  $^{26}\text{Al}$  is only produced through the  $^{26}\text{Mg}(p, n)$  channel, while  $^{21}\text{Ne}$  is supplied by the important  $^{24}\text{Mg}(n, \alpha)$  reaction among others. Also  $^{24}\text{Mg}$  is the most abundant isotope of Mg. Neutrons are not able to produce  $^{26}\text{Al}$  from Mg, except for very rare reactions like  $^{26}\text{Mg}(n, n\pi^-)^{26}\text{Al}$ . However there exists a great deal of both  $^{21}\text{Ne}$  and  $^{26}\text{Al}$  data in meteorites and we investigate in what extent the  $^{26}\text{Al}/^{21}\text{Ne}$  method is reliable in considerations about nuclear reactions. In figure 3, we show the cross section ratio  $\sigma(^{26}\text{Al})/\sigma(^{21}\text{Ne})$  versus energy in Mg to Fe targets. The ordinate covers more than 6 orders of magnitude although the neutron channels in Mg are not reported (they give near 0 values).  $^{26}\text{Al}$  excitation functions were largely deduced from the Saclay compilations [12, 18, 19] and some unpublished results from this laboratory [29]. The  $^{24}\text{Mg}(n, \alpha)^{21}\text{Ne}$  excitation function deduced by Reedy *et al.*, 1979 [28], from their measurements at 14.1 and 14.7 MeV was adopted, but the shape of the peak is quite uncertain. The  $^{27}\text{Al}(n, 2n)^{26}\text{Al}$  channel was estimated at low energy from the compilation of Stehn *et al.*, 1964 [30] about the  $^{26}\text{Al}^m$  data. According to Norman, 1979 [31], the relative yields of  $^{26}\text{Al}^s$  and  $^{26}\text{Al}^m$  deduced from the  $^{25}\text{Mg}(p, \gamma)$  reaction in the 0.3-1.72 MeV energy range are in the ratio  $Y(m)/Y(g) \simeq 1/3$ . However, Reedy [32], examining the first 52 levels of  $^{26}\text{Al}$ , found that the ratio of levels decaying to the ground state is about the same as those going to the meta state when the excitation energies are less than 6 MeV, so that the  $Y(m)/Y(g)$  ratio would be near unity. The last point of view was adopted in figure 3 because it applies to  $(n, 2n)$  reactions specifically.

When stony meteorites are considered, Al and Si target elements (high ratio) compete with Mg (low ratio) whatever the energy range. A knowledge of the balance between chemical and energy effects requires the use of models of interaction of cosmic rays and meteorites, but that is not the purpose of the present paper.

In iron meteorites there are two important sources :

i) Fe and Ni targets give ratios between 0.3 and 0.4, and only more than 500 MeV particles are effective.

ii) S and P targets are only present at the 0.2-0.5 % concentrations (except in big inclusions) but they yield a cross section ratio 1 to 3. Particles with energies as low as 100 MeV are effective and they favour  $^{26}\text{Al}$  over  $^{21}\text{Ne}$ . In iron meteorites, we thus expect a cross section ratio  $^{26}\text{Al}/^{21}\text{Ne}$  that depends on both chemical abundances of phosphorus and sulfur and shielding conditions (i.e. the number of 100 to 500 MeV secondary particles). However, high abundances of phosphorus or sulfur are exceptional [33] and from simple assumptions a cross section ratio  $\sigma(^{26}\text{Al})/\sigma(^{21}\text{Ne}) =$

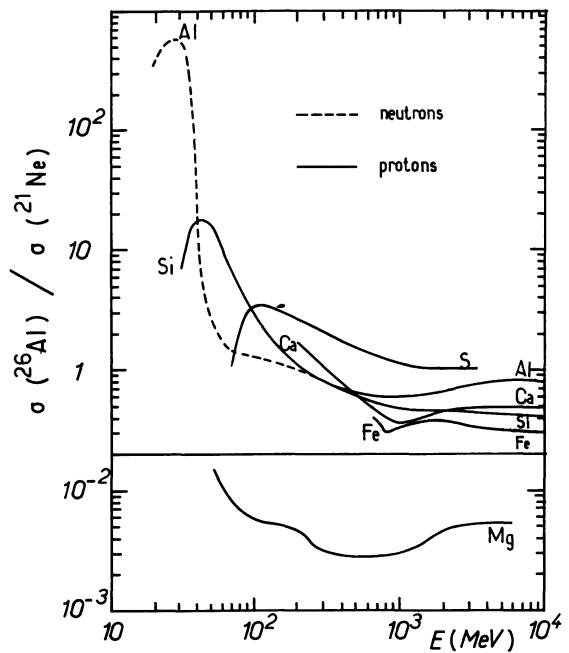


Fig. 3. — Cross section ratio  $\sigma(^{26}\text{Al})/\sigma(^{21}\text{Ne})$  versus incident energy  $E$  for protons (solid lines) and neutrons (dashed lines) in several targets,

$0.37 \pm 0.05$  was deduced, in good agreement with the 0.38 value of Hampel and Schaeffer [6].

4.3  $^{39}\text{Ar}/^{38}\text{Ar}$ . — The most important targets for regulating the  $\sigma(^{39}\text{Ar})/\sigma(^{38}\text{Ar})$  cross section ratio are K, Ca and Fe. The ratio is represented in figure 4 versus incident energy. Note the very different behaviour of neutron- and proton-induced reactions in Ca or K targets. The importance of the  $^{39}\text{K}(n, p)^{39}\text{Ar}$  reaction channel was first recognized in chondrites

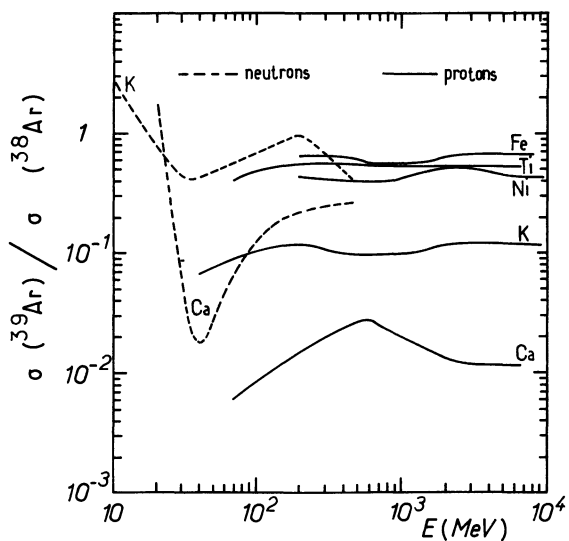


Fig. 4. — Cross section ratio  $\sigma(^{39}\text{Ar})/\sigma(^{38}\text{Ar})$  versus incident energy  $E$  for protons and neutrons in several targets. The neutron induced reactions are mostly evaluated and should be checked by some measurements.

by Begemann *et al.*, 1967 [34]. It has a zero threshold and its cross section is 15 to 400 mb between 1 and 20 MeV. However its excitation function is not very well known between 4 and 12 MeV, that is the region of the maximum. The situation in chondrites is quite complicated since the cross section ratio depends on both the nature of the target (K, Ca or Fe) and the energy range of interest. In iron meteorites, disregarding Ca- or K-rich inclusions, only Fe and Ni produce significant amounts of Ar. The  $^{39}\text{Ar}/^{38}\text{Ar}$  ratio is well known [14] in both targets and is fixed between 0.42 and 0.58 at all energies. From excitation functions only, a cross section ratio of  $0.50 \pm 0.05$  is deduced in iron meteorites, in agreement with the 0.52 value in use for years in literature [9].

4.4  $^{36}\text{Cl}/^{36}\text{Ar}$ . — This pair of isotopes is not very well suited in chondrites because the cross-section ratio  $^{36}\text{Cl}/^{36}\text{Ar}$  surely depends on both chemistry and shielding conditions through the  $^{39}\text{K}(n, \alpha)^{36}\text{Cl}$ ,  $^{40}\text{K}(n, p\alpha)^{36}\text{Cl}$  reactions, and the  $^{35}\text{Cl}(n, \gamma)^{36}\text{Cl}$  reaction in big meteorites. Also it is very difficult to correct for trapped  $^{36}\text{Ar}$  in chondrites. This method is well suited for iron meteorites and Schaeffer and Heymann, 1965 [9] proposed a cross section ratio of 0.82. From new measurements in Fe and Ni [14, 29, 35, 36] the cross section ratio  $\sigma(^{36}\text{Cl})/\sigma(^{36}\text{Ar})$  is fixed between 0.78 and 0.81 at all energies in iron meteorites.

## 5. Conclusions.

We measured new cross sections for  $^{22}\text{Na}$ ,  $^{20-22}\text{Ne}$  and  $^{36-42}\text{Ar}$  in Mg, Al, Si, Ca and Fe. Incident proton energies were 0.059 to 24 GeV. From our measurements, others from this laboratory, and a survey of literature we built excitation functions for  $^{22}\text{Na}$ ,  $^{22}\text{Ne}$ ,  $^{26}\text{Al}$ ,  $^{21}\text{Ne}$ ,  $^{39}\text{Ar}$ ,  $^{38}\text{Ar}$ ,  $^{36}\text{Cl}$  and  $^{36}\text{Ar}$  in targets that produce significant amounts of these isotopes through cosmic ray induced nuclear reactions. We then deduced the  $\sigma_R/\sigma_S$  cross section ratios *versus* energy for the radioactive-stable pairs of isotopes  $^{22}\text{Na}$ - $^{22}\text{Ne}$ ,  $^{26}\text{Al}$ - $^{21}\text{Ne}$ ,  $^{39}\text{Ar}$ - $^{38}\text{Ar}$  and  $^{36}\text{Cl}$ - $^{36}\text{Ar}$ .

With respect to nuclear reactions only, the corresponding production ratios  $P_R/P_S$  commonly used to calculate the exposure age of meteorites are expected to depend on both the concentrations of the main target elements and the shielding conditions (thus, the distribution of interacting protons and neutrons *versus* energy) in a particular sample. However the cases of stony and iron meteorites are quite different.

Iron meteorites are chemically well differentiated. They are mostly composed of Fe-Ni alloys with some inclusions. Isotopes with  $A = 21$  to 39 are all high-energy products except in some specific elements where they could be produced with high yields in inclusions at energies lower than 300 to 500 MeV, such as  $^{26}\text{Al}$  in phosphorus and sulfur. Keeping this in mind, the limits for iron meteorites deduced from this work are  $0.24 \leq P(^{22}\text{Na})/P(^{22}\text{Ne}) \leq 0.30$ ;

$0.32 \leq P(^{26}\text{Al})/P(^{21}\text{Ne}) \leq 0.42$ ;  $0.45 \leq P(^{39}\text{Ar})/P(^{38}\text{Ar}) \leq 0.55$  and  $0.78 \leq P(^{36}\text{Cl})/P(^{36}\text{Ar}) \leq 0.81$ .

The main target elements for producing  $^{22}\text{Na}$ ,  $^{20-22}\text{Ne}$  and  $^{26}\text{Al}$  in stony meteorites are Mg and Si. Magnesium plays a prominent part in regulating the  $^{22}\text{Na}/^{22}\text{Ne}$  and  $^{26}\text{Al}/^{21}\text{Ne}$  production ratios, and the low energy neutron induced reactions in this element are still insufficiently known. From nuclear reactions only, no good limits can be deduced for the  $^{26}\text{Al}/^{21}\text{Ne}$  production ratio. It is unlikely that this ratio is constant in the different classes of chondrites and for varying shielding conditions. In fact, there is no way to find a single  $P(^{26}\text{Al})/P(^{21}\text{Ne})$  value that fits exposure ages obtained from others methods [37], in a wide selection of chondrites. We offer some reservations regarding the  $^{22}\text{Na}/^{22}\text{Ne}$  ratio, since the importance of neutron induced reactions on Mg has still to be clarified. Also,  $^{22}\text{Na}$  (half-life = 2.6 years) integrates the cosmic radiation over a very short time and appears to vary over a solar cycle as measurements by Evans *et al.*, 1982 [38] in meteorite falls have shown. The  $^{39}\text{Ar}/^{38}\text{Ar}$  cross section ratio appears also very sensitive to chemical abundances and energy.

More information about the above production ratios in meteorites could be obtained from models of interaction of cosmic rays with spherical or nearly spherical objects. We are presently working on such models, inspired by the Reedy-Arnold model of interaction between cosmic rays and the Moon surface [39]. Another useful result of the present work was to identify the reaction channels for which experimental information is most needed, such as the low-energy neutron-induced reactions in K and Mg, and also in Na, Si and Ca. Such measurements are planned in the near future, and will provide a better knowledge of neutron-induced reactions in meteorites.

## Acknowledgments.

We are indebted to G. N. Simonoff for advising and support, to B. Lavielle, J. C. Lorin, K. Marti and R. C. Reedy for stimulating discussions, and to H. Sauvageon for help with experiments. We thank the following accelerator teams : Louvain-la-Neuve, Orsay, Saturne and CERN for their cooperation. Valuable technical assistance was provided by F. Brout. The research was sponsored by the Centre National de la Recherche Scientifique (ERA 144 et ATP Planétologie).

Appendix. — *Production of  $^{36}\text{Cl}$  by high energy spallation.* — Due to a mistake in the calculation of the proton fluences, the  $^{36}\text{Cl}$  cross section reported from this laboratory [36] are incorrect. Corrected values, together with some new measurements [29], also from this laboratory, are given in table IV.

Table IV. — Measured production cross sections  $\sigma$  of  $^{36}\text{Cl}$  in Fe, Ti and Ca as functions of the incident energy  $E_p$ ,  $N$  is the number of independent measurements. The  $\text{CaF}_2$  target irradiated at 0.15 GeV showed possible evidence for overheating in the accelerator.

$E_p$ (GeV)	Target	N	$\sigma(^{36}\text{Cl})$ (mb)
0.15	Fe	1	$0.21 \pm 0.07$
	Ti	2	$3.1 \pm 0.6$
	Ca	1	$\geq 10.7$
1.05 24	Fe	3	$11.0 \pm 2.2$
	Fe	3	$7.1 \pm 1.9$
	Ti	3	$17.5 \pm 1.8$
	Ca	2	$12.8 \pm 2.5$

### References

- [1] SPANNAGEL, G., SONNTAG, C., in *Radioactive Dating and Methods of low level counting* (IAEA, Vienna) 1967, p. 231-238.
- [2] MARTI, K., SHEDLOVSKY, J. P., LINDSTROM, R. M., ARNOLD, J. R., BHANDARI, N. G., in *Meteorite Research*, Millman ed., 1969 p. 246-266.
- [3] BOGARD, D. D., CLARK, R. S., KEITH, J. E., REYNOLDS, M. A., *J. Geophys. Res.* **76** (1971) 4076.
- [4] CRESSY, P. J., RANCITELLI, L. A., *Earth Planet. Sci. Lett.* **22** (1974) 275.
- [5] CRESSY, P. J., *J. Geophys. Res.* **80** (1975) 1551.
- [6] HAMPEL, W., SCHAEFFER, O. A., *Earth Planet. Sci. Lett.* **42** (1979) 348.
- [7] NISHIZUMI, K., REGNIER, S., MARTI, K., *Earth Planet. Sci. Lett.* **50** (1980) 156.
- [8] GOEBEL, K., SCHULTES, H., ZHRINGER, J., CERN report 64-12 (1964).
- [9] SCHAEFFER, O. A., HEYMANN, D., *J. Geophys. Res.* **70** (1965) 215.
- [10] MARTI, K., *Phys. Rev. Lett.* **18** (1967) 264.
- [11] VOSHAGE, H., FELDMANN, H., *Earth and Planet. Sci. Lett.* **45** (1979) 293.
- [12] TOBAILEM, J., DE LASSUS ST GENIES, C. H. LEVEQUE, L., report CEA-N-1466-1 (1971).
- [13] BAROS, F., Thèse n° 1850 (1983) Bordeaux.
- [14] REGNIER, S., *Phys. Rev. C* **20** (1979) 1517.
- [15] SAUVAGEON, H., to be published.
- [16] RAISBECK, G. M., YIOU, F., *Phys. Rev. C* **12** (1975) 915.
- [17] KORTELING, R. G., CARETTO, A. A., *Phys. Rev. C* **1** (1970) 193 and *Phys. Rev. C* **1** (1970) 1960.
- [18] LASSUS ST GENIES, C. H. (de), TOBAILEM, J., report CEA-N-1466-2 (1972). This review concerns F, Ne, Na and Mg targets. Volumes 3 and 4 concern respectively Fe and Al targets.
- [19] TOBAILEM, J., report CEA-N-1466-5 (1981). This volume compile cross sections in Si targets. Volumes 6, 7 and 8 concern respectively P, S and Cl targets.
- [20] KORTELING, R. G., CARETTO, A. A., *J. Inorg. Nucl. Chem.* **29** (1967) 2863.
- [21] WALTON, J. R., HEYMANN, D., YANIV, A., EDGERLEY, D., ROWE, M. W., *J. Geophys. Res.* **79** (1974) 314.
- [22] PULFER, P., Thesis, Bern (1979).
- [23] BIERI, R. H., RUTSCH, W., *Helvetica Phys. Acta* **35** (1962) 553.
- [24] STOENNER, R. W., LYMAN, W. J., DAVIS, R. Jr, *Proc. of the Apollo 11 Lunar Sci. Conf.* **2** (1970) 1583.
- [25] WALTON, J. R., HEYMANN, D., YANIV, A., EDGERLEY, D., ROWE, M. W., *J. Geophys. Res.* **81** (1976) 5689.
- [26] SILBERBERG, R., TSAO, C. H., *Astrophys. J. Supp.* **220** 25 (1973) 315.  
Improvements of the Silberberg and Tsao spallation formula were published in *Spallation nuclear reactions and their applications*, Shen, Merker, Eds, 1976, p. 49; and later in *Proceed. 16th Intern. Cosmic Ray Conf.*, Kyoto, 1979, p. 202 and *proceed. 19th Intern. Cosmic Ray Conf.*, India, 1983, paper OG 522.
- [27] KELLER, K. A., LANGE, J., MÜNDEL, H., Landolt Bornstein, vol. 5, part c (Springer-Verlag) 1974.
- [28] REEDY, R. C., HERZOG, G. F., JESSBERGER, E. K., *Earth Planet. Sci. Lett.* **44** (1979) 341.
- [29] DEDIEU, J., Thèse n° 1525, Bordeaux, 1979.
- [30] STEHN, J. R., GOLDBERG, M. D., MAGURMO, B. A., WIENER-CHASMAN, R., Neutron cross sections BNL 325, 1964.
- [31] NORMAN, E. B., *Astrophys. J.* **231** (1979) 198.
- [32] REEDY, R. C., private communication.
- [33] MOORE, C. B., LEWIS, C. F., NAVA, D., in *Meteorite Research*, Millman Ed., 1969, p. 738-748.
- [34] BEGEMANN, F., VILCSEK, E., WANKE, H., *Earth Planet. Sci. Lett.* **3** (1967) 207.
- [35] CHENG, A., Ph. D. Thesis (1972) Stony Brook.
- [36] REGNIER, S., BAKLOUTI, M., SIMONOFF-LAGARDE, M., SIMONOFF, G. N., *Phys. Lett.* (1977) 202. Erratum privately circulated. 1979. All  $^{36}\text{Cl}$  cross sections in the 1977 paper are too high. Corrected values are given in Appendix.
- [37] REGNIER, S., BAROS, F., *Meteoritics* **17** (1982) 274.
- [38] EVANS, J. C., REEVES, J. H., RANCITELLI, L. A., BOGARD, D. D., *J. Geophys. Res.* **87** (1982) 5577.
- [39] REEDY, R. C., ARNOLD, J. R., *J. Geophys. Res.* **77** (1972) 537.
- [40] SCHAEFFER, O. A., ZHRINGER, J., *Phys. Rev.* **113** (1959) 674.