

SEARCH FOR EXTINCT ALUMINUM-26 AND TITANIUM-44 IN DIAMONDS FROM THE ALLENDE AND MURCHISON METEORITES. A. Besmehn, P. Hoppe, R. Strebel, and U. Ott, Max-Planck-Institute for Chemistry, P.O. Box 3060, D-55020 Mainz, Germany (abesmehn@mpch-mainz.mpg.de).

Introduction: Presolar diamonds are the most abundant presolar grains (up to ≈ 1000 ppm) identified in primitive meteorites. Their origin is still controversial, particularly because their $^{12}\text{C}/^{13}\text{C}$ ratio is close to solar [1]. From high-resolution TEM studies Daulton et al. [2] argued that most of the diamonds were formed by chemical vapor deposition in circumstellar environments and not by shock-induced grain-grain collisions in the interstellar medium [3]. At least a fraction of the diamonds appears to have formed in the ejecta of supernova (SN) explosions as indicated by the isotopic compositions of Xe [4] and Te [5].

Presolar circumstellar grains exhibit isotopic compositions which are characteristic for their stellar sources. Particularly diagnostic are the elements Mg and Ca. Large excesses in ^{26}Mg can be attributed to the in situ decay of radioactive and now extinct ^{26}Al ($T_{1/2} \approx 700000$ years) which is produced by nuclear reactions in the interior of stars. Aluminum-26 is most abundant in supernova grains (SiC X grains, Si_3N_4 , graphite) with inferred initial ^{26}Al concentrations of ≈ 100 - 1000 ppm and $^{26}\text{Al}/^{27}\text{Al}$ ratios of $\approx 10^{-2}$ to 10^{-1} [e.g., 6-9]. On the other hand, presolar grains from AGB and red giant stars (SiC mainstream, corundum, and spinel) have less ^{26}Al with $^{26}\text{Al}/^{27}\text{Al}$ ratios of $\approx 10^{-4}$ to 10^{-3} [e.g., 10, 11]. Another diagnostic isotope with respect to the origin of presolar grains is radioactive ^{44}Ti ($T_{1/2} \approx 60$ years). Supernovae are the only stellar sources that produce ^{44}Ti and, provided that the mixing in the ejecta is deep, radiogenic ^{44}Ca can be expected to be present in meteoritic SN grains. In fact, radiogenic ^{44}Ca is observed in some SiC X and graphite grains. Inferred initial $^{44}\text{Ti}/^{48}\text{Ti}$ ratios of SiC X grains are, on average, between $4 \cdot 10^{-3}$ and $5 \cdot 10^{-2}$ depending on grain size and the inferred average ^{44}Ti concentrations range from 10 to 40 ppm [e.g., 9, 12]. Similar $^{44}\text{Ti}/^{48}\text{Ti}$ ratios are inferred for SN graphite grains [e.g., 12, 13].

In order to get additional information on the origin of meteoritic diamonds we were searching for extinct ^{26}Al and ^{44}Ti in acid-resistant residues from the Allende CV3 and Murchison CM2 meteorites.

Experimental: Both meteorites were treated with the standard chemical procedure [14] in order to isolate the nanodiamonds. One additional Murchison sample (further called Murchison microwave) was prepared by a new microwave technique [15]. The isotopic composition of Mg together with ^{27}Al was measured in all three samples. In addition, the two samples prepared by

the standard method were analyzed for their Ca-Ti isotopic compositions. The isotopic measurements were made with the modified Cameca IMS3f ion microprobe at the Max-Planck-Institute for Chemistry in Mainz, using a 10-20 nA primary O^- beam. The sensitivity factors of Mg, Al, Ca, and Ti relative to C which are required to determine elemental abundances and $^{26}\text{Al}/^{27}\text{Al}$ and $^{44}\text{Ti}/^{48}\text{Ti}$ ratios were measured on a 9:1 mixture (by weight) of synthetic diamond powder and fine-grained NIST SRM 611 glass.

Results: Mg and Al abundances are low in the diamond samples with solar-normalized Mg/C and Al/C ratios of 10^{-6} and $4 \cdot 10^{-4}$, respectively, for Allende, and $1.7 \cdot 10^{-5}$ and $2.7 \cdot 10^{-2}$, respectively, for Murchison. The $^{26}\text{Mg}/^{24}\text{Mg}$ ratios of the Allende and Murchison microwave diamonds are essentially solar with $\delta^{26}\text{Mg}$ values of 4 ± 7 ‰ for Allende and 5 ± 13 ‰ for Murchison. From these data we infer an upper limit (2σ) for $\delta^{26}\text{Mg}^*$ (from the decay of ^{26}Al) in the Allende diamonds of 18‰ corresponding to an $^{26}\text{Al}/^{27}\text{Al}$ ratio of $9 \cdot 10^{-5}$ and an ^{26}Al concentration of < 1 ppb (Fig. 1). For the Murchison microwave sample we get an upper limit of $\delta^{26}\text{Mg}^* = 31$ ‰, an $^{26}\text{Al}/^{27}\text{Al}$ ratio of $< 1.4 \cdot 10^{-4}$, and an ^{26}Al concentration of < 53 ppb (Fig. 1).

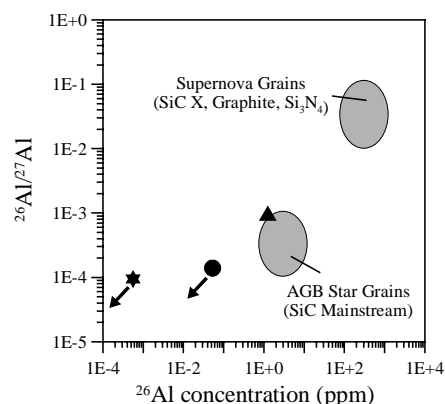


Fig.1: $^{26}\text{Al}/^{27}\text{Al}$ and ^{26}Al concentrations in meteoritic diamonds. Asterisk = Allende, triangle = Murchison, circle = Murchison microwave

On the other hand, the Murchison diamond sample treated with the standard chemical procedure is highly enriched in ^{26}Mg with $\delta^{26}\text{Mg} = 3830 \pm 530$ ‰. It is well known that Murchison diamonds prepared by the standard chemical procedure contain fine-grained presolar SiC [2]. From the ^{26}Mg excess we calculate

$^{26}\text{Al}/^{27}\text{Al} \approx 10^{-3}$ and $[\text{Al}] \approx 1$ ppm (Fig. 1), which corresponds to about 1-2 wt% SiC in the diamond sample.

Solar-normalized Ca and Ti concentrations are between $3 \cdot 10^{-4}$ (Allende) and $9 \cdot 10^{-4}$ (Murchison) and, respectively, between $4 \cdot 10^{-3}$ (Allende) and $4 \cdot 10^{-2}$ (Murchison). The $^{44}\text{Ca}/^{40}\text{Ca}$ ratios are close to solar in both samples with $\delta^{44}\text{Ca} = 8 \pm 8$ ‰ for Allende and $\delta^{44}\text{Ca} = 10 \pm 7$ ‰ for Murchison. From that we infer upper limits (2σ) for $\delta^{44}\text{Ca}^*$ (from the decay of ^{44}Ti) of 24 ‰, corresponding to initial $^{44}\text{Ti}/^{48}\text{Ti}$ ratios of $1 \cdot 10^{-3}$ (Allende) and $5 \cdot 10^{-4}$ (Murchison) and ^{44}Ti concentrations of 4 ppb and 12 ppb, respectively (Fig. 2).

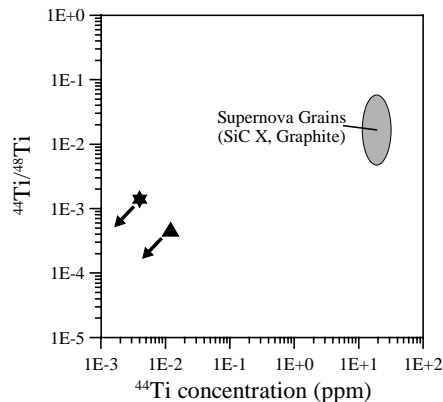


Fig.2: $^{44}\text{Ti}/^{48}\text{Ti}$ and ^{44}Ti concentrations in meteoritic diamonds. For symbols see Fig.1.

Discussion: The inferred upper limits of the $^{26}\text{Al}/^{27}\text{Al}$ and $^{44}\text{Ti}/^{48}\text{Ti}$ ratios are clearly lower than those of SN grains (Figs. 1 and 2). The Al and Ti isotopic data give thus no evidence for a SN origin of a large fraction of the diamonds. On the other hand, the upper limits on the $^{26}\text{Al}/^{27}\text{Al}$ ratios and the missing evidence for the presence of now extinct ^{44}Ti are compatible with an AGB star origin for the majority of the diamonds. The inferred upper limits on the $^{26}\text{Al}/^{27}\text{Al}$ and $^{44}\text{Ti}/^{48}\text{Ti}$ ratios would be higher if Al- and Ti-bearing grains contributed to the measured Al and Ti signals. Ion imaging and high resolution SEM/EDX investigations have shown the presence of only a few Al-bearing grains (> 500 nm). They are easily recognized in the ion microprobe by an increase in the Al signal and these measurement cycles have been removed from our data before the calculation of the $^{26}\text{Al}/^{27}\text{Al}$ ratios. During most measurement cycles constant Al/C and Ti/C ratios were observed, indicative of homogeneous distribution of Al and Ti in the diamond samples. Especially the Allende diamonds exhibit very low solar-normalized Al/C and Ti/C ratios that are comparable or even lower than the solar-normalized Xe/C ratio (Fig. 3), suggesting that a major fraction of

Al and Ti is intrinsic to the diamonds and not from Al- and Ti-bearing grains in the diamond samples.

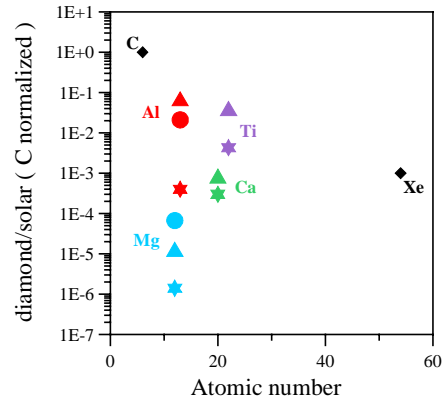


Fig.3: Elemental abundances in Allende and Murchison diamonds normalized to carbon and solar. For symbols see Fig.1.

It is possible that a fraction of originally present radiogenic ^{26}Mg and ^{44}Ca got lost from the diamonds by recoil after radioactive decay of ^{26}Al and ^{44}Ti , respectively. The medium range of ^{26}Mg with recoil energy of 66 eV (from the 1.8 MeV γ transition of ^{26}Mg) is ≈ 0.4 nm, that of ^{44}Ca with recoil energy of 17 eV (from the 1.2 MeV γ transition of ^{44}Ca) is ≈ 0.2 nm. When these numbers are compared with the typical diamond diameter of ≈ 2 -3 nm then it can be inferred that ≈ 75 % of radiogenic ^{26}Mg and ≈ 90 % of radiogenic ^{44}Ca is preserved in the diamonds. We thus conclude that $^{26}\text{Al}/^{27}\text{Al}$ and $^{44}\text{Ti}/^{48}\text{Ti}$ ratios characteristic for presolar SN grains should have been recognizable in the meteoritic diamonds.

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