

## Iron and nickel isotopic compositions of presolar SiC grains from supernovae

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Approximately 1% of presolar SiC grains found in primitive meteorites condensed in the ejecta of Type II supernovae. To explain the isotopic signatures of these so-called X grains, material from different SN zones had to contribute to the mix from which the grains formed. We measured the Fe and Ni isotopic ratios of individual X grains with the NanoSIMS. Most grains have enhanced  $^{57}\text{Fe}/^{56}\text{Fe}$  ratios ranging up to 2×solar. A few grains have depleted  $^{57}\text{Fe}/^{56}\text{Fe}$  ratios down to 0.55×solar. In contrast,  $^{54}\text{Fe}/^{56}\text{Fe}$  ratios are close to solar. Anomalies in the Ni isotopes are even more extreme.  $^{61}\text{Ni}/^{58}\text{Ni}$  ratios range up to 3.5×solar,  $^{62}\text{Ni}/^{58}\text{Ni}$  ratios up to 2×solar, but  $^{60}\text{Ni}/^{58}\text{Ni}$  ratios only up to 1.1×solar. One of the grains with  $^{57}\text{Fe}$  deficits has a  $^{61}\text{Ni}$  deficit, the others have normal  $^{61}\text{Ni}/^{58}\text{Ni}$  ratios. The grain with the largest  $^{57}\text{Fe}$  deficit has  $^{62}\text{Ni}/^{58}\text{Ni}$  of 1.8×solar but solar  $^{61}\text{Ni}/^{58}\text{Ni}$ . The excesses in  $^{57}\text{Fe}$  and  $^{61,62}\text{Ni}$  observed in most of the grains can be explained by admixture of material from the He/C zone to material from the He/N (or outer) zones. In the He/C zone, neutron capture results in  $^{57}\text{Fe}$  and  $^{61,62}\text{Ni}$  excesses, and the grain data can be fairly well reproduced by variable admixture from different layers of the He/C zone. The lack of large  $^{54}\text{Fe}$  excesses in the grains is puzzling in view of the fact that material from the Si/S zone, rich in  $^{54}\text{Fe}$ , is required to explain the  $^{28}\text{Si}$  excesses of X grains. We still do not have a good explanation for the  $^{57}\text{Fe}$  deficits of several of the grains; they could reflect the initial isotopic compositions of the parent stars.

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## 1. Introduction

Primitive meteorites contain grains that condensed in the outflows of evolved stars and in SN ejecta [e. g., 1]. These grains are characterized by large isotopic anomalies and their study in the laboratory provides information on stellar nucleosynthesis and evolution, Galactic chemical evolution, physical and chemical conditions of stellar atmospheres, and conditions in the early Solar nebula and on meteorite parent bodies [2]. Among the different types of presolar grains, SiC has been studied in most detail because it is chemically very refractory and by chemical separation can be isolated in almost pure form [3]. While most SiC grains come from AGB stars, ~1% of them originated in Type II supernovae. These grains, called X grains, are characterized by  $^{15}\text{N}$  and  $^{28}\text{Si}$  excesses, mostly isotopically light C, high inferred  $^{26}\text{Al}/^{27}\text{Al}$  ratios, and evidence for the initial presence of the short-lived radioisotopes  $^{44}\text{Ti}$  and  $^{49}\text{V}$  [1].

We measured the Fe and Ni isotopic ratios of 37 X grains from the Murchison carbonaceous meteorite, identified from their C, N, and Si isotopic compositions, by secondary ion mass spectrometry (SIMS) with the NanoSIMS. In addition to  $^{54,56,57}\text{Fe}$  and  $^{58,60,61,62}\text{Ni}$ , we measured the  $^{52}\text{Cr}$  signal in order to make a correction for  $^{54}\text{Cr}$  interference. We made also a correction for the  $^{58}\text{Fe}$  isobar, which cannot be separated from  $^{58}\text{Ni}$ .

## 2. Isotopic ratios and comparison with SN models

### 2.1. Iron isotopic ratios

The Fe isotopic ratios of the X grains are shown in Fig. 1 as so-called delta-values, deviations from the solar ratios in permil (‰).

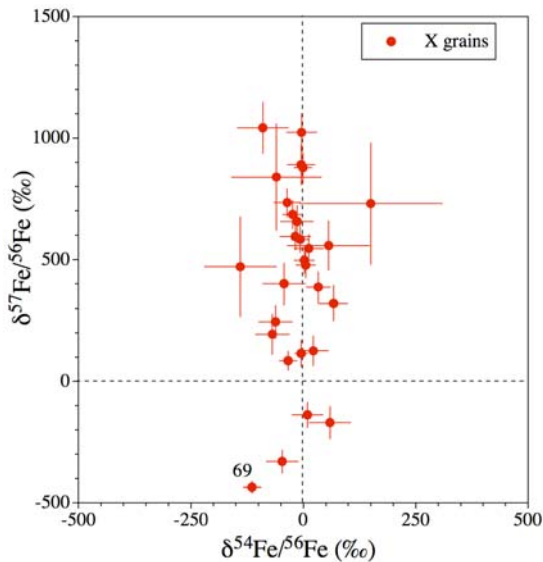


Figure 1. Iron isotopic ratios of X grains.

Most grains have  $^{57}\text{Fe}$  excesses, a few have  $^{57}\text{Fe}$  deficits. In contrast to the  $^{57}\text{Fe}$  anomalies, the  $^{54}\text{Fe}/^{56}\text{Fe}$  ratios are close to

solar. In Type II supernovae  $^{57,58}\text{Fe}$ , as well as  $^{61,62}\text{Ni}$ , are enhanced in the regions that experienced neutron capture (the weak s-process). These are the zones below the H-rich envelope and the He/N zone. Figure 2 shows the abundances of the Fe and Ni isotopes in the  $25M_{\odot}$  SN model by Rauscher et al. [4]. In the H and He/N zones the Fe and Ni isotopic ratios are normal, but the  $^{57,58}\text{Fe}$  and  $^{61,62}\text{Ni}$  abundances suddenly increase at the boundary to the He/C zone and keep increasing throughout this zone toward the O/C zone, while the  $^{54,56}\text{Fe}$  and  $^{58,60}\text{Ni}$  abundances decrease. The grains' Fe isotopic compositions can be reproduced by mixing of material from the He/N zone with material from the He/C and from the O/C zone. Figure 3 shows lines obtained by such mixing with variable mixing ratios. Of the He/C zone we chose two layers, one just below the He/N-He/C boundary (Mix He/C-a) and the other a layer close to the O/C zone (Mix He/C-b).

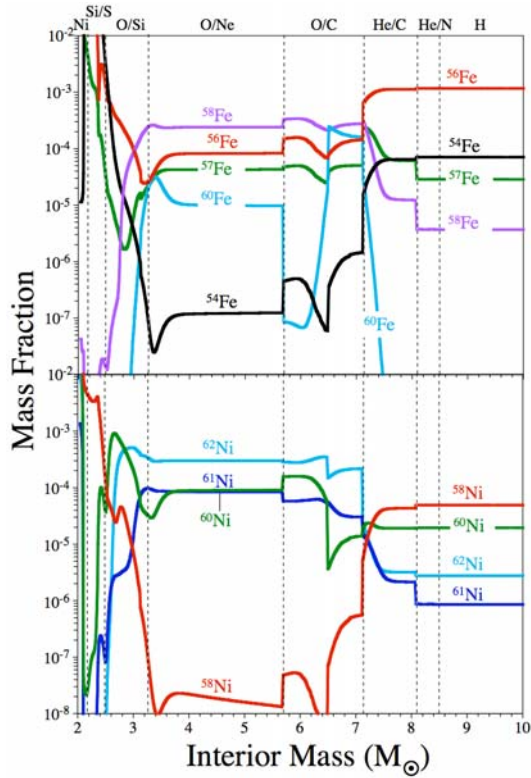


Figure 2. Theoretically predicted mass fractions of the Fe and Ni isotopes inside the  $25M_{\odot}$  SN model by Rauscher et al. (2002).

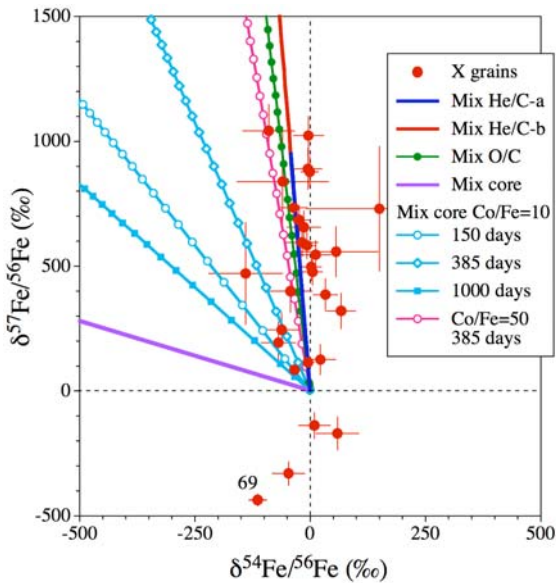


Figure 3. Fe isotopic ratios of X grains and predicted lines of mixtures between the He/N zone and the He/C, O/C zones and the Ni core, respectively.

The Ni core is rich in all Ni isotopes and in  $^{56,57}\text{Fe}$  from the decay of  $^{56,57}\text{Ni}$  (Fig. 4). We thus also considered mixing between the He/N

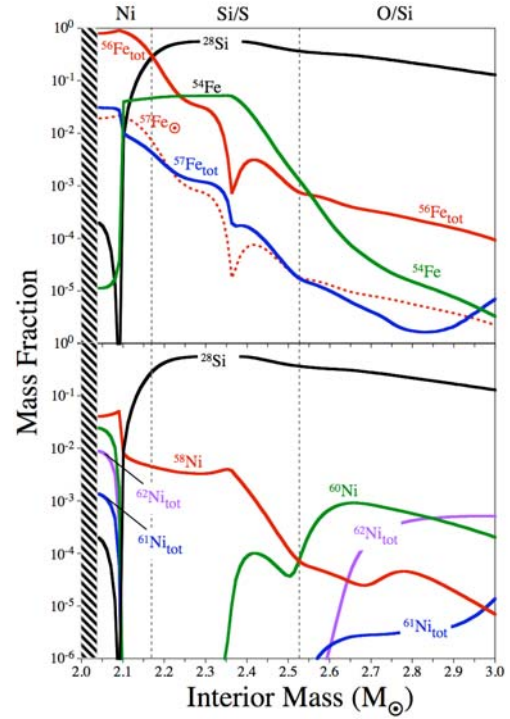


Figure 4. Predicted mass fractions of Fe and Ni isotopes in the interior zones of a  $25M_{\odot}$  SN. The abundances of the nuclides labelled “tot” are those after the decay of radioactive precursors. The line labelled  $^{57}\text{Fe}_{\odot}$  depicts the abundance of  $^{57}\text{Fe}$  if the  $^{57}\text{Fe}/^{56}\text{Fe}$  ratio is solar.

zone and the Ni core. The mixing line misses the grain data (Fig. 3) because  $^{54}\text{Fe}$  in the core is extremely low. However, the Co/Fe ratios measured in the grains are larger than solar and since the precursors  $^{56}\text{Co}$  and  $^{57}\text{Co}$  have half lives of 77 and 272 days, respectively, we have to consider that the  $^{57}\text{Fe}$  condensed as  $^{57}\text{Co}$  into the grains. Fig. 3 shows mixing lines for a fractionation factor of ten and varying grain condensation times. The maximum (negative) slope is reached after 385 days, but this line misses most data points. One has to increase the fractionation factor (to 50 in the figure) to get close to the data. However, most grains have Co/Fe ratios of  $\sim 10$ , thus mixing with core material cannot explain their Fe isotopic ratios.

The  $^{54}\text{Cr}$  interference correction for the data in Figs. 1 and 3 was made with the assumption of a solar  $^{54}\text{Cr}/^{52}\text{Cr}$  ratio. Since

$^{54}\text{Cr}$  is enriched in the He/C zone, we improved the correction by scaling the assumed  $^{54}\text{Cr}/^{52}\text{Cr}$  ratios to the  $^{57}\text{Fe}/^{56}\text{Fe}$  ratios measured in the grains and the ratios in the SN model. The effect is small and, with two exceptions, the decrease in the  $^{54}\text{Fe}/^{56}\text{Fe}$  ratios is smaller than the experimental uncertainties.

## 2.2. Nickel isotopic ratios

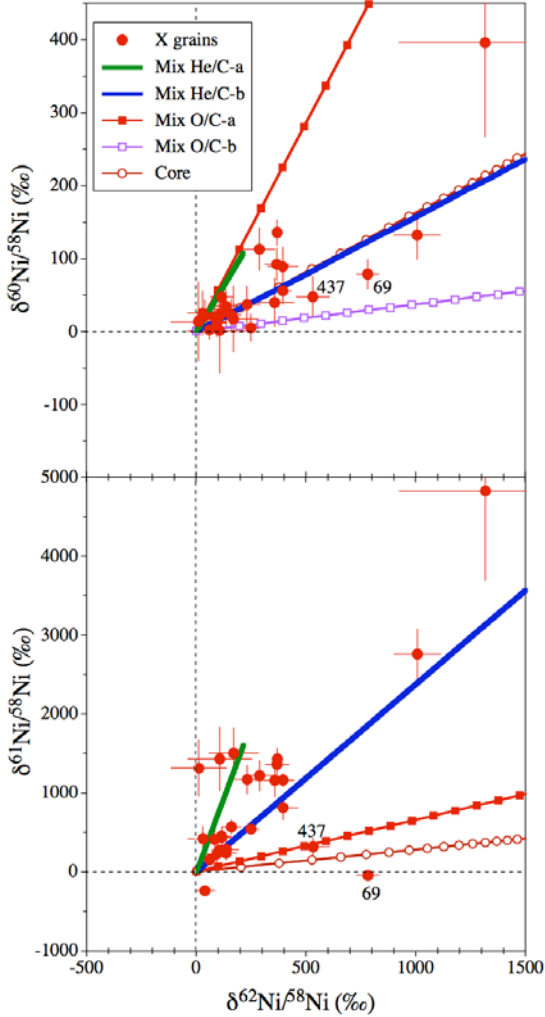


Figure 5. Ni isotopic ratios of X grains and predicted lines of mixtures between the He/N zone and the He/C, O/C zones, and the Ni core, respectively.

Nickel isotopic ratios of the X grains are plotted in Fig. 5. For  $^{58}\text{Ni}$  we have to make a correction for the  $^{58}\text{Fe}$  interference. Fortunately, Ni/Fe ratios in the grains are much higher than solar (Fig. 6), so that the  $^{58}\text{Fe}$  correction is

relatively small. Similar to the  $^{54}\text{Cr}$  correction, we scaled the assumed  $^{58}\text{Fe}/^{56}\text{Fe}$  ratios to the  $^{57}\text{Fe}/^{56}\text{Fe}$  ratios measured in the grains and the model ratios in the He/C zone (Fig. 2).

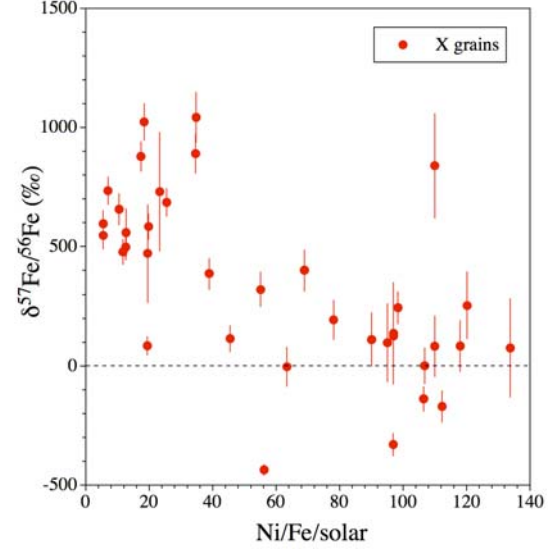


Figure 6.  $^{57}\text{Fe}/^{56}\text{Fe}$  ratios of X grains vs. solar-normalized Ni/Fe ratio.

The Ni isotopic ratios in the grains are characterized by large excesses in  $^{61}\text{Ni}$  and  $^{62}\text{Ni}$ , and by smaller excesses in  $^{60}\text{Ni}$ . Grain 69, which has a  $^{57}\text{Fe}$  deficit (Fig. 1), is an exception, having a large  $^{62}\text{Ni}$  excess but no  $^{61}\text{Ni}$  excess. As with the Fe isotopic ratios, we compare the Ni isotopic ratios with the results of mixing calculations for the Rauscher et al.  $25M_{\odot}$  SN model. We again use two layers for the He/C zone. The outer O/C zone has a high abundance of  $^{60}\text{Fe}$ , which decays into  $^{60}\text{Ni}$ . The  $^{60}\text{Ni}/^{58}\text{Ni}$  ratio thus depends on the Fe/Ni ratios of the grain. In Fig. 5 we plot mixing lines for the extreme Fe/Ni ratios observed in the grains. The Rauscher et al. (2002) models used the nuclear cross sections given by Bao et al. [5]. Recently, new neutron-capture data on  $^{60}\text{Ni}$  and  $^{62}\text{Ni}$  became available [6, 7], indicating an increase of the  $^{62}\text{Ni}$  cross section by almost a factor of two. To account for this change, we estimated the  $^{62}\text{Ni}$  abundances in the SN layers that

experienced neutron capture by using the  $^{54}\text{Fe}$  as an indicator of total neutron exposure [8]. The resulting decrease in  $^{62}\text{Ni}$  abundance means that in Fig. 5 the mixing lines involving the He/C and O/C zones are steeper than they would be with the old cross sections.

As can be seen in Fig. 5, in the  $\delta^{60}\text{Ni}/^{58}\text{Ni}$  vs  $\delta^{62}\text{Ni}/^{58}\text{Ni}$  plot all mixing lines cover the grain data fairly well. However, in the  $\delta^{61}\text{Ni}/^{58}\text{Ni}$  vs  $\delta^{62}\text{Ni}/^{58}\text{Ni}$  plot the O/C and core mixing lines are close only to grains 69 and 437 but miss all the other data points. We conclude that the Fe and Ni isotopic ratios of X grains are best explained by mixing of material from the He/N zone with varying amounts of material from different layers of the He/C zone and that contributions from the O/C zone and the Ni core must be limited.

### 3. Remaining problems

There are several remaining problems. One is the  $^{57}\text{Fe}$  deficits observed in a few grains (Fig. 1). The only regions with  $^{57}\text{Fe}$  deficits in the SN model ( $^{57}\text{Fe}_{\text{tot}}$  is smaller than  $^{57}\text{Fe}_{\odot}$ ) are between 2.1 and 2.2 and between 2.52 and 2.93 interior mass (Fig. 4). However, in the first region  $^{54}\text{Fe}$  is too high and all Ni isotopes are much too low; in the second region the Fe abundance as a whole is too low relative to the abundance in the He/N zone and the Ni isotopic ratios are wrong (Figs. 2 and 4). In addition, this region is extremely O-rich, making it doubtful that SiC could condense from such a mixture.

Another problem is posed by the fact that X grains have sizeable  $^{28}\text{Si}$  excesses but lack any  $^{54}\text{Fe}$  excesses. Admixture from the Si/S zone, rich in  $^{28}\text{Si}$ , has been advocated to explain the Si isotopic ratios of X grains [9], and is necessary to account for  $^{44}\text{Ti}$  [10]. However, this zone is very rich in  $^{54}\text{Fe}$  (Fig. 4). In the grains, we find large fractionation between Ni and Fe, and between Co and Fe, but we do not understand such processes well enough to be able to say whether fractionation between Si and Fe from the Si/S zone can solve the  $^{54}\text{Fe}$  problem.

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