Presence of ^{60}Fe in eucrite Piplia Kalan: a new perspective to the initial $^{60}Fe/^{56}Fe$ in the early solar system

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Fe–Ni isotope measurements of ferrous pyroxenes of the Piplia Kalan eucrite using Secondary Ion Mass Spectrometer revealed the presence of ^{60}Ni excess corresponding to the initial $^{60}Fe/^{56}Fe$ of $(5.2 \pm 2.4) \times 10^{-9}$. Combining this ratio with the inferred initial $^{26}Al/^{27}Al$ ratio of $(7.5 \pm 0.9) \times 10^{-7}$ in plagioclase of the Piplia Kalan that has been already reported in the literature suggests initial $^{60}Fe/^{56}Fe$ of $(5.2 \pm 2.4) \times 10^{-8}$ in the early solar system, which is significantly lower than the value inferred from Fe–Ni isotope measurements of chondrules in silicates and sulphides from unequilibrated ordinary chondrites. We attribute the difference in the initial $^{60}Fe/^{56}Fe$ to be solely due to the closure temperature of Fe–Ni isotope systematics, which is less compared to Al–Mg isotope systematics. With the initial $^{60}Fe/^{56}Fe$ values found in the Piplia Kalan, it is possible that the Fe–Ni isotope systematics was disturbed for another ~ 5 Ma (million years) after closure of the Al–Mg isotope systematics. The closure temperature of ~ 450–650°C for Fe–Ni isotope system seems feasible and we anticipate a cooling rate of ~ 20–60°C/Ma in the crust region of the parent body of Piplia Kalan, thereby matching the initial $^{60}Fe/^{56}Fe$ to ~ 5×10^{-7}. This is consistent with the initial value found in the silicate phases in chondrules of least metamorphosed meteorites. However, the presence of ^{60}Ni excess in Piplia Kalan does not confirm ^{60}Fe to be a major heat source for the early thermal evolution of meteorite parent bodies. Also, it seems that a massive star probably has contributed the two short-lived nuclides, ^{26}Al and ^{60}Fe, to the early solar system.

Keywords: Cooling rate, early solar system, eucrite, Fe–Ni isotope, planetary differentiation.

THE conventional astrophysical scenario for the formation of the solar system involves a supernova-triggered initiation of the gravitational collapse of the pre-solar molecular cloud, forming a proto-sun surrounded by cloud of gas and dust, the so-called solar nebula. This scenario also led to the seeding of the short-lived radionuclides such as ^{41}Ca, ^{26}Al, ^{60}Fe, ^{53}Mn, etc. into the parent molecular cloud of the solar system[1]. ^{26}Al, with a half-life of 0.72 Ma (million years), is widely studied in early solar system objects such as CAIs (Ca–Al rich inclusions), chondrules, ureilites, pallasites, angrites and eucrites[2–14]. The Al–Mg isotope systematics led to the major understanding of the onset and duration of chondrule formation, lifetime of the active solar nebula, formation of the parent body(ies) of eucrites, etc. However in recent times, ^{60}Fe with a half-life of ~ 1.5 Ma has emerged as a promising chronometer for the study of the timescale and processes in early history of the solar system[8–10]. The presence of ^{60}Fe in the early solar system was resolved by the excess of ^{60}Ni in CAIs by Birck and Lugmair[11] with an upper limit of ~ 1.6×10^{-6} for initial $^{60}Fe/^{56}Fe$. Subsequent studies carried out on an eucrite sample of Chervony Kut and Juvinas indicated an initial $^{60}Fe/^{56}Fe$ ratio of $(3.9 \pm 0.6) \times 10^{-9}$ and $(4.3 \pm 1.5) \times 10^{-10}$ respectively[12,13]. These Fe–Ni isotope systematics studies on achondrites[12–14] showed that the inferred initial $^{60}Fe/^{56}Fe$ values in the solar system fell short by more than an order of magnitude than those inferred by silicate phases of chondrule and troilite.

Previous studies of Fe–Ni isotope systematics performed on Fe-rich silicate phases of chondrule and troilite from unequilibrated ordinary chondrites (UOCs)[4,8–10] provided an initial $^{60}Fe/^{56}Fe$ from $(1–10) \times 10^{-7}$. The wide range does not allow us to constrain the initial $^{60}Fe/^{56}Fe$ values for the early solar system considering the time difference between CAIs and chondrules to be ~ 1 Ma, like its counterpart, initial $^{26}Al/^{27}Al$ (= 5×10^{-5}) and hence may not assist towards the understanding of the chronology of early solar system objects. It also creates a dilemma in the choice of the right stellar source for injection of these two short-lived radionuclides. The primitive samples such as Semarkona (LL3.0), Bishunpur (LL3.1) and Krymka (LL3.1) even after having the least metamorphic grade did have a widespread distribution of initial $^{60}Fe/^{56}Fe$. In fact, the spread in the initial $^{60}Fe/^{56}Fe$ does seem to gain support for the heterogeneous distribution in the solar system, unlike that of ^{26}Al. Whether ^{60}Fe was homogenously distributed in the early solar system cannot be concluded at this point. Eucrites such as Piplia Kalan provide an opportunity to study the initial $^{60}Fe/^{56}Fe$ as these differentiated meteorites have well-documented initial $^{26}Al/^{27}Al$ (ref. 3). The essential aim of the present study is to look for evidence of live ^{60}Fe in eucrite Piplia Kalan, thereby providing an initial $^{60}Fe/^{56}Fe$ in the early solar system, and further to deduce any correlation that exists between ^{60}Fe and ^{26}Al.

The eucrite sample, Piplia Kalan, which is an equilibrated, noncumulate, monomict breccia with clasts was chosen for Fe–Ni isotope systematics measurements. The age of this meteorite Piplia Kalan by various techniques is reported to be 4.57 and 4.56 Ga by Sm–Nd age and Pu–Xe model age respectively[15,16]. The early formation of the Piplia Kalan parent body is evident from its deduced age and consequently is an exemplary sample that has provided with a major contribution by indicating ^{26}Al to be a major heat source[3]. The major minerals in the sample

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consist of pyroxene, plagioclase along with some minor ilmenite and iron sulphide. This sample proved to be just right due to the presence of the large Fe-rich pyroxene crystals of about a millimetre in size with high Fe/Ni ratio. The back-scattered electron image of a portion of the polished section of Piplia Kalan is shown in Figure 1.

The identified Fe-rich phases of pyroxene in the section of Piplia Kalan were analysed for Fe–Ni isotopic measurement using Secondary Ion Mass Spectrometer (SIMS Cameca IMS-4f); usually called as ion microprobe that was operated at a mass resolution ($M/\Delta M$) of ~ 4500, sufficient to resolve the major interference at masses ^{57}Fe, ^{60}Ni and ^{62}Ni of interest. Contributions from hydrides existed which cannot be avoided and were confirmed to be less than per mil level. A 10–20 μm focused primary O^- beam with current ≥ 5 nA accelerated to 17.5 kV was used to sputter secondary ions from the sample surface using the largest contrast aperture to maximize transmission. Measurements were carried out in peak jumping mode by cycling the magnet through the mass sequence of $^{57}Fe^+$, $^{60}Ni^+$, $^{62}Ni^+$ and counted on electron multiplier for 1, 20 and 60 s in each cycle operated in the pulse-counting mode. The large size of the pyroxene grains that have uniform composition provides an advantage to do repeated measurements in many cases. The tail corrections for ^{60}Ni and ^{62}Ni were made from their interference peaks of ^{44}CaO and ^{46}TiO (refs 9, 14). A relative ion yield for both elements, Fe and Ni, is close to the unity. The relative sensitivity estimated is similar to that found by Sugiura *et al.*[14].

The results of the Fe–Ni isotope systematics study performed on the Fe-rich phases of pyroxene of Piplia Kalan are given in Figure 2. The isochron plotted in Figure 2 is ^{60}Ni/^{62}Ni versus ^{56}Fe/^{62}Ni; the error bars shown are 1σ for clarity. The results indicate the presence of ^{60}Fe in Piplia Kalan eucrite, yielding an initial ^{60}Fe/^{56}Fe value of $(5.2 \pm 2.4) \times 10^{-9}$ (the errors are 2σ). The horizontal dashed line in Figure 2 is for the terrestrial line. The other two dashed lines that have initial ^{60}Fe/^{56}Fe value of 5×10^{-7} and of 5×10^{-8} respectively, are shown for comparison. The scattering in the ^{60}Ni/^{62}Ni at low Fe/Ni may be the result of disturbance in the Fe–Ni isotope systematics. In spite of the disturbance that was present in the Al–Mg isotope systematics and Fe–Ni isotope systematics, it was not sufficient to completely equilibrate the Mg and Ni isotopic composition in the plagioclase and pyroxene respectively[3]. The isochron shown in Figure 2 for Fe–Ni isotope systematics supports the possibility of the presence of excess ^{60}Ni in Piplia Kalan.

The inferred initial ^{60}Fe/^{56}Fe obtained from Fe-rich phases in Piplia Kalan is much lower than that measured by troilite phases in Semarkona[9] and Fe-rich silicate phases in chondrules from least metamorphosed chondrites[4,8,10] using ion microprobe. Nevertheless, the initial ^{60}Fe/^{56}Fe value in the Fe-rich phases of Piplia Kalan sample falls within the range of different achondrites that were previously analysed for Fe–Ni isotope systematics[12–14]. The compiled data of all the achondrites studied for the Fe–Ni isotope systematics are shown in Figure 3. The data of Fe–Ni isotope systematics from D'Orbigny, Sahara 99555 and NWA 1670 and Asuka 881371 are from the angrites, whereas Asuka 881394, Stannern, Chervony Kut, Juvinas and Piplia Kalan are from the eucrites. Many data has high errors with some are provided by upper limits[12–14]. Figure 3 signifies that angrites and eucrites are consistent and fall within the narrow band has the error been better, except in the case of Juvinas

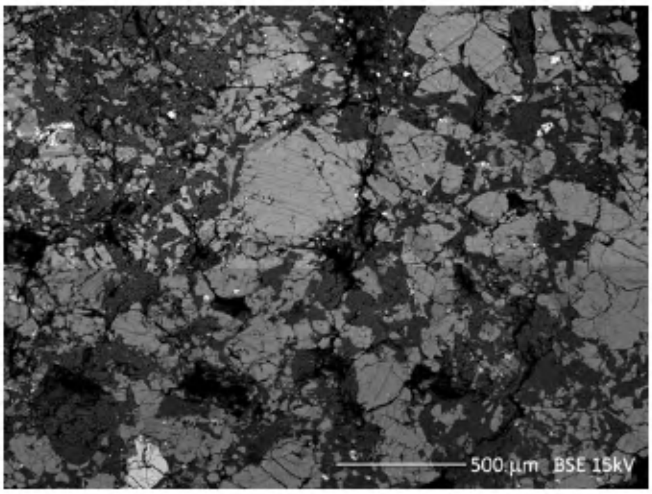

Figure 1. A section of back-scattered electron images of eucrite Piplia Kalan. The plagioclase grains are dark grey, while the pyroxenes are light grey. The pyroxene grains are large enough for ion microprobe analysis that has high Fe/Ni ratio.

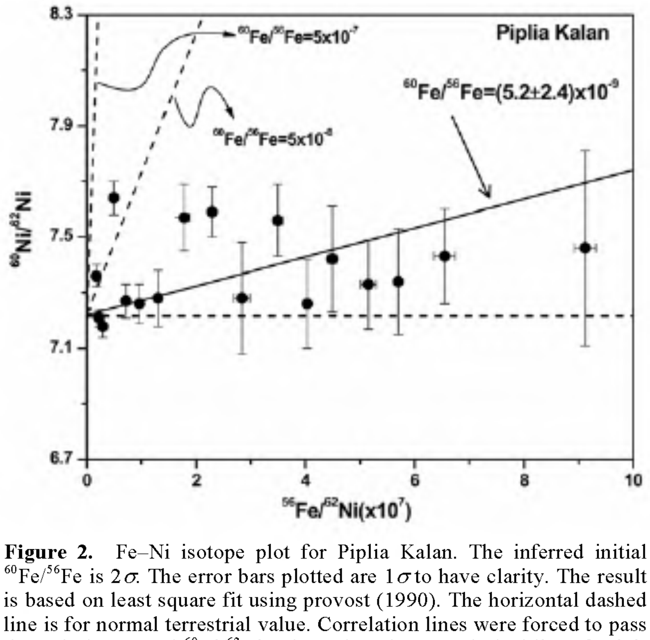

Figure 2. Fe–Ni isotope plot for Piplia Kalan. The inferred initial ^{60}Fe/^{56}Fe is 2σ. The error bars plotted are 1σ to have clarity. The result is based on least square fit using provost (1990). The horizontal dashed line is for normal terrestrial value. Correlation lines were forced to pass through the normal ^{60}Ni/^{62}Ni value. The other two dashed lines for initial ^{60}Fe/^{56}Fe of 5×10^{-7} and 5×10^{-8} respectively, are shown for comparison.

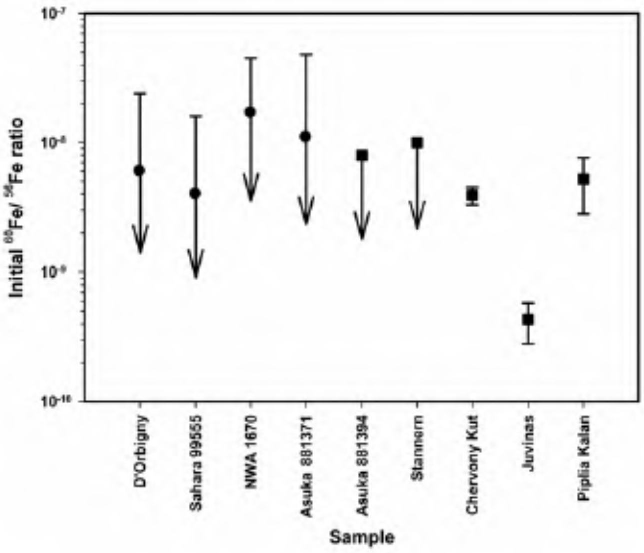

Figure 3. Distribution of the initial ^{60}Fe/^{56}Fe for achondrites. All errors plotted are 2σ. The angrites samples are D'Orbigny, Sahara 99555, NWA 1670, Asuka 881371 (circles). The eucrites are Asuka 881394, Stannern, Chervony Kut, Juvinas and Piplia Kalan (square)[12–14]. Most of the data fall within the narrow band except in Juvinas. Data with high error are provided by upper limits.

that has much lower initial ^{60}Fe/^{56}Fe value. However, the resulting ^{60}Fe/^{56}Fe ratio of $(5.2 \pm 2.4) \times 10^{-9}$ measured in Piplia Kalan is the best among the eucrites so far and is relatively well behaved when compared with other differentiated meteorites. The initial ^{60}Fe/^{56}Fe at the beginning of the solar system calculated from both angrites and eucrites (after ~ 5 Ma of CAIs age) approximately ranges from $(1–6) \times 10^{-8}$. The compilation of the angrites data (four in number) from the Sugiura *et al.*[14] gives a solar system initial ^{60}Fe/^{56}Fe value of $(6 \pm 9) \times 10^{-8}$. This is much larger with large errors than those from the eucrites data compilation (five in number), which provides an initial ^{60}Fe/^{56}Fe of $(6.5 \pm 1.4) \times 10^{-9}$. The eucrites have a larger spread in the initial ^{60}Fe/^{56}Fe values compared to the angrites, which has resulted in lower values. In the case of Piplia Kalan samples, the time difference between CAIs formation and crust formation in the parent body of Piplia Kalan is ~ 5 Ma from Al–Mg isotope systematics[3]. Taking 5 Ma as the benchmark for the Fe–Ni isotope systematics, the initial ^{60}Fe/^{56}Fe at the time of the solar system is $(5.2 \pm 2.4) \times 10^{-8}$. This is the best and the highest value till now among the eucrites. We do not have a suitable explanation for the spread in the eucrite values, but it seems at present that various minerals analysed by various authors have different resetting time and hence one should be careful while interpreting the data[14].

The initial ^{26}Al/^{27}Al in the Al-rich phases of plagioclase in Piplia Kalan has resulted in an upper limit of ~ 5 Ma for the formation, melting and crystallization of the eucrite parent body[3]. The deduced age of ~ 5 Ma for the Piplia Kalan parent body crust formation, if extrapolated for Fe–Ni isotope systematics, infers an initial ^{60}Fe/^{56}Fe of $(5.2 \pm 2.4) \times 10^{-8}$ at the beginning of the solar system. The inferred initial ^{60}Fe/^{56}Fe values found in the silicate phases of chondrules are larger by more than an order of magnitude than the above-mentioned value for Piplia Kalan. In such a situation the values found in the achondrites cannot be directly used for chronological interpretation. The smaller initial ^{60}Fe/^{56}Fe values found in achondrites compared to silicate phases in chondrules of least metamorphosed meteorites have prompted Sugiura *et al.*[14] to suggest that ^{60}Fe was heterogeneously distributed in the solar nebula.

The presence of the short-lived nuclides in the early solar system has been generally argued to be either due to irradiation or a late stage injection of the nuclides in the early solar system by an evolved star. We rule out the substantial irradiation contribution of ^{26}Al by the X-wind irradiation scenario, as it not possible to explain the confined spread in the initial canonical value of ^{26}Al/^{27}Al observed in the majority of the unaltered CAIs[17]. Further, the recent titanium isotopic analysis[18] indicates a direct genetic relationship between the CAIs and the bulk composition of the carbonaceous chondrites, thereby suggesting that CAIs were probably formed locally in the nebula by local transient thermal episodes rather than in the vicinity of the sun as proposed by the X-wind model. Hence, it is unlikely that the X-wind irradiation contributed to the thermal processing of CAIs and irradiation production of the short-lived nuclides. We favour massive star(s)[17,19–21] to be the source of the two short-lived nuclides, ^{26}Al and ^{60}Fe. Further, detailed theoretical analyses by Boss[22] suggest that stellar injection results in heterogeneity less than ~ 10% in the solar nebula. Even though this analysis was performed for ^{26}Al, it is applicable for ^{60}Fe. Apart from the dynamical processes operating in the solar nebula that result in the homogenization of the stellar ejecta, e.g. the extent of turbulence, the degree of heterogeneity of a short-lived nuclide would depend upon the nature of the carrier dust grain that brings in the short-lived nuclide to the solar system[20]. Among the two short-lived nuclides, ^{26}Al and ^{60}Fe, the former could be carried by comparatively more refractory grains. Hence, it is more likely that in case the two short-lived nuclides were injected into the solar system, the latter would homogenize earlier due to the comparatively less refractory nature of the carrier dust grain. This makes them less resistant against vaporization by the local transient thermal events in the solar nebula that aid in homogenization.

As already mentioned, there is a discrepancy in establishing the initial ^{60}Fe/^{56}Fe values present in the early solar system as one makes an effort to measure Fe–Ni isotope systematics in CAIs, troilite, chondrules, eucrites, etc. The inferred initial ^{60}Fe/^{56}Fe of $\sim 10^{-6}$ in troilite phases as measured by Mostefaoui *et al.*[9] is the highest so far, whereas the troilite phases measured by Tachibana and Huss[8] yield an order of magnitude lower value. The inferred initial $^{60}Fe/^{56}Fe$ in silicate phases[8] of chondrules is $(2.2–3.7) \times 10^{-7}$, whereas the present study on Piplia Kalan eucrite yields an order of magnitude lower value, but is nonetheless within the range of achondrites (Figure 3). The Al–Mg and Fe–Ni isotope data of Mishra *et al.*[23] has shown that chondrule formation was over by ~3 Ma from both the systematics (the initial $^{26}Al/^{27}Al$ and $^{60}Fe/^{56}Fe$ at the time of CAI formation were taken as $\sim 5 \times 10^{-5}$ and $\sim 5 \times 10^{-7}$ respectively). The lower values of the initial $^{60}Fe/^{56}Fe$ in the achondrites, in general, could be attributed either to the late Fe–Ni closure compared to Al–Mg, or alternatively, that both of the systematics were disturbed to different extents by thermal metamorphism subsequent to the crustal formation of the eucrite parent body. It seems that the higher initial $^{60}Fe/^{56}Fe$ in troilite phases is due to mild parent-body metamorphism where Fe gets more diffused compared to Ni, whereas in case of silicate both Fe and Ni are mobile[24]. The diffusion of Fe and Ni from silicate during metamorphism can lower the original isochron as it is seen in Piplia Kalan and different achondrites. These chondrules with the initial $^{60}Fe/^{56}Fe$ of $(2–4) \times 10^{-7}$, allow us to draw a conclusion that initial $^{60}Fe/^{56}Fe$ in the early solar system was probably $\sim 5 \times 10^{-7}$. Considering the above as initial $^{60}Fe/^{56}Fe$ value for the early solar system, the time difference drawn from Piplia Kalan yields ~10 Ma till the closure of the Fe–Ni isotope systematics. This suggests that the diffusion of Ni in pyroxene phase went on for another ~5 Ma after closure of Al–Mg isotope systematics. The closure temperature of Ni in pyroxene is difficult to know experimentally. However, it is reasonable to believe that ~450–650°C is the closure temperature for Ni for the time period of ~1 Ma, where it will not retain its heterogeneity. This reduces the temperature to ~100–300°C over a period of ~5 Ma, allowing us to estimate roughly that the cooling rates lie somewhere between 20 and 60°C/Ma. The ~5 Ma of cooling will raise the initial $^{60}Fe/^{56}Fe$ ratio to an order of magnitude from its present values, placing them with the values found in the silicate phases of chondrules. ^{26}Al is mostly concentrated in the plagioclase phases of eucrite whose grains sizes are ~1 mm. The heterogeneity in the plagioclase grain would be lost for these grain sizes at a temperature of ~750°C within a time period of ~1 Ma (ref. 25). But even for temperature <750°C, ^{60}Ni will diffuse easily, or the Fe–Ni systematics would be reset till it reaches a particular critical temperature where no diffusion of ^{60}Ni takes place. We believe that the critical temperature to prevent ^{60}Ni diffusion is far less compared to ^{26}Mg.

The initial $^{60}Fe/^{56}Fe$ ratio of $\sim 5 \times 10^{-7}$ in the early solar system, deduced from the achondrites by taking cooling rate into consideration, is not exclusively significant for the heating and differentiation of the parent bodies of eucrites[26,27]. In order to be an efficient heat source, an initial $^{60}Fe/^{56}Fe > 10^{-6}$ would be essential[26,27]. The time difference between CAIs and chondrule formation and the duration of chondrule formation estimated from Al–Mg isotope systematics is ~2–3 Ma, done on the primitive samples by various group where one does on expect any ^{26}Mg diffusion[4–6]. Hence, major accretion of chondrite parent bodies may have occurred ~2–3 Ma subsequent to the formation of the CAIs, while the accretion and differentiation of differentiated planetesimals commenced within a million years. While ^{26}Al powered the essential thermal energy for the planetary differentiation, the accumulation of ^{60}Fe in the growing iron core could have maintained heat for an extended time. The Al–Mg isotope systematics by Srinivasan *et al.*[3] suggests that the formation, melting and crystallization was over within 5 Ma, but the Fe–Ni isotope systematics suggests that the activity of metamorphism went on for another 5 Ma, after which the diffusion or elemental mobility stopped in Piplia Kalan. This can also be interpreted as the parent body of eucrite being active at least for ~10 Ma after CAI formation. Hence, the initial $^{60}Fe/^{56}Fe$ values obtained from various achondrites previously and by us are too small to be given serious consideration in terms of heating the meteorite parent body during the early evolution.

The source of ^{60}Fe along with the other short-lived nuclides has been debated for a long time. Although the source of the short-lived nuclides $^{7,10}Be$, ^{53}Mn and ^{36}Cl was probably irradiation in the early solar system, the canonical value of $^{26}Al/^{27}Al$ would be difficult to obtain by an irradiation scenario, e.g. the X-wind irradiation scenario[17,20–22]. Further, it is highly unlikely that ^{60}Fe was produced by irradiation. Hence, it is quite plausible that the two short-lived nuclides, ^{26}Al and ^{60}Fe, were produced by stellar source(s)[17,20–22]. The two short-lived nuclides, along with the other short-lived nuclides, e.g. ^{41}Ca, were either injected into the proto-solar molecular cloud or the proto-planetary disc. Numerous stellar sources have been proposed by different workers. These include a Thermally-Pulsing Asymptotic Giant Branch (TP-AGB) star, Nova, supernova SN II and Wolf-Rayet star evolved through SN Ib/c (ref. 20). Even though the lower value of 5×10^{-7} for $^{60}Fe/^{56}Fe$ compared to an initial value suggested by Mostefaoui *et al.*[9] makes the case for a low metallicity TP-AGB star a little more tempting, the various dynamical constraints associated with the scenario involving an AGB-triggered gravitational collapse of proto-solar molecular cloud make the AGB star an unlikely source of the short-lived nuclides[17,20]. It would be extremely difficult for the low-velocity winds associated with an evolving low-mass TP-AGB star to trigger star formation and inject short-lived nuclides[20]. There does not exist any stellar scenario involving a slow mixing of TP-AGB winds with a proto-solar molecular cloud model, eventually leading to the gravitational collapse of the cloud core. However, in case we do not impose constraints on the stellar source of the short-lived nuclides to be responsible for triggering the gravitational collapse of the presolar cloud, TP-AGB winds can provide the short-lived nuclides. Further, among the massive stellar scenarios, stars with mass > 30 $M_\odot$ seem to be the most plausible source(s). These massive stars evolve through the Wolf–Rayet (WR) stage, eventually leading to SN Ib/c supernova explosion[17]. ^{26}Al is produced in these stars during the WR stages and SN Ib/c, whereas ^{60}Fe is produced during SN Ib/c supernova. Estimates based on the integrated yields of these radionuclides for the entire evolution of these massive stars can explain the presence of the short-lived nuclides ^{26}Al and ^{60}Fe (ref. 20). The half-life of ^{60}Fe was revised to 2.6 Ma by Rugel *et al.*[28]. This may not have serious implications on our understanding of ^{60}Fe as a heat source in the early solar system, even after using 2.6 Ma as the half-life. However, the revised half-life will change our understanding of the initial ^{60}Fe/^{56}Fe in the early solar system.

The Fe–Ni isotope measurements in eucrite Piplia Kalan have revealed the presence of ^{60}Ni excess. The inferred initial ^{60}Fe/^{56}Fe value of $(5.2 \pm 2.4) \times 10^{-9}$ in Piplia Kalan is the best so far among the eucrites, measured using ion microprobe. The initial ^{60}Fe/^{56}Fe value found in Piplia Kalan is consistent with the silicates study carried out on the chondrules, provided the cooling scenario is invoked, as Fe–Ni isotope systematics is easily disturbed at 750°C, which is the closure temperature of the Al–Mg isotope systematics. But the closure temperature of Fe–Ni is ~ 450–650°C and the time required to cool will be ~ 5 Ma, thereby suggesting a cooling rate of ~ 20–60°C/Ma. The initial ^{60}Fe/^{56}Fe value inferred from eucrite Piplia Kalan rules out ^{60}Fe as a major heat source in the early solar system. A massive star injected the short-lived radionuclides, ^{26}Al and ^{60}Fe, into the solar system and the nuclides were rapidly homogenized in the solar nebula.

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