Introduction: The origins of short-lived radionuclides ($t_{1/2} \leq 100$ Myr) have important implications for the chronology and birth environment of the early Solar System. Of all shortest-lived ($t_{1/2} < 10$ Myr) radioactivities, $^{26}$Al ($t_{1/2} = 0.7$ Myr) and $^{41}$Ca ($t_{1/2} = 0.1$ Myr) are of special interest as they were found to have been correlated with one another in terms of the absence or presence in meteoritic refractory inclusions, such as Ca-Al-rich inclusions (CAIs) and hibonite (CaAl$_2$O$_4$) grains [1]. This correlation has been often used as an argument for co-injection of the two radioisotopes from a common stellar source after the solar nebula and a small fraction of high-temperature solids had already formed (“late injection”) [2]. It should however be noted that $^{26}$Al correlating with $^{41}$Ca has remained fairly qualitative (i.e., they either co-existed or were both absent in refractory inclusions). Although the study of [3] revealed a hint that there could have been concordant decay between $^{26}$Al and $^{41}$Ca, the small number (~3) of samples in which this observation was made and large analytical uncertainties associated with the initial $^{41}$Ca/$^{40}$Ca values (up to 40%, 2σ) precluded a definitive answer.

In this study, we attempt to reinvestigate the correlation between $^{26}$Al and $^{41}$Ca more quantitatively by analyzing the potassium isotope compositions of a suite of CAIs characterized by $^{26}$Al/$^{27}$Al ratios ranging from $5.2 \times 10^{-5}$ to $3 \times 10^{-5}$. The expected $^{41}$Ca/$^{40}$Ca values from this suite of samples should fall between $4.2 \times 10^{-9}$ and essentially 0. The result will provide a constraint on the astrophysical origin(s) of $^{26}$Al and $^{41}$Ca and the timing of injection. Here we report some preliminary results on the $^{26}$Al/$^{27}$Al and $^{41}$Ca/$^{40}$Ca values in a Leoville CAI.

Experimental: The sample studied here, Leoville 2-A, is a compact Type A CAI primarily composed of melilitite and spinel. The magnesium and potassium isotopic compositions of this CAI were measured with the UCLA ims-1270 ion microprobe. The analytical setting for high precision Mg isotopic measurements was similar to that described in [4]. A 20–25 nA $^{16}$O$^+$ primary beam ($\phi \sim 30 \mu m$) was used to generate sufficient Mg signals ($^{24}$Mg$^+ \geq 10^7$ cps) for accurate current measurements by Faraday cups. The mass resolution ($M/\Delta M$) was set at ~ 4200 to completely separate molecular and major doubly charged ion interferences ($^{40}$Ca$^{2+}$ and $^{48}$Ti$^{2+}$). The instrumental mass fractionation was characterized by using a suite of terrestrial standards (a synthetic fassaite glass, San Carlos olivine, Burma spinel and Madagascar hibonite) and corrected. The deviation of measured Mg isotopic ratio from the reference value is expressed with the modified delta notation: $\delta^{25,26}$Mg = $1000 \times \ln([^{25,26}$Mg/$^{24}$Mg]_s/[^{25,26}$Mg/$^{24}$Mg]_ref). By assuming an exponential mass fractionation law and an exponent of 0.514 [5], the radiogenic excesses of $^{26}$Mg were then calculated with the relationship: $\Delta^{26}$Mg* = $\delta^{25}$Mg – $\delta^{26}$Mg/0.514. The true $^{27}$Al/$^{26}$Mg$^+$ ratio of each spot was derived by applying the relatively sensitivity factor determined on a synthetic glass of fassaite composition.

The potassium isotope analysis of the CAI was performed by following the protocol established in [3]. The sample was sputtered by a 10 nA $^{16}$O$^+$ primary beam, and the secondary ions were collected by the axial electron multiplier in a peak-jumping mode, with the mass sequence of $38.7, \ 39$K, $^{41}$K$^+$, $^{40}$CaH$^+$, ($^{40}$Ca$^{45}$Ca$^+$), ($^{40}$Ca$^{47}$Al$^{16}$O$^+$)$^+$, $^{42}$Ca$^+$ and $^{43}$Ca$^+$. For sufficient separation between $^{40}$CaH$^+$ and $^{41}$K$^+$, mass resolution of ~7000 was set. Prior to each analysis, ~15–25 minute presputtering was applied to the sample surface to minimize contaminations. All isotopic ratios were calculated after summing up all the counts to avoid ratio bias [6].

Results: The result of Al-Mg isotopic measurements is shown in Fig 1. All the points were measured in melilitite, except one ($^{27}$Al/$^{24}$Mg – 3) being a mixture of spinel and melilitite. A free fit through all the data yields an $^{26}$Al/$^{27}$Al ratio of $(3.58 \pm 0.15) \times 10^{-5}$ (2σ), with an intercept ($\Delta^{26}$Mg*$_0$) = $(0.74 \pm 0.11)_\sigma$ (2σ) and a reduced $\chi^2 = 30.3$. Such a low $^{26}$Al/$^{27}$Al ratio with a large scatter along the isochron is likely to have resulted from partial isotopic resetting of the CAI, as some of the spots are consistent with having incorporated canonical $^{26}$Al/$^{27}$Al = 5.2x$10^{-5}$ (reference line).

The $^{41}$Ca-$^{41}$K isotope result is shown in Fig 2. Only in melilitite crystals could reasonably high $^{40}$Ca/$^{39}$K ratios be obtained, albeit all below 10$^6$. However, all the spots are devoid of radiogenic excesses of $^{41}$K that could be derived from the decay of $^{41}$Ca. The slope of a least-squares regression line indicates a $^{41}$Ca/$^{40}$Ca ratio of $(0.79 \pm 2.09) \times 10^{-9}$ (2σ, $\chi^2 = 3.3$) in the CAI. The intercept ($^{41}$K/$^{39}$K) being 0.0724±0.0003 is consistent with the terrestrial value (= 0.072).

Discussion and Conclusion: According to the Mg isotope result, it is clear that the Leoville 2-A CAI had undergone post-formation isotopic disturbance. However, it is unlikely that isotopic closure was comprised when $^{26}$Al/$^{27}$Al was at $(3.58 \pm 0.15) \times 10^{-5}$ because of the very large scatter of points about the isochron (reduced
\(\chi^2 = 30.3\). As there is no clear correlation between the spots that have been reset and their locations within the CAI, it is not possible to assess how many post-formation thermal events this CAI experienced (i.e., no well-defined isochrons can be designated to specific areas), thus the timing of last isotopic closure. However, one could still infer that this CAI must have formed early. This is indicated by the fact that 9 out of 19 spots are plotted along the \(^{26}\text{Al}/^{27}\text{Al} = 5.2 \times 10^{-5}\) reference line. The best fit through these 9 spots results in \(^{26}\text{Al}/^{27}\text{Al} = (4.95 \pm 0.21) \times 10^{-5}\) (2\(\sigma\)) and \(\Delta^{26}\text{Mg}^* = (0.33 \pm 0.1)\%\) (\(\chi^2 = 1.3\)). It is therefore reasonable to deduce that this CAI could have formed with \(^{26}\text{Al}/^{27}\text{Al} \sim 5 \times 10^{-5}\) and the large \(\Delta^{26}\text{Mg}^*\) scatter seen in the data resulted from isotopic disturbance that only reset part of the inclusion.

Under the assumption that Leoville 2-A formed while \(^{26}\text{Al}/^{27}\text{Al} \sim 5 \times 10^{-5}\) and that \(^{41}\text{Ca}\) and \(^{26}\text{Al}\) were derived from the same source, this CAI should have also incorporated \(^{41}\text{Ca}\) at the level of \(^{41}\text{Ca}/^{40}\text{Ca} \sim (3-4) \times 10^{-9}\). However, there are no resolvable excesses of \(^{41}\text{K}\) in the CAI that can be attributed to the decay of \(^{41}\text{Ca}\). This can also be understood in the context of isotopic resetting. The K self-diffusivity in melilite is marginally slower than that of Mg (6.74 \times 10^{-19} and 9.43 \times 10^{-19} m^2 s^{-1} at 1200 °C, respectively, [7]), so that the thermal event(s) that largely disturbed the Mg isotopes of the CAI could have easily erased \(^{41}\text{K}\) excesses. If the time at which the last perturbation took place was \(> 0.3\) Myr (consistent with the time difference calculated between \(^{26}\text{Al}/^{27}\text{Al} = 5 \times 10^{-5}\) and \(3.58 \times 10^{-5}\)) after CAI formation, \(^{41}\text{Ca}/^{40}\text{Ca}\) would be too low (< \(5 \times 10^{-10}\)) and \(^{41}\text{K}\) excesses would be essentially undetectable in this CAI (all spots have \(^{40}\text{Ca}/^{39}\text{K} < 10^6\)).

Isotopic analyses of the Leoville 2-A CAI have shown that the Mg and K isotopes have been largely reset in part of the inclusion and therefore does not provide constraints on the concordant decay between \(^{26}\text{Al}\) and \(^{41}\text{Ca}\). Having said that, it still falls within the expectation of \(^{41}\text{Ca}/^{40}\text{Ca}\) being 0 in a CAI with \(^{26}\text{Al}/^{27}\text{Al} \sim 3 \times 10^{-5}\). More CAIs that have well-characterized \(^{26}\text{Al}\) isochrons will be examined for the K isotopes in future studies.

References:


Fig 1. The Al-Mg plot for the Leoville 2-A CAI. The solid line is the best fit through all the data points, whereas the dashed line is the reference line representing \(^{26}\text{Al}/^{27}\text{Al} = 5.2 \times 10^{-5}\). All errors are 2\(\sigma\).

Fig 2. The \(^{41}\text{Ca}/^{41}\text{K}\) plot for the Leoville 2-A CAI. The solid line is the best fit through all the data points, whereas the dotted line stands for \(^{41}\text{Ca}/^{40}\text{Ca} = 4 \times 10^{-9}\). The dashed line indicates the terrestrial \(^{41}\text{K}/^{39}\text{K}\) value of 0.072. All errors are 2\(\sigma\).