

**A LATE EPISODE OF IRRADIATION IN THE EARLY SOLAR SYSTEM: EVIDENCE FROM EXTINCT  $^{36}\text{Cl}$  AND  $^{26}\text{Al}$  IN METEORITES.** Weibiao Hsu<sup>1</sup>, Yunbin Guan<sup>2</sup>, L. A. Leshin<sup>2</sup>, T. Ushikubo<sup>2</sup> and G. J. Wasserburg<sup>3</sup>, <sup>1</sup> Laboratory for Astrochemistry and Planetary Sciences, Purple Mountain Observatory, Nanjing, 210008, China: [wbxu@pmo.ac.cn](mailto:wbxu@pmo.ac.cn), <sup>2</sup> Dept. of Geological Sci., ASU, Tempe, AZ 85287, USA. <sup>3</sup> Lunatic Asylum, Div. of Geological and Planet. Sci., Caltech, Pasadena, CA 91125, USA.

**Introduction:** A recent report showed a linear correlation of  $^{36}\text{S}/^{34}\text{S}$  with  $^{35}\text{Cl}/^{34}\text{S}$  in late-formed halogen-rich phases of a Ningqiang CAI [1]. The inferred  $^{36}\text{Cl}/^{35}\text{Cl}$  is very high ( $\sim 5 \times 10^{-6}$ ) and exceeds the range for an AGB or SNe source [2]. In addition, the  $^{36}\text{Cl}$  is decoupled from  $^{26}\text{Al}$ . This imposes a serious challenge to our current understanding of how  $^{36}\text{Cl}$  and other short-lived nuclei were produced.

Here we present petrographic and isotopic (S, Mg, and O) studies of the Allende meteorite in an attempt to address the following questions: (a) Was  $^{36}\text{Cl}$  widespread present in the early solar system (ESS) and at what level? (b) How does  $^{36}\text{Cl}$  correlate with other short-lived nuclides in the ESS? (c) What is the most plausible source of  $^{36}\text{Cl}$ ? (d) Under what conditions did the secondary alteration of CAIs and chondrules occur?

**Petrography:** We studied one altered CAI, named Pink Angel [3], and a nearby porphyritic olivine chondrule from Allende (Fig. 1). Pink Angel is a  $\sim 2$  cm diameter CAI which has a large pinkish interior surrounded by a whitish rim. The interior is a powdery, porous aggregate of spinel, sodalite, grossular, and some anorthite. In the thin section studied, two porphyritic olivine chondrules lie close to Pink Angel (Fig. 1). The lower left chondrule also contains numerous sodalite grains, which occur interstitially between olivine grains. Sodalite does not appear in the lower right chondrule and in matrix. Pink Angel and two chondrules are close to each other within a distance of 500  $\mu\text{m}$ , and the two chondrules have the same texture. If sodalite formed within the Allende parent body by secondary processes, sodalite would also likely occur in both chondrules and matrix. The observation suggests that sodalite formed before the CAI and chondrule were accreted.

**Isotopes:** Sulfur, Mg, and O isotope compositions of sodalite and anorthite were analyzed with the CAMECA ims-6f ion probe at Arizona State University. The relative sensitivity factor of Cl/S (0.68) was determined on a terrestrial hauyne standard. Sodalite grains in Pink Angel and in the chondrule show clear  $^{36}\text{S}$  excesses (up to 760 ‰) that linearly correlate with  $^{35}\text{Cl}/^{34}\text{S}$  (Fig. 2). This is essentially the same as that previously observed in a Ningqiang CAI [1]. The inferred  $^{36}\text{Cl}/^{35}\text{Cl}$  ratio for the combined data set (Allende and Ningqiang) is  $(4.4 \pm 0.4) \times 10^{-6}$ .

Sodalite and anorthite of Pink Angel show large variations of  $^{27}\text{Al}/^{24}\text{Mg}$ , but contain no resolvable  $^{26}\text{Mg}$  excesses (Fig. 3). This is consistent with previous data for the same inclusion [4]. A few analyses of sodalite in the chondrule yield low Al/Mg ( $< 20$ ) and exhibit normal Mg. An upper limit of  $^{26}\text{Al}/^{27}\text{Al}$  for Pink Angel is set at  $1.7 \times 10^{-6}$ . If we consider the canonical value of  $^{26}\text{Al}/^{27}\text{Al}$   $5 \times 10^{-5}$  as the initial state, it implies the time interval for the sodalite formation  $\geq 3.5$  Myr. This is consistent with the conventional belief on the time interval ( $\sim 2$  Myr) between the formation of CAIs and chondrules [5].

In Pink Angel, spinel and diopside show typical  $^{16}\text{O}$ -enrichments with  $\delta^{17}\text{O}$  and  $\delta^{18}\text{O}$  of  $\sim -40$  ‰ and the secondary phases, sodalite, anorthite, grossular, and hedenburgite, are relatively  $^{16}\text{O}$ -depleted. The data plot along the CCAM line in the three-O-isotope diagram. Their oxygen isotope compositions are compatible with extensive alteration of an original CAI with  $\delta^{18}\text{O} = \delta^{17}\text{O} = -50$  ‰. The relict phases are spinel and diopside. This then points to an environment of alteration with “normal” planetary oxygen which may represent a nebular environment [6]. It appears that CAIs and chondrules interacted with halogen-rich material to form sodalite long after their original formation. This must have occurred in a common site for both the precursors to Pink Angel and the chondrule before they accreted into a planetary body.

**Discussions:**  $^{36}\text{Cl}$  ( $\tau = 0.43$  Myr) has a major branched decay to  $^{36}\text{Ar}$  (98.1%,  $\beta^-$ ) and a minor one to  $^{36}\text{S}$  (1.9%,  $\epsilon$  and  $\beta^+$ ) [7]. The  $^{36}\text{Ar}$  produced by the decay of  $^{36}\text{Cl}$  must have been almost completely lost after the formation of the sodalite. This must also have occurred without any major  $^{129}\text{Xe}$  loss from the same phase [4]. The sodalite family has an open aluminosilicate framework structure closely related to the zeolites. Sulfur is bound in the sodalite crystal structure where  $[\text{SO}_4]^{2-}$  substitutes for 2  $\text{Cl}^-$  to form solid solutions of  $([\text{SO}_4]^{2-}, \text{Cl}^-)$ -sodalite-nosean with a large miscibility gap [8]. Assuming  $^{36}\text{Cl}$  was the source of the  $^{36}\text{S}$  excesses, to interpret the observations concerning  $^{36}\text{Ar}$  and  $^{129}\text{Xe}$ , we must infer that there was extensive diffusive loss of  $^{36}\text{Ar}$  (presumably due to an episode of heating) that occurred after the sodalite formed and that the duration of this process was not much longer than 3 – 5 Myr. As the mean life of  $^{129}\text{I}$  is 23 Myr, there would not be much loss of  $^{129}\text{Xe}$ . The  $^{129}\text{Xe}$  disturbance inferred by [9] may be due to longer term diffusion or late metamorphism that would also cause

Ar loss. With regard to sulfur, it is structurally bound so the possibility of a diffusive loss would arguably be small. Diffusion of noble gases from the sodalite open structure is to be expected.

These data show that halogen-rich phases in Pink Angel and the chondrule contain large  $^{36}\text{S}$  excesses that correlate well with the chlorine concentration and strongly support the argument for *in situ* decay of  $^{36}\text{Cl}$ . However, these phases are devoid of any significant  $^{26}\text{Mg}$  excesses from the decay of  $^{26}\text{Al}$  [1,4]. If the  $^{36}\text{Cl}$  is decoupled from  $^{26}\text{Al}$ , then this requires a mechanism that produces abundant  $^{36}\text{Cl}$  and negligible  $^{26}\text{Al}$ . If the time interval for the sodalite formation is  $\sim 3.5$  Myr and the observed ratio represents the residue of a much higher  $^{36}\text{Cl}$  inventory coupled with  $^{26}\text{Al}$  production, it would require a source that is capable to produce  $^{36}\text{Cl}/^{35}\text{Cl} \sim 10^{-2}$  and that would presumably also have to be responsible for a substantial fraction of  $^{26}\text{Al}$ . In both of these considerations, we conclude that no stellar nucleosynthetic source can be responsible for the  $^{36}\text{Cl}$  inventory, even at the level of  $10^{-4}$  [2,10]. A SN source would only give  $^{36}\text{Cl}/^{35}\text{Cl} \sim 10^{-6}$  and an AGB source would give  $^{36}\text{Cl}/^{35}\text{Cl} 4.7 \times 10^{-7}$ , correlating with  $^{26}\text{Al}$  [2,10]. Accounting for the  $^{26}\text{Al}$  by a SNeII source would give extremely high  $^{53}\text{Mn}$  and  $^{60}\text{Fe}$  in the ESS which are not observed [2,10]. The  $^{36}\text{Cl}$  would exacerbate this situation as a SNeII source would only provide  $^{36}\text{Cl}/^{35}\text{Cl} \sim 10^{-6}$  in this case [2,10]. We conclude that the  $^{36}\text{Cl}$  case is similar to that for  $^{10}\text{Be}$  discovered by McKeegan *et al.* [11]. The  $^{10}\text{Be}$  can only be explained by spallation reactions.  $^{36}\text{Cl}$  can be produced abundantly in particle bombardment by an active early sun [12,13].

We provide strong evidence in support of the existence of  $^{36}\text{Cl}$  in CAIs as observed by Lin *et al.* [1] but also in chondrules as well.  $^{36}\text{Cl}$  is not correlated with  $^{26}\text{Al}$ . These results indicate that intense late irradiation processes occurred in the ESS.

**References:** [1] Lin Y. *et al.* (2005) *PNAS* **102**, 1306. [2] Wasserburg G. J. *et al.* (2006) *Nuclear Physics A*, in press. [3] Armstrong J. T. and Wasserburg G. J. (1981) *Lunar Planet. Sci.* **XII**, 25. [4] Villa I. M. *et al.* (1981) *Lunar Planet. Sci.* **XII**, 1115. [5] Huss G. R. *et al.* (2001) *MAPS* **36**, 975. [6] Clayton R. N. and Mayeda T. K. (1999) *GCA* **63**, 2089. [7] Endt P. M. (1990) *Nuclear Physics* **521**, 1. [8] Kotel'nikov A. R. *et al.* (2005) *Geochem. Intern.* **43**, 544. [9] Swindle T. D. *et al.* (1988) *GCA* **52**, 2215. [10] Busso M. *et al.* (1999) *ARA&A* **37**, 239. [11] McKeegan K. D. *et al.* (2000) *Science* **289**, 1334. [12] Goswami J. N. *et al.* (2001) *ApJ* **549**, 1151. [13] Leya I. *et al.* (2003) *ApJ* **594**, 605.

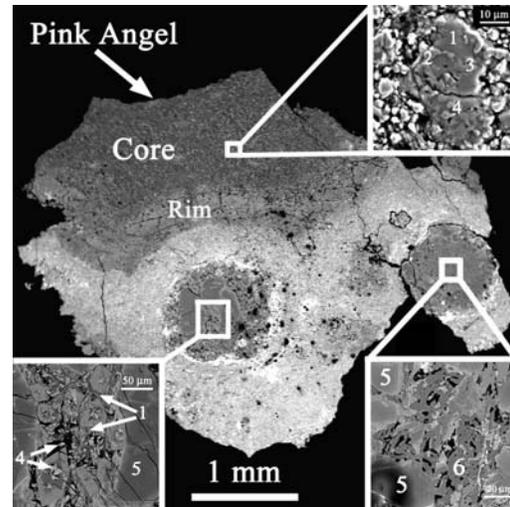


Fig. 1 BSE images of Pink Angel and two nearby porphyritic olivine chondrules in Allende. Mineral phases: 1. sodalite; 2. spinel; 3. grossular; 4. diopside; 5. olivine; 6. anorthite.

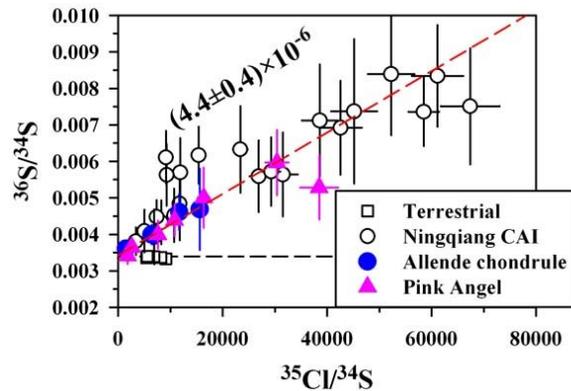


Fig. 2  $^{36}\text{S}/^{34}\text{S}$  vs.  $^{35}\text{Cl}/^{34}\text{S}$  in sodalite from Pink Angel and a chondrule of Allende and a CAI of Ningqiang [1]. Errors are  $2\sigma$ . The inferred  $^{36}\text{Cl}/^{35}\text{Cl}$  is  $(4.4 \pm 0.4) \times 10^{-6}$ .

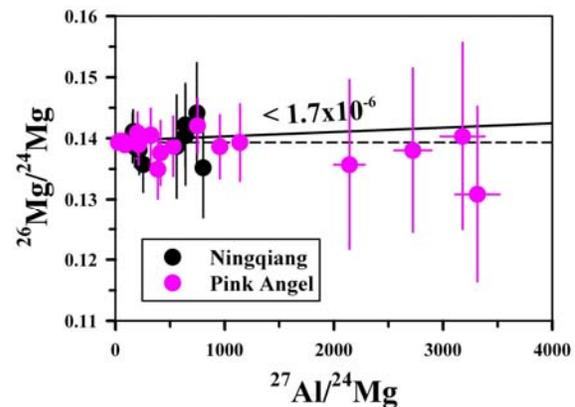


Fig. 3 Mg isotopic compositions and Al/Mg ratios of sodalite and anorthite in Pink Angel and a Ningqiang CAI [1]. Errors are  $2\sigma$ . An upper limit of  $^{26}\text{Al}/^{27}\text{Al}$  for Pink Angel is  $1.7 \times 10^{-6}$ .