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LATE-STAGE FORMATION OF SHORT-LIVED RADIONUCLIDES BY SOLAR ENERGETIC PARTICLE IRRADIATION IN THE EARLY SOLAR SYSTEM. B. Jacobsen¹, J. Matzel¹, I. D. Hutcheon¹, A. N. Krot², Q.–Z. Yin³, and K. Nagashima². ¹Lawrence Livermore National Laboratory, CA 94550, USA, ²University of Hawai^ci at Mānoa, Honolulu, HI 96822, USA, ³University of California, Davis, CA 95616, USA.

Introduction: The origin of short-lived ($\tau_{1/2} < 5$ Myr) and now extinct radionuclides (¹⁰Be, ²⁶Al, ³⁶Cl, ⁴¹Ca, ⁵³Mn, ⁶⁰Fe; hereafter SLRs) is fundamental to understanding the formation of the early solar system as they provide a unique source of information about the astrophysical environment in which the solar system formed, as well as a high-resolution chronology of early solar system events [1, 2]. The origin of SLRs in the early solar system, however, remains controversial [2] with two main classes of models proposed – injection of SLRs from a stellar source (e.g., supernova, asymptotic giant branch star or Wolf-Rayet star) and solar energetic particle (SEP) irradiation of dust and gas near the proto-Sun [2–7].

Excesses of ³⁶S correlated with ³⁵Cl/³⁴S ratios were previously reported in sodalite (Na₈Al₆Si₆O₂₄Cl₂), a secondary mineral in Ca-Al-rich Inclusions (CAIs) and chondrules from the Allende (CV) and Ninggiang (CV anomalous) carbonaceous chondrites [8-10]. The highest reported ³⁶Cl levels in sodalite ³⁶Cl/³⁵Cl ~ 5×10^{-6} [8, 9] are consistent with levels predicted for energetic particle irradiation of a reservoir with solar composition, but exceed by several orders of magnitude the values predicted for any stellar source [2]. Irradiation models predict that the production of ³⁶Cl by SEP irradiation cannot occur in isolation but would be coupled to the production of other SLRs such as ²⁶Al, ⁵³Mn, and ¹⁰Be [5–7]. These data underscore the importance of ³⁶Cl and its relationship to other SLRs for understanding the origin of SLRs in the early solar system.

Timing of ³⁶Cl production: Recently, we reported extremely large 36S excesses correlated with the re-³⁵Cl/³⁴S spective ratios in wadalite (Ca₆(Al,Si,Mg)₇O₁₆Cl₃), a Cl-rich secondary mineral, in Allende CAI AJEF [11]. The inferred ³⁶Cl/³⁵Cl ratio of wadalite of $(1.81\pm0.13)\times10^{-5}$ (Fig. 1a) represents the highest initial abundance of ³⁶Cl reported in any meteorite; four times greater than the highest ³⁶Cl/³⁵Cl initial ratio observed in sodalite in CAIs and chondrules [8, 9]. The absence of radiogenic ²⁶Mg in secondary grossular (Fig. 1b) contrasts with the wellconstrained primary mineral internal isochron in AJEF [12] yielding an initial ${}^{26}Al/{}^{27}Al$ ratio of $\sim 5 \times 10^{-5}$ and suggests that the wadalite-grossular paragenesis in AJEF formed >2.6 Myr after crystallization of the CAI. The well-defined ²⁶Al-²⁶Mg chronology, for both primary and secondary minerals within AJEF place

important constraints on the origin of ³⁶Cl. If ³⁶Cl was produced together with ²⁶Al, the late formation of wadalite inferred from the low (²⁶Al/²⁷Al)₀ ratio in cogenetic grossular, would require an unrealistically high initial 36 Cl/ 35 Cl ratio of >8.7×10⁻³ at the time the primary CAI mineral assemblage crystallized. This value is more than sixty times the maximum level that can be produced by SEP irradiation of gas and/or dust of solar composition [5, 6]. This suggests that production of 36 Cl by SEP irradiation must have occurred late, >2 Myr after the formation of the first solar system solids and provides the first conclusive evidence that ³⁶Cl found in secondary, low temperature minerals in CAIs and chondrules was produced in processes unrelated to those responsible for the SLRs (²⁶Al, ⁴¹Ca, ¹⁰Be) observed in primary, high temperature minerals in the same objects.



Fig. 1. (a) ³⁶Cl-³⁶S isochron diagram of wadalite from the Allende CAI AJEF (solid diamonds). The dashed line the inferred (³⁶Cl/³⁵Cl)₀ ratio for sodalite (open squares) from the Allende CAI Pink Angel [10]. Terrestrial wadalite is shown as open diamond. (b) ²⁶Al-²⁶Mg isochron diagram for grossular in the Allende CAI AJEF. The uncertainties in both panels (a) and (b) and inset are 2σ .

Production of SLRs during late-stage irradiation: Assuming *late-stage* irradiation of a reservoir with solar composition and a particle fluence sufficient to produce ³⁶Cl corresponding to the inferred ³⁶Cl/³⁵Cl ratio in wadalite ($\sim 2 \times 10^{-5}$), we use the approach of [5-7] to estimate relative abundances of co-produced ²⁶Al, ⁵³Mn and ¹⁰Be. The range in predicted abundances reflects different assumptions among the models regarding production cross-sections, ³He/H and ³He/⁴He ratios of SEP, hardness of the energy spectrum, and the relative importance of gradual to impulsive SEP events. The abundances of the three SLRs are compared against observed abundances in bulk meteorites [13, 14; Fig. 2]. In nearly all cases, the amounts of ²⁶Al and ⁵³Mn produced by SEP irradiation of a solar composition reservoir are significantly greater than the values observed in bulk meteorites, and an irradiation model accounting for ³⁶Cl, ²⁶Al and ⁵³Mn in a selfconsistent manner is difficult to achieve. Only in the case of an extremely hard SEP spectra, $p \ge 5$, where p is the spectral exponent of the SEP power law [6], is a self-consistent solution achievable (Fig. 2). If the initial ³⁶Cl abundance, however, was any higher than the assumed value $({}^{36}\text{Cl}/{}^{35}\text{Cl} > 2 \times 10^{-5})$, the problem will be exacerbated. The ³⁶Cl abundance assumed for the SLR abundance calculations is likely a lower limit for the amount produced by a late irradiation. Thus, ³⁶Cl production by late-stage SEP irradiation of a reservoir with solar composition would very likely overproduce both 26 Al and 53 Mn (Fig. 2).

Late-stage irradiation of a volatile-rich reservoir: Overproduction of ²⁶Al and ⁵³Mn can be avoided if the reservoir irradiated to produce ³⁶Cl was depleted in refractory elements (enriched in volatile elements) relative to solar composition due to CAI and chondrule formation. In particular, irradiation of a reservoir enriched in chlorine - a primary target element for SEP production of ³⁶Cl – would significantly enhance the production of 36 Cl relative to 26 Al and 53 Mn. During the lifetime of the protoplanetary disk, chlorine is present mainly as HCl gas and will condense as solid HCl hydrates (HCl•3H₂O) when temperatures fall below ~160 K and may adhere to mineral grains and water ice particles [15]. Solar energetic particle irradiation of either an HCl-rich gas or dust particles mantled by HCl hydrates would significantly enhance the production of ³⁶Cl relative to ²⁶Al and ⁵³Mn.



Fig. 2. Ratio of calculated to observed abundances of ¹⁰Be, ²⁶Al, and ⁵³Mn assuming a particle fluence sufficient to produce ³⁶Cl corresponding to (³⁶Cl/³⁵Cl) = 2×10^{-5} . The calculated ²⁶Al and ⁵³Mn abundances are normalized to the inferred upper limits of ²⁶Al and ⁵³Mn abundances for bulk meteorite [13, 14]. As there are no constraints on the ¹⁰Be abundance in bulk meteorites the calculated abundance for ¹⁰Be is normalized to the inferred solar initial value.

Since oxygen is the primary target element, ¹⁰Be will be co-produced with ³⁶Cl in any late SEP irradiation scenario. The most sensitive test for the late addition of ¹⁰Be is determination of boron-isotope abundances in late-forming secondary phases in CAIs or chondrules (e.g., wadalite or grossular). On the basis of the model presented here, we predict ¹⁰Be/⁹Be ratios exceeding 10⁻⁴ will be found.

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