Introduction: Chondrules and CAIs are the major high temperature chondritic components formed during transient heating events in the solar nebula ~4.57 billion years ago [1,2]. One of the major questions in meteoritics concerning the origin of CAIs and chondrules is their relative chronology. Most CAIs show large 26Mg excesses (26Mg*) corresponding to an initial 26Al/27Al ratio [(26Al/27Al)0] of ~5×10⁻⁵ [3,4], whereas most chondrules have smaller 26Mg* corresponding to (26Al/27Al)0 of ~1.2×10⁻⁵ [5-7]. Based on these observations and the assumption that 26Al was uniformly distributed in the solar nebula, it is generally inferred that CAIs formed at least 1-1.5 Myr before chondrules. This conclusion has recently been questioned based on new Pb [2] and Mg isotope measurements [4]. The 207Pb-206Pb ages of the Allende chondrules (4566.7±1.0 Ma) cannot be distinguished from those of the CV CAIs (4567.2±0.6 Ma) [1,2]. Bizzarro et al. [4] reported a range of (26Al/27Al)0 from (5.66±0.80) to (1.36±0.52)×10⁻⁵ in the bulk Allende chondrules and concluded that chondrule formation began contemporaneously with the formation of CAIs, and continued for at least 1.4 Myr. We note, however, that the (26Al/27Al)0 ratios inferred from bulk Mg isotope measurements of chondrules may date the time for the formation of chondrule precursor materials, not the time of chondrule melting; the latter requires Mg isotope measurements of mineral separates or individual mineral grains, which have not been done yet. In addition, spatial heterogeneity of 26Al in the solar nebula cannot be ruled out.

The relative chronology of CAI and chondrule formation can be resolved by studying compound objects composed of chondrule and CAI, because both constituents of such objects were affected by the same heating episode. Based on the presence of chondrule fragments in an igneous CAI having an 16O-poor melilite core and an 16O-rich diopside-anorthite mantle, [8] recently concluded that some chondrules formed contemporaneously with or even before CAIs, contrary to the previously accepted general view. This conclusion appears to be inconsistent with the common presence of relict CAIs inside chondrules, indicating that chondrules formed after CAIs [9-14]. Here, we report new detailed study of two chondrule-bearing, igneous CAIs in Allende (TS26 and ABC) that may resolve the apparent inconsistency.

Results: ABC is a coarse-grained, igneous, anorthite-rich (Type C) CAI fragment composed of lath-shaped anorthite (An₉₉) and Cr-poor Al-Ti-diopside, both poikilitically enclosing spinel grains, and interstitial, akermanite-rich (Åk₇₄₄) melilite [15]. Melilite is replaced by fine-grained grossular, monticellite, and wollastonite; anorthite is slightly corroded by sodalite and nepheline. A coarse fragment of forsteritic olivine (Fa₅) intergrown with low-Ca pyroxene (Fs₇₅Wo₃⁵) occurs in the CAI portion containing Cr-rich, Al-Ti-poor diopside. The olivine-pyroxyene fragment is corroded by the diopside and surrounded by a halo of high-Ca pyroxene (Fs₉₀.₃₈₅₃₉₅Wo₃⁵.₄₇). The abundances of Al₂O₃ and TiO₂ in Al-Ti-diopside decrease towards the forsterite grain; no such depletion is observed in Al-Ti-diopside occurring between anorthite laths. Olivine and low-Ca pyroxene have 16O-poor compositions; spinel and Al-Ti-diopside are moderately 16O-enriched, whereas Cr-spinel, Al-Ti-poor diopside, high-Ca pyroxene, anorthite, and melilite are 16O-depleted to various degrees (Fig. 1). The CAI shows a resolvable 26Mg* corresponding to (26Al/27Al)0 ratio of (4.7±1.4)×10⁻⁶ (Fig. 2).

TS26 is an irregularly-shaped Type C CAI showing a well-defined core-mantle structure, but lacking Wark-Lovering (WL) rim layers observed around most coarse-grained CAIs from Allende [16]. It has a coarse-grained core composed of lath-shaped anorthite (An₉₀) and sector-zoned Al-Ti-diopside, both poikilitically enclosing spinel grains, and interstitial akermanite-rich (Åk₇₂) melilite, sodalite, and ferrous olivine. Melilite is partly replaced by grossular, monticellite, and wollastonite. The finer-grained mantle, separated from the core by a discontinuous layer of Fe-Ni-sulfides, is composed of Al-Ti-diopside, lath-shaped anorthite, and abundant coarse grains of forsteritic olivine (Fa₈-₁₇) and low-Ca pyroxene (Fs₇₅Wo₃⁵.₄₂). The olivine and low-Ca pyroxene grains are corroded by the diopside and surrounded by haloes of high-Ca pyroxene (Fs₉₀.₃₈₅₃₉₅Wo₃⁵.₄₂). Olivine and high-Ca pyroxene have 16O-poor compositions; spinel is 16O-rich, whereas Al-Ti-diopside and anorthite are 16O-depleted to various degrees; the coarse Al-Ti-diopside grains in the core are less 16O-depleted compared to those in the finer-grained mantle (Fig. 1). The CAI anorthite, spinel, and Al-Ti-diopside show no resolvable 26Mg*; the inferred (26Al/27Al)0 ratio is <1.2×10⁻⁵ (Fig. 2).

Discussion: The corroded appearance of olivine-pyroxyene fragments in ABC and TS26 and the presence of high-Ca pyroxene haloes surrounding them suggest that these grains were present inside the host CAIs during final solidification and were partly dissolved in the
CAI melts. The relict origin of the olivine-pyroxene fragments is consistent with their dissolution textures and with the absence of olivine and low-Ca pyroxene in the crystallization sequence predicted for a melt having ABC or TS26-like bulk composition. The coarse-grained nature and $^{16}$O-poor compositions of relict forsteritic olivine associated with low-Ca pyroxene suggest that these grains are probably fragments of Fe-Mg chondrules. Although coarse olivine grains occasionally associated with low-Ca pyroxene are also found in AOAs and in forsterite-rich accretionary rims around CAIs, these olivines and low-Ca pyroxenes have $^{16}$O-rich compositions.

Most coarse-grained igneous CAIs in Allende show O-isotope heterogeneity: spinel and Al-Ti-diopside are typically $^{16}$O-rich ($\Delta^{17}$O ~ -20‰), whereas melilite and anorthite are $^{16}$O-depleted to various degrees ($\Delta^{17}$O up to 5‰) [17,18]. This heterogeneity has been attributed to O-isotope exchange between an $^{16}$O-poor nebular gas and initially uniformly $^{16}$O-rich CAIs during their incomplete melting [18]. TS26 and ABC show, in addition, significant $^{16}$O-depletion in Al-diopside; this depletion increases towards the relict chondrule fragments and the CAI peripheries. We infer that ABC and TS26 experienced incomplete O-isotope exchange during melting in an $^{16}$O-poor gas and dilution by $^{16}$O-poor relict chondrule materials, probably in the chondrule-forming region.

The observed differences in grain sizes between the core and the mantle of TS26 may indicate that melting was incomplete and followed by relatively fast cooling. The absence of WL-rim layers could be due to the inferred melting episode as well. The high abundance of relict chondrule-like material in the outer portion of TS26 suggests that there was a high abundance of dust in the region where melting occurred, consistent with dusty environment inferred for chondrule formation. The low ($^{26}$Al/$^{27}$Al)$_0$ ratios observed in ABC and TS26 may record their late-stage re-melting during incorporation of the chondrule fragments. We note, however, that because Allende experienced thermal metamorphism that may have disturbed the $^{26}$Al-$^{26}$Mg systematics in CAIs and chondrules, the exact age difference between the formation of CAIs ABC and TS26 and their re-melting should be considered with caution.

The proposed multistage formation history of ABC and TS26 is consistent with the extended (~ 2 Myr) formation time of several other igneous CAI from CV chondrites inferred from a range of the ($^{26}$Al/$^{27}$Al)$_0$ ratios within a single inclusion and petrographic observations. The late-stage melting and O-isotope exchange of ABC and TS26 is also consistent with the recently proposed model for the global evolution of the O-isotope composition of the inner solar nebula gas from $^{16}$O-rich to $^{16}$O-poor with time [19,20].