

# Clustering and Precipitation during Early-Stage Artificial Aging of Al–Si–Mg(–Cu) Foundry Alloys

## Note on isotope ranging in APT

The  $\text{Ti}^{++}$  and  $\text{Mg}^+$  mass peaks have partial overlap. We made a choice to range  $^{24}\text{Mg}^+ / ^{48}\text{Ti}^{++}$  as Mg, and  $^{25}\text{Mg}^+ / ^{50}\text{Ti}^{++}$  as Ti. This reproduces the expected bulk Ti concentration, which is assumed to be uniform throughout the material, and we include the most abundant Mg isotope for cluster identification. Due to the low amount of Ti present, the overlap is not expected to pose a problem.

## APT dataset parameters

Table S1: Acquisition parameters for all conditions investigated with APT.

Condition	Number of atoms ( $10^6$ )	Analyzed clusters	Laser pulse energy (pJ)
Mg0.45, 28 d RT	2.2	129	42
Mg0.45Cu0.5, 28 d RT	3.8	200	54
Mg0.45, 30 min AA	12.7	473	89
Mg0.45Cu0.5, 30 min AA	11.9	359	57
Mg0.45, 28 d RT + 30 min AA	11.5	385	62
Mg0.45Cu0.5, 28 d RT + 30 min AA	12.0	490	59

## Composition uncertainty in APT

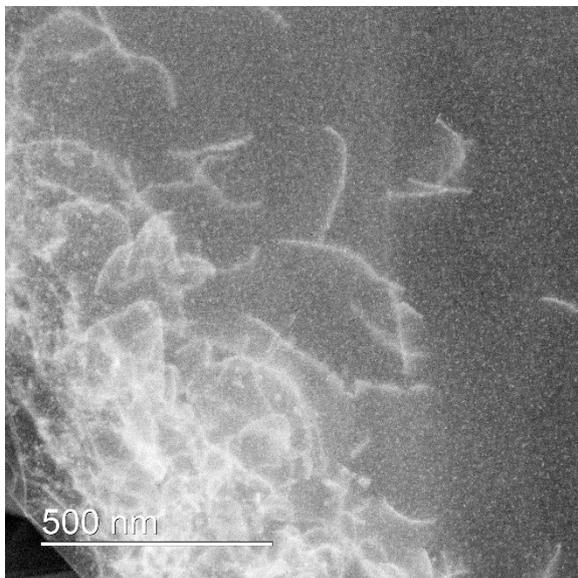
Tables 2 and 3 give compositional estimates from atoms detected in APT. The following sources of errors are deemed as significant:

- Sampling errors due to material inhomogeneities (random). Probably the largest error in the bulk composition. The magnitude is difficult to estimate without acquiring many datasets from different locations.
- Preferential evaporation (systematic). Depending on analysis conditions, Mg and Si may be over- or under-estimated compared to Al. Evaporation of atoms in-between laser pulses will make elements part of the background, which is subtracted from the isotope peak signals. With a reasonably low temperature (40 K) and high laser energy (20% equivalent pulse fraction), this is not thought to be a large problem [1], but it is the greatest systematic error, possibly shifting the composition of all acquired datasets.

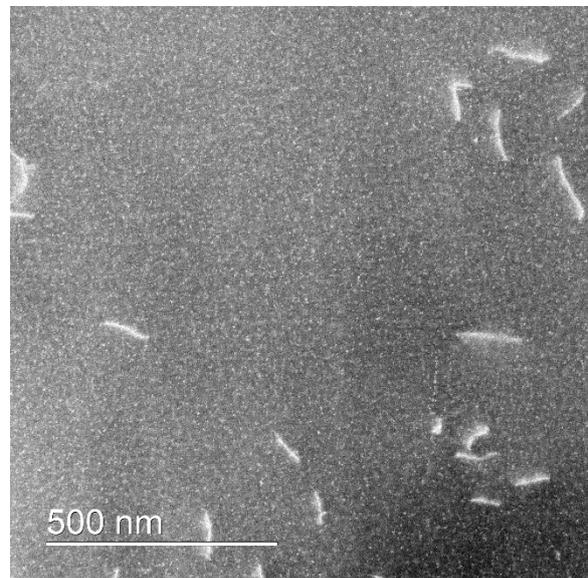
- Atom selection error (random). Due to limited detector efficiency (70–80%), some atoms from each species will not be detected, leading to a binomial distribution for the measured composition. For Mg and Si content in bulk volumes, this leads to a relative error of <1%, and for the cluster compositions the error is about 1/10 of the standard deviations of the cluster populations.

## TEM images of area near eutectic Si particle

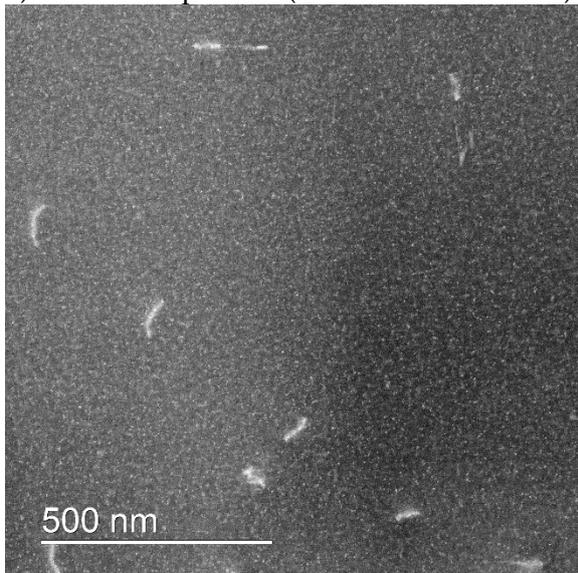
Figure S1 shows LAADF-STEM images from the FIB-prepared sample of Mg<sub>0.45</sub>Cu<sub>0.5</sub>, at low magnification and different distances from an eutectic Si particle. There is no long-range variation in precipitate microstructure. The many dislocations (bright lines) close to the Si particle probably originated from sample preparation, as the sample may bend during ion thinning, and the hard Si particle has caused deformations in the softer aluminium matrix.



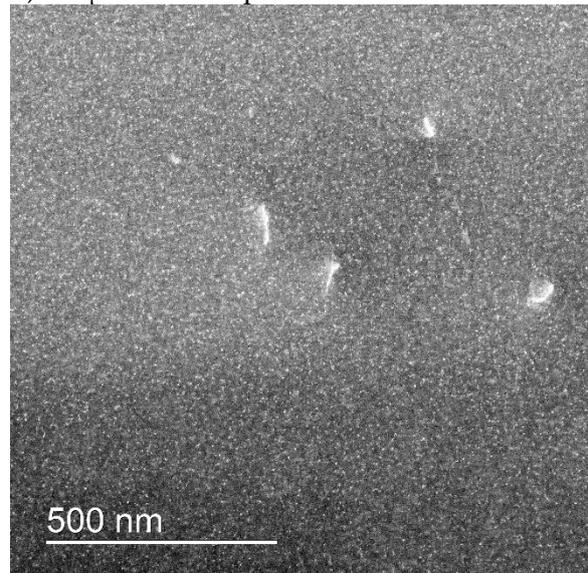
a) Close to Si particle (in bottom left corner)



b) 1.5 μm from Si particle



c) 3 μm from Si particle



d) 10 μm from Si particle

Figure S1: LAADF-STEM images of microstructure at different distances from eutectic Si particle.

- [1] C. Hatzoglou, S. Rouland, B. Radiguet, A. Etienne, G.D. Costa, X. Sauvage, P. Pareige and F. Vurpillot, *Preferential Evaporation in Atom Probe Tomography: An Analytical Approach*, *Microscopy and Microanalysis*, **26**, 689-698 (2020).