

Supplementary Materials – Analytical Run Conditions

Analytical run conditions for both specimens are provided in the table below, after Blum et al. (2018).

Supplementary Table 1: Atom Probe Tomography Data Acquisition Settings and Data Summary

Specimen	M1	M2
Instrument model	LEAP 4000X HR	LEAP 4000X HR
Laser wavelength (nm)	355	355
Laser pulse energy (pJ)	300	300
Pulse frequency (kHz)	200	200
Evaporation control	Detection rate	Detection rate
Target detection rate (ions/pulse)	0.01	0.01
Nominal flight path (mm)	382	382
Set point temperature (K)	60	60
Sample temperature (K)	69.2	69.2
Chamber pressure (Torr)	3.1×10^{-11}	3.3×10^{-11}
Data Summary		
LAS Root version	15.41.342l	15.41.342l
CAMECAROOT Version	18.46.452d	18.46.452d
Analysis software	IVAS 3.8.4sp1	IVAS 3.8.4sp1
Total ions (millions):	56.9	100.0
Single (%)	70.6	71.0
Multiple (%)	29.2	28.8
Partial (%)	0.2	0.2
Reconstructed ions (millions)	55.2	98.2
(complete dataset):		
Ranged (%)	89.2	83.2
Unranged (%)	10.8	16.8
Volt./bowl corr. peak (Da)	16	16

Mass calibration (peaks/interp.)	8/Linear	10/Linear
$(M/\Delta M)$ for $^{16}\text{O}^+$	951	979
Time-dependent background (ppm/ns)	16.8	17.1
Reconstruction		
Final specimen state	Fractured	Intact
Pre-/post-analysis imaging	SEM/SEM	SEM/SEM
Radius evolution model	Voltage evolution	Voltage evolution
Field factor (k)	3.3	3.3
Image compression factor	1.65	1.65
Assumed E-field (V/nm)	28	28
Detector Efficiency (%)	36	36
Average atomic volume (nm^3)	0.01076	0.01076
V_{initial} (V)	4866	5456
V_{final} (V)	8135	10252

ΔM is full width at half maximum

Pb isotopic ratio calculations and corrections

The $^{207}\text{Pb}/^{206}\text{Pb}$ ratio was determined from the Pb^{++} ion peaks located in the time-of-flight mass spectrum at 103 and 103.5 Da. Equivalent proportions of each peak were measured by selecting the counts in a central 0.1 Da mass range. Count values were then corrected for the local background noise using an estimate of the background between 100.5 and 102.5 Da, where there are assumed to be no mass peaks. A further correction was applied to the ^{207}Pb signal to account for an overlap between the 103.5 Da peak ($^{207}\text{Pb}^{++}$) and the extended leading-edge of the 104 Da peak ($^{28}\text{Si}_2^{16}\text{O}_3^{++}$). The degree of this overlap was estimated by examining the spectrum from the entire dataset (M2), in which the contribution of leading-edge counts in the 103.5 Da peak range was found to be 20% of those found centered in the 104 Da peak. This correction further reduced the ^{207}Pb counts, resulting in a lower $^{207}\text{Pb}/^{206}\text{Pb}$ ratio than would otherwise be measured.