**Electronic Supplementary Information**

# Aluminium metal–insulator–metal structure fabricated by the bottom-up approach

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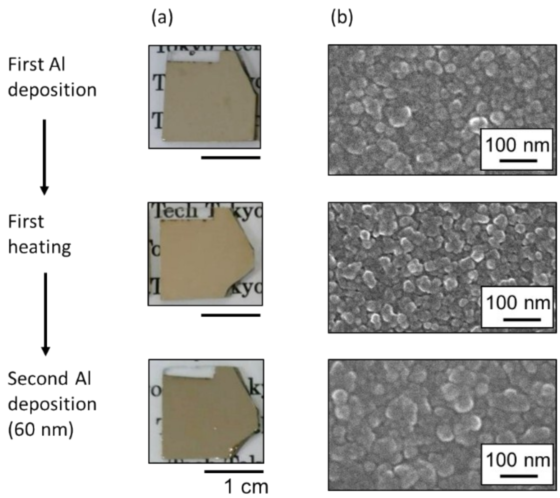


**Fig. S1** Visual appearance and SEM images of a sample before the second heating. (a) Appearance of the sample in each process. The film thickness of the second Al layer was 60 nm, as measured using a film thickness monitor. (b) SEM images showing the appearance of Al particles.

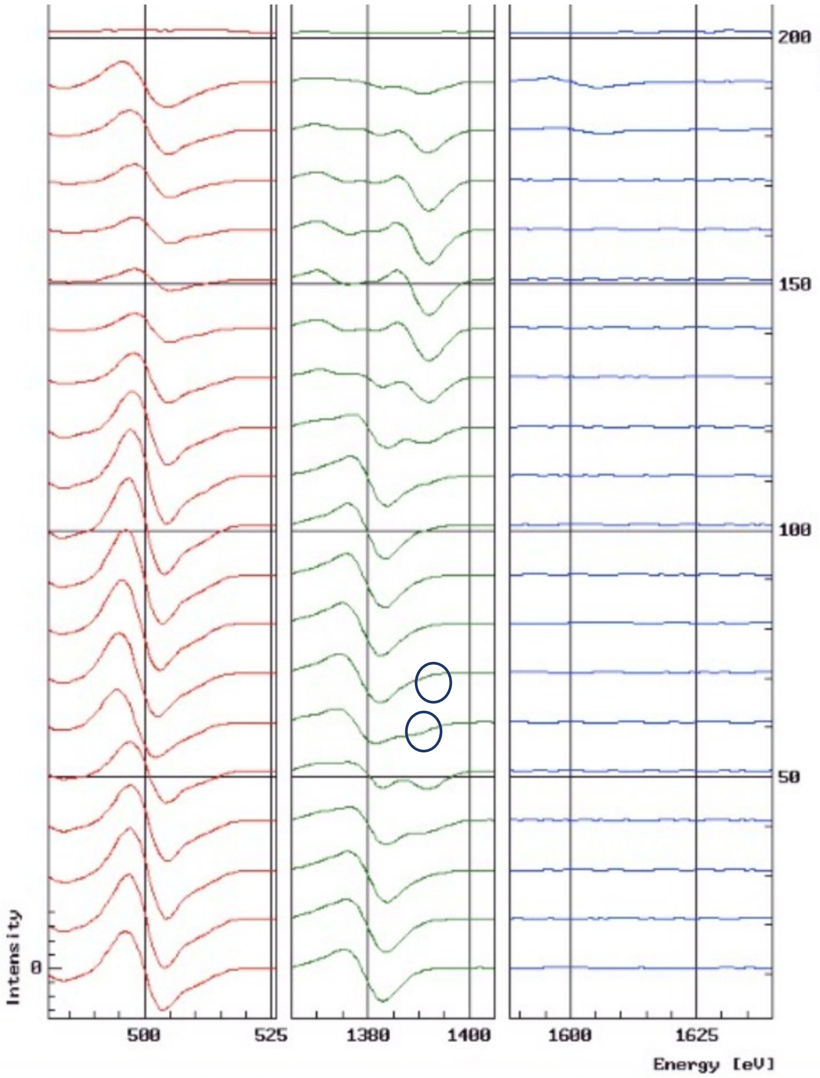


**Fig. S2** The results of Auger electron spectroscopic analysis of sample Al60-600. The larger the number of analysis cycles, the deeper the sample data was detected. The dip near 1383 eV is due to $Al_2O_3$, and the dip near 1392 eV is due to Al. Both dips appear approximately 50 and 120 cycles.

**Supporting Information 3. How to set the simulation model?**

The oxidation of Al nanoparticles proceeds from the top surface in contact with the atmosphere and from grain boundaries. However, the bottom surface of the Al second layer was less oxidized because there was no oxygen. Thus, the depth of Al nanoparticles was determined and subsequently confirmed by the cross-sectional EDX mapping. Therefore, we speculate that a bowl or deep dish-shaped metal shape is preferable; however, for simplicity of calculation, we here selected a spheroid.

The size of the calculation model was obtained by performing the following processing on particles observed by EDX mapping. Particles with an approximate elliptical area of 300 $nm^2$ or less were excluded because the peak of the scattering cross section was small and the contribution to color was relatively small. We selected six particles.

The long axis and the short axis for the obtained six particles were measured. Because of large size variations, we set two models: large spheroids, where the major and minor axes are the averages for three large particles, and small spheroids, where the major and minor axes are the averages for three small particles.

In the model, a sufficient thickness with no light transmission was used for the first Al layer. As the thickness from the lower part of the metal particle to the first Al film, an average value obtained by randomly taking 16 points was used. As the thickness of the $Al_2O_3$ layer on the upper side of the particles, the distance from the top of the particle was measured at 10 random points and the average was used. For the gap between nanoparticles, the horizontal distance between the edges of particles was taken at four points and the average value was used.