

1 SUPPLEMENTARY MATERIAL CAPTIONS

2 TABLE A1. Spectral Library

3 Figure A. X-ray diffraction patterns of synthetic allophanic products. The pattern from the Si/Al
 4 = 0.58 allophane sample was not included because it overlaps that of the Si/Al = 0.44 allophane
 5 sample. Each pattern displays broad absorptions, indicating samples are X-ray amorphous.
 6 Broad reflections at 0.35, 0.22, and 0.14 nm confirm the structure of allophane, while an intense
 7 reflection at 0.37 and weaker reflections at 0.22 and 0.14 nm are consistent with a high-Si
 8 aluminosilicate gel.

9 Figure B. Bright field transmission electron microscope images of synthetic allophanic products
 10 on holey carbon grids. Samples are composed of gel-like particles or aggregates of roughly
 11 spherical particles ~5 nm in diameter, consistent with the morphology of natural allophane. a, b)
 12 Aluminosilicate gel (Si/Al = 5.6) is composed of aggregates of spherical particles ~5 nm in
 13 diameter and has a mottled appearance compared to the carbon grid. d) Al-rich product (Si/Al =
 14 0.44) is composed of irregularly shaped gel-like particles. f) Products with intermediate Si/Al
 15 ratios (Si/Al = 0.92) are composed of irregularly shaped gel-like particles as in Al-rich products,
 16 but on closer inspection, reveal a spheroidal morphology (g). Electron diffraction patterns of all
 17 products show they are amorphous (c,e,h).

Plagioclase group Albite WAR-0244 Oligoclase WAR-5804 Andesine BUR-240 Labradorite WAR-4524 Bytownite WAR-1384 Anorthite BUR-340 Shocked Anorthite 22.6 GPa Shocked Anorthite 56.3 GPa Low-calcium pyroxene group Bronzite NMNH-93527 Enstatite HS-9.4B Bronzite BUR-1920 Avg. Lindsley pigeonite High-calcium pyroxene group Diopside WAR-6474 Augite NMNH-9780 Augite NMNH-122302 Hedenbergite (manganoan) DSM-HED01 Olivine group Forsterite BUR-3720A Fayalite WAR-RGFAY01 KI 3362 Fo60 KI 3115 Fo68 KI 3373 Fo35 KI 3008 Fo10	High-silica group Biotite BUR-840 Muscovite WAR-6474 Serpentine HS-8.4B Illite IMt-1 <0.2 microns Montmorillonite (Ca) STx-1 Saponite <0.2 microns K-rich volcanic glass SiO ₂ glass Opal-A Aluminous opal Antigorite NMNH-47108 Crystalline heulandite Crystalline stilbite Allophane group Allophane, molar Si/Al ratio 0.44 Allophane, molar Si/Al ratio 0.58 Allophane, molar Si/Al ratio 0.92 Allophanic gel, molar Si/Al ratio 5.6 Other group Quartz BUR-4120 Microcline BUR-3460 Orthoclase WAR-RGSAN01 Magnesiohastingsite HS-115.4B Actinolite HS-116.4B Magnesiohornblende WAR-0354 Hematite BUR-2600 Calcite C40 Dolomite C20 Anhydrite S9 Gypsum S6
---	--



