Effect of maghemization on the magnetic properties of non-stoichiometric pseudo-single-domain magnetite particles

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During formation, magnetic minerals record the direction and intensity of the Earth's magnetic field. Paleomagnetists use this information to investigate, for example, past tectonic plate motion and geodynamo evolution. However, subsequent to formation the constituent magnetic minerals are commonly exposed to a range of weathering conditions and environments. One of the most common weathering processes is maghemization, which is the oxidation of magnetite (Fe_3O_4) at ambient temperatures, i.e., the slow oxidation of Fe_3O_4 to maghemite (γ-Fe_2O_3), and is known to alter the original remanent magnetization.

Of the constituent magnetic minerals, particles in the single domain (SD) grain size range (< 100 nm) are regarded as ideal paleomagnetic recorders because of their strong remanence and high magnetic stability, with potential relaxation times greater than that of the age of the Universe. However, magnetic signals from rocks are often dominated by small grains with non-uniform magnetization that exhibit magnetic recording fidelities similar to those of SD grains (termed pseudo-SD (PSD)).

In this context, the effect of maghemization on the magnetic properties of Fe_3O_4 grains in the PSD size range is investigated as a function of annealing temperature. X-ray diffraction and transmission electron microscopy confirms the precursor grains as Fe_3O_4 ranging from ~ 150 nm to ~ 250 nm in diameter, whilst Mössbauer spectrometry suggests the grains are initially near-stoichiometric. The Fe_3O_4 grains are heated to increasing reaction temperatures of 120 – 220 °C to investigate their oxidation to γ-Fe_2O_3. High-angle annular dark field imaging and localized electron energy-loss spectroscopy reveals slightly oxidized Fe_3O_4 grains, heated to 140 °C, exhibit higher oxygen content at the surface. Off-axis electron holography allows for construction of magnetic induction maps of individual Fe_3O_4 and γ-Fe_2O_3 grains, revealing their PSD (vortex) nature, which is supported by magnetic hysteresis measurements, including first-order reversal curve analysis. The coercivity of the grains is shown to increase with reaction temperature up to 180 °C, but subsequently decreases after heating above 200 °C; this magnetic behavior is attributed to the growth of a γ-Fe_2O_3 shell with magnetic properties distinct from the Fe_3O_4 core.