Quantitative methods to analyse superparamagnetic and single domain mixtures of magnetite

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Abstract: Mixtures of superparamagnetic (SP) and single domain (SD) magnetite frequently occur in natural material, which are often subjected to environmental magnetic studies, e.g. soils, dust or sediments. Failure to identify and quantify the potential presence of SP particles cannot only undermine conclusions drawn from otherwise straightforward approaches, but also overlook physical and chemical processes involved. Although this is problem is commonly recognized, few methods exist that can be invoked cost-effectively in order to determine the potential presence of SP particles. Obtaining an estimate of the relative proportion of SP to SD particle sizes in a material can be useful in evaluating environmental conditions in natural materials, or in understanding the homogeneity of particle size and the degree of agglomeration in synthesized particles. This study is concerned with evaluating methods that can aid in estimating the relative proportions of the two size fractions. The methods include frequency-dependent susceptibility, hysteresis properties, analysis of first-order reversal curves (FORC), and temperature dependent saturation remanent magnetization (M_{rs}). The boundary between SP and SD behaviour is considered normally to be 30 nm for magnetite (and greigite), assuming a measurement time of 100 s. If measurement acquisition is faster, then this boundary will be lower, e.g., 24.5 nm when averaging over 100 ms. Frequency dependent susceptibility is one of the most common methods used to identify SP particles in a material. The ability to detect SP particles will be dependent on the frequencies that can be applied. The AGICO MFK1 susceptibility bridge has three applied frequencies, 975 Hz, 3.904 kHz and 15.616 kHz. The highest frequency will block in SP particles with a diameter down to 17 nm (Hrouda and Jezek, 2014). Using a differential analysis, it is in theory possible to obtain an estimate of the SP grain size fraction (Kodama, 2013). Magnetization ratio, M_{rs}/M_s, and coercivity ratio, H_{cr}/H_c, which are obtained from hysteresis measurements and DC backfield demagnetization, can also provide information on the relative proportion of the SP and SD grain size fractions, by using Langevin theory (Dunlop and Carter Stiglitz, 2006). FORC analysis can provide information on the reversible and irreversible particle size fractions, thus providing a

relative estimate of the two particle sizes (Kumari, 2014). At low temperature the magnetite grains undergo blocking and acquire a remanent magnetization. Theoretically the change in the blocked volume provides an estimate of the SP fraction, under the assumption that the particles are non-interacting. A combination of methods is tested on a synthetic mixture of magnetotactic bacteria that contain a chain of SD magnetosomes with the incremental addition of a strain of bacteria whose magnetosomes are SP in size. The particle size distribution of the SP magnetosomes is known from TEM images. Hysteresis properties and FORC analysis both show a progressive change related to the increasing SP content. A second example examines lacustrine sediments from Southern Norway, for which no information is available on the actual particles size of the magnetite in the sediments. Hysteresis parameters and FORC analysis indicate a varying amount of SP grain size in the lacustrine sediments, which will be compared to the change in M_{rs} from room temperature and 77 K. Frequency dependent susceptibility was measured on an AGICO MFK1 and found to be less effective in the above cases due to the limited frequency range.

Keywords: superparamagnetic, single domain, particle size, magnetite, sediments

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