

## Short Communication

## Magnetically-responsive electrophoretic silica organosols



Gregory N. Smith, Julian Eastoe\*

School of Chemistry, University of Bristol, Cantock's Close, Bristol BS8 1TS, UK

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## ABSTRACT

**Hypothesis:** Silica nanoparticles can be dispersed in organic solvents (organosols) using surfactants, such as didodecyltrimethylammonium bromide (DDAB). DDAB analogs prepared with lanthanide tetrahalide counterions, either a high-magnetic moment ion ( $\text{HoCl}_3\text{Br}$ , DDAH) or low-magnetic moment one ( $\text{NdCl}_3\text{Br}$ , DDAN), are expected to produce charged particles, but only DDAH-stabilized dispersions are expected to be magnetically responsive.

**Experiments:** Phase-analysis light scattering (PALS) measurements have been performed to determine the charge on DDAH- and DDAN-stabilized organosols. Magnetic sedimentation experiments have been performed to determine whether or not the silica dispersions are magnetically responsive. Sedimentation was monitored both by visual observations and UV-vis spectroscopy.

**Findings:** Both DDAH and DDAN organosols are negatively charged, the same as DDAB-stabilized nanoparticles. The DDAH-stabilized nanoparticles are found to respond to a magnetic field, whereas the DDAN-stabilized nanoparticles do not. This opens up possibilities for creating nanoparticle dispersions in nonpolar solvents which can be tailored to respond to desired external stimuli.

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## 1. Introduction

The term “organosol” is used to describe a classic system in colloid science, consisting of inorganic nanoparticles stabilized in a nonaqueous, organic solvent [1]. Organosols are of general interest, for example, as inorganic nanoparticles as acid-neutralizers in petrochemical fluids [2]. Furthermore, mineral oxides can be charged by surfactants in nonpolar solvents [3–8]. Of the many possible inorganic nanoparticles, silica has by far been the most studied [7,9–15]. Despite the extensive studies of charged nanoparticles in organic solvents, the detailed origin of the production of charge on the particle surfaces is still not fully understood [16,17].

The use of charged nanoparticles has been motivated in recent years by their applications in electrophoretic displays, which are used to form the electronic paper screens used in e-Reader devices [18]. This initial report has been cited over 500 times and receives approximately 50 citations per year. Additionally, there have been approximately 2000 US and EU patents registered for electrophoretic display particles. Many of the proposed technologies for next-generation electronic paper, with lower power consumption and a full color gamut, require control of the charge of either the particles or the fluid [19]. While previous electronic paper technologies have relied on electrophoresis, a

recent idea is to develop dual-functionality, magnetic–electric electronic paper [20].

Recently, a simple method to prepare surfactant-stabilized silica organosols through phase transfer from aqueous to organic solvents has been demonstrated [21]. Of the particle–surfactant combinations presented, didodecyltrimethylammonium bromide (DDAB) stabilized silica particles in toluene–heptane solvent mixtures are most promising for generating electrophoretic and magnetophoretic dual-functionality. Quaternary ammonium surfactants with lanthanide tetrahalide anions have previously been prepared and shown to exhibit magnetic responsiveness [22–24]. In this study, the  $\text{HoCl}_3\text{Br}$  analog of DDAB (DDAH) was chosen due to the high magnetic moment of  $\text{Ho}^{3+}$  ( $10.6 \mu_B$ ), and the  $\text{NdCl}_3\text{Br}$  analog (DDAN) was prepared as a non-magnetic lanthanide control due to the much lower magnetic moment of  $\text{Nd}^{3+}$  ( $3.58 \mu_B$ ) [25]. Both surfactants result in charged particles with larger magnitude electrophoretic mobilities than DDAB-stabilized particles; only DDAH, however, shows a response to magnetic fields. This demonstrates that it is possible to design small-molecule dispersants which can impart both magnetic and electrophoretic mobility to mineral oxide nanoparticles.

## 2. Materials and methods

DDAH and DDAN were synthesized according to literature procedures for preparing magnetic cationic surfactants [23]. (The

\* Corresponding author. Fax: +44 (0)117 92 77985.

E-mail address: julian.eastoe@bristol.ac.uk (J. Eastoe).

details and analysis of the surfactants are shown in the [Supporting Information](#).) The silica organosols were prepared in toluene-heptane (70:30 w/w) using a drying method previously described [21]. The transfer efficiency (mass of dispersed surfactant and silica over the whole solid mass) was determined after gentle centrifugation for 300 s at 3000 rpm (DDAB 97%, DDAH 59%, and DDAN 75%). The DDAH and DDAN organosols were not as stable as those with DDAB, but sufficient mass was transferred to enable studies of the response of the particles to electric and magnetic fields. (Further details of the preparation procedures are provided in the [Supporting Information](#).)

### 3. Results and discussion

The electrophoretic mobilities ( $\mu$ ) of DDAH- and DDAN-stabilized dispersions were measured using phase-analysis light scattering, and the results of these measurements are shown in [Table 1](#). (Experimental details are provided in the [Supporting Information](#).) As has previously been determined for DDAB-stabilized

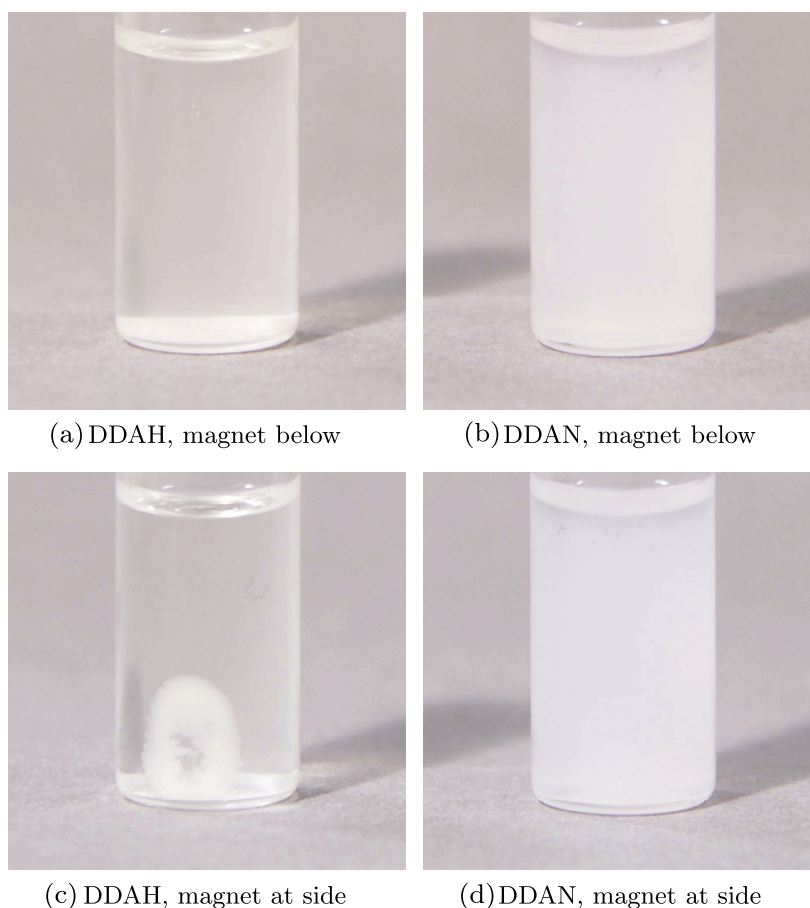
silica organosols [21], both of the lanthanide surfactants also induce a negative charge on the silica particles. Additionally, the magnitude of  $\mu$  is greater for both lanthanide surfactants, indicating that the two are more effective charge control agents than DDAB. The DDAH-stabilized dispersions are more highly charged than the DDAN-stabilized ones, but the value of  $\mu$  when using DDAN is more similar to DDAH than DDAB. In terms of the electrophoretic response, there is little difference between the two lanthanide tetrahalide surfactants.

This demonstrates that the particles respond to the application of electric fields. In addition to this, the response of the dispersions to magnetic fields has been studied to determine the effect of the Nd and Ho counterions. NdFeB rod magnets were used to apply to a magnetic field to the sample. (Details of the magnets are provided in the [Supporting Information](#).) In the absence of a magnetic field, the particles do not sediment significantly over 1 h. More occurs in the DDAH-stabilized organosol, which can be expected due to the decreased stability reflected by the lower transfer efficiency. The organosols were redispersed after all sedimentation experiments (magnetic or non-magnetic) using vortex mixing. The stability and performance of the dispersions were similar regardless of the number of times the particles had been sedimented and redispersed. (Through the course of this study, particles were cycled through sedimentation and redispersion approximately 10 times.)

In the first set of magnetic sedimentation experiments, the magnets were placed at the bottom of the vials, either under the bottom of the vial or set to the side. As can be seen in [Fig. 1](#), there

**Table 1**  
Electrophoretic mobility ( $\mu$ ) for silica organosols.

Dispersant	$\mu/(10^{-10} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1})$
DDAB	$-5.9 \pm 0.3$
DDAH	$-17.8 \pm 0.7$
DDAN	$-12.1 \pm 0.6$



**Fig. 1.** Photographs of silica organosols after 1 h sedimentation in the presence of a magnetic field (either below the vial or set to the side). The photographs were taken immediately after the magnets were removed. The magnetic field causes no visible difference to the DDAN-stabilized dispersion. For the DDAH-stabilized dispersion, however, enhanced sedimentation is observed, causing either near-total sedimentation or a ring of sediment to form on the wall of the vial.

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