

Diffusiophoretic enhancement of mass transfer by nanofluids

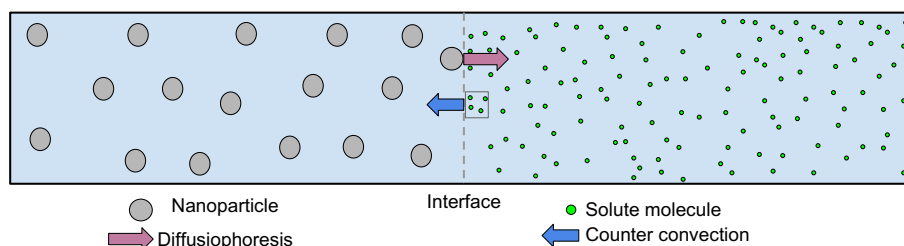
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HIGHLIGHTS

- Mass transfer enhancement by nanofluids is demonstrated in a capillary tube.
- The enhanced motion of the solute is shown to be due to diffusiophoresis.
- Diffusiophoresis of nanoparticles induces a counter-convective motion of the dye.
- Earlier observations of mass transfer effects in nanofluids explained.

GRAPHICAL ABSTRACT



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ABSTRACT

Observations of an enhanced mass transfer and an irregular mixing when a small drop of dye is gently placed on a nanofluid dispersion have led to several propositions for the underlying cause, but none of them have been conclusively established. Here, we reproduce the enhancement phenomenon within a glass capillary containing fluorescein di-sodium dye solution on one side and alumina nanoparticle suspension on the other, avoiding the inertial interference present earlier. The enhancement is explained by the convective motion (and not a diffusion) of the dye solution counter to the direction of the diffusiophoretic motion of the nanoparticles towards a higher concentration of the dye. The initial velocity of the dye front in the capillary and the dye-drop experiment agree quantitatively with independent theoretical estimates from the diffusiophoretic velocity of alumina nanoparticles. With a suitably chosen nanofluid, it should now be possible to effect a desired level of enhancement (or suppression) of mass transfer of any solute. In micro-environments requiring fast mixing, such as in nanofabrication, lab-on-chip, drug-delivery, and chemotaxis, the effects can be substantial.

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1. Introduction

The presence of a small fraction of nanoparticles as a suspension in a fluid (also referred to as nanofluids) immensely influences various phenomenon like diffusion (Krishnamurthy et al., 2006; Fang et al., 2009; Veilleux and Coulombe, 2010), absorption (Komati and Suresh, 2008, 2010; Pang et al., 2012; Olle et al., 2006; Esmaili-Faraj and Nasr Esfahany, 2016; Moraveji et al., 2013; Rahmatmand et al., 2016), extraction (Mirzazadeh Ghanadi et al., 2015; Saien and Bamdadi, 2012; Ashrafmansouri et al., 2016), in radiation (Waheed et al., 2015), electric conductivity

(Liu et al., 2016), evaporation (Javed et al., 2013) and reaction kinetics of chemical processes. Mass transfer studies in dye-diffusion by Krishnamurthy et al., 2006 and others (Fang et al., 2009; Komati and Suresh, 2010; Veilleux and Coulombe, 2010) have demonstrated, through dramatic visual effects, that the presence of nanoparticles increases the effective diffusivity of a solute up to 48 folds. However, when carried out in other system configurations, there are some cases where no enhancement (Ozturk et al., 2010; Subba-Rao et al., 2011; Feng and Johnson, 2012) and even a marginal lowering of the diffusivity (Turanov and Tolmachev, 2009; Gerardi et al., 2009) is observed. Various possibilities have been advanced in an attempt to explain these intriguing observations: Brownian motion induced micro-convection (Fang et al., 2009; Krishnamurthy et al., 2006), interfacial complex-

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ation (Ozturk et al., 2010), and a dispersion model (Veilleux and Coulombe, 2011). We can discount Brownian motion induced micro-convection or dispersion as it is in qualitative disagreement with direct measurements of self-diffusion coefficients using Nuclear Magnetic Resonance (NMR) (Turanov and Tolmachev, 2009; Gerardi et al., 2009) and Fluorescence Correlation Spectroscopy (FCS) (Subba-Rao et al., 2011) on a spatially uniform sample. Ozturk et al. (2010) only observed the presence of interfacial complexation between fluorescein and alumina, but there was no reason provided (phenomenological or mathematical) as to why that explains enhancement. Ironically, in the system they studied (parallel miscible streams co-flowing in a microchannel) they only observed a reduction in the measured diffusivity. In summary, there is no explanation, on a sound theoretical footing, for the observed enhancement in mass-transfer in nanofluids.

Here, we explore our hypothesis that the enhancement is because of a diffusiophoretic motion (Prieve et al., 1984) of the nanoparticles giving rise to a convective motion (counter to the direction of diffusiophoresis) of the surrounding fluid containing the dye (or other solute) molecules, that is measured as an increase in the apparent diffusivity of the solute. That is, the observed motion of the dye (or solute) is not an enhanced diffusion but a bulk directed (not random) motion of the solution.

We first elaborate the contrasting observations on the effect of nanoparticles. For a spatially uniform distribution of the nanoparticles and the solute, the self-diffusion coefficient measured using FCS (Subba-Rao et al., 2011) or NMR (Turanov and Tolmachev, 2009; Gerardi et al., 2009), shows no enhancement in the presence of nanoparticles. There is a marginal decrease that is explained by a hindered motion encountered due to the nanoparticles, in reasonable agreement with an effective medium theory. A significant enhancement is observed only in an inhomogeneous system (some of the configurations are shown in Fig. 1). In all these systems the solute, whose diffusivity or mass-transfer coefficient is measured, is initially located in a region of finite concentration adjacent to another fluid with zero concentration of the solute.

Given the ubiquitous presence of a gradient in the dye or other solute concentrations, we propose that the anomalous increase in diffusivity is due to diffusiophoresis. Diffusiophoresis is the

phoretic motion of colloidal particles in the presence of a gradient in the concentration of a solute that interacts with the surface of the colloidal particle. Diffusiophoresis is a well-known phenomenon (Derjaguin et al., 1961); Derjaguin provided a simple expression for the phoretic velocity of the particle. The most comprehensive derivation of the equations of motion of the particles for Brownian and non-Brownian suspensions has been recently published (Brady, 2011) for non-electrolyte suspensions. Depending on the nature of the interaction, attractive or repulsive, the particle experiences a positive or negative velocity, respectively, along the direction of the concentration gradient. The micro-mechanical origin of this velocity is due to a solute-induced gradient in the normal stresses (fluid pressure) in the fluid surrounding the particle, that is balanced by the viscous forces at the surface of the solid (Anderson et al., 1982; Prieve, 2008). Our postulate is that the drift motion of nanoparticles in a concentration gradient induces a counter-convective motion of the solvent containing the dye molecules leading to an increase in their spread, which is often reported as an increase in the diffusivity.

In this work, we construct a system identical in the fundamental phenomenology to the system used in the dye-drop experiments (Krishnamurthy et al., 2006; Fang et al., 2009), where a drop of dye is placed gently on a nanofluid inside a small cavity. It is possible that due to this action of dropping the dye, there could be interfacial instabilities resulting from the inertial motion of the drop (Ozturk et al., 2010). To avoid this, we recognise that when a drop of dye is placed in a suspension of nanoparticles, an interface between two nearly “stationary” fluids is created—one side being the solution of dye in water and the other side being the suspension of nanoparticles. By using a controlled system without requiring to drop a liquid, we avoid the irregular motion observed in the earlier work (Krishnamurthy et al., 2006), measure the initial velocity of the dye front and compare it with the estimate obtained using the diffusiophoretic velocity of the nanoparticles. The method is detailed in Section 2, the measured velocities are presented in Section 3, and estimates of diffusiophoretic velocities are made and compared with the measured velocities in Section 4. Finally, in Section 5 we present a brief discussion on other systems where enhancement has been reported, and how it may be related to diffusiophoresis.

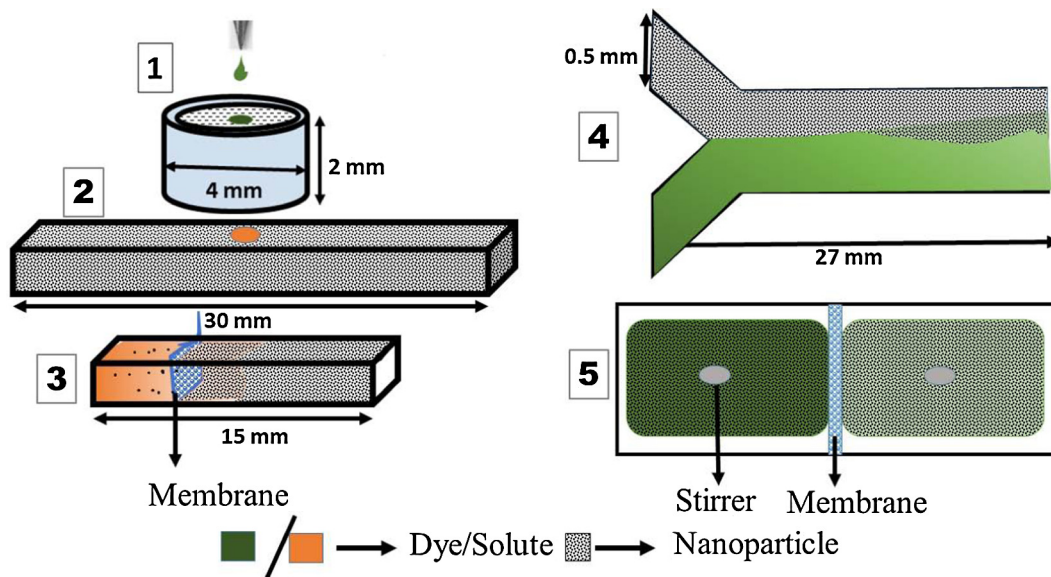


Fig. 1. Schematic of a few inhomogeneous systems where the effect of nanoparticles on the diffusivity of a solute was studied. 1: Dye-drop set up of Krishnamurthy et al. (2006) and Fang et al. (2009). 3: Membrane barrier diffusion of Veilleux and Coulombe (2010). 4: Microfluidic setup of Ozturk et al. (2010). 5: Diffusion cell of Feng and Johnson (2012). While in the first three cases, an enhancement in the measured diffusivity is noted, in the last two cases no enhancement has been reported.

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