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Distribution of Gd(III) ions at the graphene oxide/water interface

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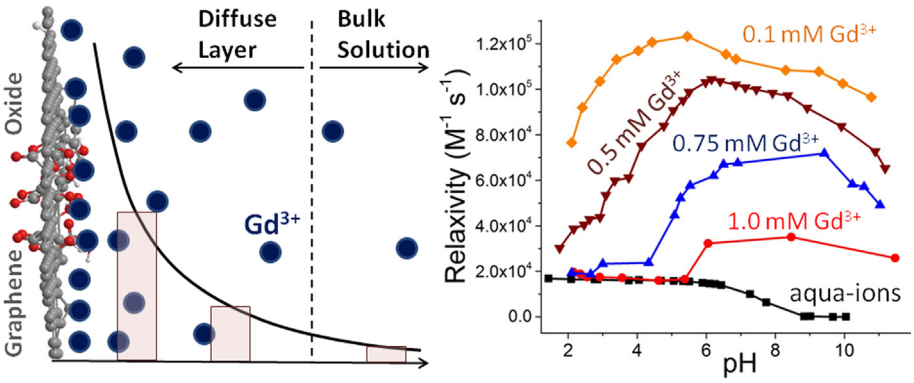
GRAPHICAL ABSTRACT



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ABSTRACT

Graphene oxide (GO) have emerged recently as a novel material for sorbing metal cations from aqueous media. However, the literature data on sorption capacity differ by more than one order in magnitude, and the nature of the chemical bonding between GO and metal cations remains unclear. In this work we show that Gd^{3+} ions are bound to GO by both coordinate-covalent bonding and electrostatic attraction with prevailing the former. We provide the complete account for the GO sorption toward Gd^{3+} as the function of the Gd^{3+}/GO ratio and pH of solution. The upper limits of the strong bonding are determined as 0.70 and 0.16 mmol(Gd^{3+})/g(GO) in the neutral and in the intrinsically acidic solutions, respectively. At large excess of Gd^{3+} in the neutral solutions, the sorption capacity reaches 1.45 mmol(Gd^{3+})/g(GO). The effectiveness of water, hydrochloric acid and EDTA as desorbing eluents is compared. We experimentally demonstrate the existence of the Gd^{3+} concentration gradient within the diffuse layer at the GO/water interface, and its exponential character on the distance from the GO surface. The thickness of the diffuse layer and the position of the slipping plane are estimated. Such characteristics, typical for colloid systems, show that in solutions, GO flakes form distinct phase, even though they are just one atom thick.

1. Introduction

Graphene Oxide (GO) gained enormous attention during the last decade due to its unique properties allowing numerous applications [1]. Unlike its non-oxidized counterpart, GO is soluble in water and in several polar organic solvents, affording solution-phase processability. Despite this apparent advantage, surprisingly little is known about the nature of the GO solutions, and especially about the GO/liquid interface, which remains *terra-incognito*. The GO solutions are very unique in terms of their classification. The lateral dimensions of GO flakes vary from hundreds of nanometers through tens of microns. Based on the lateral size, the GO solutions

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