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Application of graphene oxide nanoplatelets for adsorption of Ibuprofen from aqueous solutions: Evaluation of process kinetics and thermodynamics

Priya Banerjee^a, Pinaki Das^b, Aisha Zaman^c, Papita Das^{c,*}

^a Department of Environmental Science, University of Calcutta, 35, Ballygunge Circular Road, Kolkata 700 019, India

^b Department of Physiology, Calcutta National Medical College, 32, Gorachand Road, Kolkata 700014, India

^c Department of Chemical Engineering, Jadavpur University, 188, Raja S.C. Mullick Road, Kolkata 700 032, India

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ABSTRACT

Ubiquitous occurrence of several pharmaceuticals in discharged sewage effluents has led to considerable deterioration of life and quality of receiving water bodies. Ibuprofen, an acidic nonsteroid drug, is one such pharmaceutical being widely used for its analgesic, antipyretic and anti-inflammatory properties. The present work investigated the efficiency of graphene oxide nanoplatelets (GONPs) in adsorption of Ibuprofen from its aqueous solutions. The GONPs were characterized by electron microscopy and X-ray diffraction to analyze changes in structure and morphology occurring due to adsorption (if any). The impact of various process parameters on percentage removal (%) of Ibuprofen was determined by batch adsorption experiments. The data obtained were subjected to isotherm and kinetic analysis in order to describe the distribution of ibuprofen between the liquid and solid phases in the batch studies. The results obtained best fitted the Langmuir isotherm model and were determined to be guided by pseudo second-order kinetics. Thermodynamic parameters such as Gibb's free energy, Enthalpy and Entropy were also evaluated and the results revealed the endothermic and spontaneous nature of the process of adsorption of ibuprofen onto graphene oxide. Hence, graphene oxide may be considered as a suitable adsorbent for large scale efficient treatment of water contaminated with ibuprofen and similar other anti-inflammatory drugs.

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1. Introduction

Several studies in the past few years have shown that the pharmaceutical drugs, sold both with prescription and over-the-counter, may end up in water bodies as a through excretion with urine and feces as products of metabolism or indiscriminate dumping of unutilized medicines (Bound and Voulvoulis, 2005). Previous investigations suggest that non-steroidal anti-inflammatory drugs (NSAIDs) are among the

most common group of pharmaceuticals detected in surface and ground water across the globe (Cleuvers, 2004). Ibuprofen (IBP) is one of that most widely consumed NSAID and is found in detectable concentrations in the effluents from sewage-treatment plants and waste water treatment facility units (Guedidi et al., 2013). It features amidst the top five most commonly consumed drugs in the United Kingdom as well (Cho et al., 2011). IBP has high mobility (Guedidi et al., 2013) in the aquatic environment but is hydrophobic

* Corresponding author. Tel.: +033 24572696; fax: +033 24146378.

E-mail address: papitasaha@gmail.com (P. Das).

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and moderately soluble in water (21 mg/L at 25 °C; [Yalkowsky and Dannenfelser, 1992](#)). According to the US and European regulatory guidance, current pharmaceuticals are subjected to standard acute toxicity tests (where algae, *Daphnia magna* and fish are selected as experimental organisms) when the concentration of the active ingredient in effluent exceeds 1 µg/L (US legislation) or 10 ng/L (European Medicines Agency) ([Gros et al., 2010](#)). However, the presence of IBP in high concentrations (0.050–100 µg/L) in effluents from wastewater treatment plants (WWTPs) has been reported by many authors ([Salgado et al., 2010](#); [Gros et al., 2012](#); [Jakimska et al., 2014](#)).

Conventional processes such as coagulation, flocculation, and sedimentation are not wholly effective in the removal of NSAIDs like IBP. Several studies have also established the biodegradation of pharmaceuticals exhibited in WWTPs ([Radjenovic et al., 2007](#); [Verlicchi et al., 2012](#)). However, the active ingredients of pharmaceuticals are specifically designed to elicit biological responses and hence widely differ from conventional pollutants in their action ([Lishman et al., 2006](#)) and pharmaceutical removal using microorganisms may develop drug resistance in the used organisms ([Martucci et al., 2012](#)). Also, if not completely removed, the parent compounds as well as the excreted or transformed products will continuously exert detrimental effects on aquatic ecosystems ([Lishman et al., 2006](#)). However, biological treatment processes incur high cost and result in formation of sludge or other undesirable byproducts ([Adebowale et al., 2014](#)). Therefore, it is imperative to design advanced treatment processes for achieving efficient removal of NSAIDs like IBP in highly reduced time and a cost effective manner.

In recent times, the removal of different contaminants through the process of adsorption has emerged as a proficient and commercial substitute to the conventional water treatment facilities ([Liu et al., 2012](#); [Banerjee et al., 2015](#)). Till date various materials like clays, hydrophobic porous polymer and carbon-based materials have been reported to have selectively removed several organic and inorganic pollutants from water. Of these the carbon-based materials are the more preferred adsorbents owing to their high specific surface area, high chemical and mechanical stabilities ([Jaegfeld et al., 1983](#)). Earlier studies have reported that carbon-based nanomaterials, such as graphite ([Jaegfeld et al., 1983](#)), fullerene ([Cheng et al., 2003](#)), and carbon nanotubes (CNT) ([Stafiej and Pyrzynska, 2007](#)) have been found to play a chief role in sorption of inorganic and organic contaminants. Nevertheless, synthesis of most of these carbon materials requires expensive techniques. In contrast, Graphene oxide (GO) and Graphene (GR) nanoplatelets (NPs) have been utilized widely for adsorption studies due to their cost effectiveness, convenience of synthesis and application in significantly low quantities ([Wang et al., 2012](#); [Yao et al., 2012](#); [Li et al., 2013](#); [Travlou et al., 2013](#); [Das et al., 2014](#)).

GONPs can be synthesized from graphite by diverse chemical oxidation routes. The strong oxidation results in the formation of multiple oxygen-containing functional groups (such as carboxyl, hydroxyl, epoxy) on different layers of GONPs. GONPs are hence characterized by a highly-oxidized planar structure holding 25–33% oxygen with good adsorption potentials in between layers ([Stafiej and Pyrzynska, 2007](#); [Wang et al., 2012](#); [Li et al., 2013](#)). These hydrophilic oxygen moieties aid GONPs to undergo comprehensive dispersion in water thereby stabilizing it in a ([Yao et al., 2012](#)). Owing to its characteristic structure, GONPs have emerged as potent

adsorbents. Its electronic properties help it to interact strongly with organic molecules, via non-covalent forces, such as hydrogen bonding, π - π stacking, electrostatic forces, van-der-Waals forces. Moreover GONPs are additionally benefited by their nano-sized structure. This provides some advantages such as rapid equilibrium rates, high adsorption capacity, and effectiveness over a wide range of pH values ([Das et al., 2014](#)).

The present work was undertaken to investigate the adsorption capacity of graphene oxide for the removal of an IBP from aqueous surface. The characterization of the synthesized graphene oxide was done by Scanning electron microscope (SEM), Transmission electron microscope (TEM), Atomic force microscope (AFM) and X-ray diffraction (XRD) analysis was carried out before and after IBP adsorption and compared for changes (if any). Data recorded in batch studies with variable experimental parameters were also subjected to calculation of adsorption isotherms, kinetics and thermodynamics.

2. Materials and methods

2.1. Preparation of adsorbent (GO)

All chemicals used in this study were of analytical grade and obtained from Merck, India. GONPs were prepared from commercially obtained graphite flakes by modified Hummers method ([Li et al., 2013](#); [Banerjee et al., 2015](#)). Graphite flakes was placed in a flask kept in an ice-water bath, to which concentrated H₂SO₄ was gently added with stirring. Potassium Permanganate (KMnO₄) which was slowly added to the aforesaid mixture for over 30 min. This mixture was then subjected to vigorous stirring for about 180 min at a temperature of 313 K. Then, the mixture was allowed to stand at room temperature for 10 min before being slowly diluted with distilled (DI) water. The remaining KMnO₄ was further decomposed by adding hydrogen peroxide (H₂O₂; 30 wt%). The insoluble precipitations were removed. The mixture thus obtained was then filtered and the residue was washed and dried in a hot air oven for 48 h. The dried residue was crushed and stored for future use as GONPs.

2.2. Preparation of adsorbate (IBP) solution

2-[4-(2-Methylpropyl) phenyl] propanoic acid, commercially available as ibuprofen, was purchased from Sigma Aldrich and used as received. This compound was moderately soluble in water ([Yalkowsky and Dannenfelser, 1992](#)) but highly soluble in a majority of organic solvents ([Lee et al., 2006](#); [Manrique and Martinez, 2007](#)). A stock solution (10 mg/L) of IBP was prepared by dissolving weighed amount of powdered IBP in 10% Methanol solution with subsequent heating and stirring. Different concentrations of IBP were obtained by diluting the stock solution so prepared with 10% Methanol.

2.3. Measurement of residual IBP concentrations in solution

A calibration curve of IBP was prepared in the concentration range of 2–10 mg/L prepared in 10 mL volumetric flasks. The IBP concentration of the standards and untreated and treated effluents was determined spectrophotometrically using a double beam UV-visible spectrophotometer (Perkin Elmer Lambda 25, USA) at an absorbance wavelength of 221 nm. In order to minimize the interference of fine GO particles, all samples

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