



A modular process for the treatment of high level liquid waste (HLLW) using solvent-impregnated graphene aerogel

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ARTICLE INFO

Keywords:

Graphene aerogel
Solvent impregnated adsorbent
Modular process
HLLW

ABSTRACT

Graphene aerogel (GA) is a kind of superhydrophobic material with low density, high specific surface area and porosity. It can be used as modular matrix material to be impregnated with extractant due to its excellent organic solvent immobilization performance. In this paper, the modular GA based solvent impregnated adsorbents (GA-SIA) with various extractants were prepared. The adsorption performance of these adsorbents for typical fission products in high level liquid waste (HLLW) was investigated, and it is found that the adsorption performance is strongly dependent on the impregnated extractants. Based on the batch adsorption experiments, a modular process for the treatment of HLLW was proposed and tested. The results demonstrate that the modular process based GA-SIA possesses advantages of high selectivity, simple operation and low cost.

1. Introduction

Nuclear energy has provided a significant part of the energy demand in the world. In order to improve the safety and environmental friendliness of nuclear energy, the reduction of radioactive polluting emissions in nuclear fuel cycle is still one of the most important tasks. The appropriate management of radioactive waste streams requires effective separation of various useful, toxic and hazardous materials. Taking high level liquid waste (HLLW) for example (Romanovskiy et al., 2001; Wei et al., 2012; Xu et al., 2014), it is a strong radioactive waste generated in nuclear fuel cycle and consists of long-life radioactive fission products such as heat emitting nuclides Sr and Cs, minor actinide and lanthanide elements. Among these radionuclides, Sr and Cs should be removed to reduce the threat to human health and environment. On the other hand, they can also be applied in several industries as a heat generator (Ma et al., 2017; Sun et al., 2012). Minor actinide should be separated for further transmutation (Heidet et al., 2017). Furthermore, there are some precious rare earth elements in the HLLW. From the view of making full use of resources, they should also be recovered (Xu et al., 2014). To achieve the partition of above elements, a number of techniques have been developed, for instance, sorption of Cs by inorganic ion-exchange media (Dashtinejad et al., 2014; Sun et al., 2012), extraction of Sr by crown-ethers (Dai et al., 1999), and extraction of the actinide and lanthanide elements by neutral phosphorus extractant (Li et al., 2016), carbamoyl phosphine oxide

derivatives (Law et al., 1999) and amide extractant (Narbutt et al., 2015). For above classical techniques, a multistep process and several unit operations are usually required to separate and recover desired radionuclides from HLLW efficiently. In addition, these conventional techniques still suffer from economical and environmental limitations due to the labor-consumption, high usage of chemicals and massive liquid waste production (Shaeri et al., 2015; Zhong and Wu, 2012). Thus, an appropriate method which is simpler, eco-friendly and less cost should be developed to achieve the effective treatment of HLLW.

Recently, a concept of solvent impregnated adsorbent has been proposed and developed (Demirel et al., 2003; Hussein, 2011; Hosseini et al., 2016; Muraviev et al., 1998; Raouf and El-Kamash, 2006; Tang et al., 2017; Van Hecke and Modolo, 2004). It can be described as a technique that combines the selectivity of extraction and the easy operation of adsorption (Tang et al., 2017). It is devisable by physically or chemically impregnating specific extractant or reactive groups into a microporous solid matrix, for the adsorption of desired target. In this approach, selecting appropriate matrix is very important for the preparation of adsorbent and further target adsorption. Several kinds of materials, such as macroporous polymeric sorbents (Demirel et al., 2003; Lin et al., 2010; Tang et al., 2017), hydrophobized silica gels (Zhang et al., 2005; 2007), and carbonaceous materials (Dashtinejad et al., 2014; Feng et al., 2010; Shao et al., 2009; Wang et al., 2009; Yang et al., 2014) have been widely studied as the matrix to be impregnated with various extractants. Among these materials, there are some

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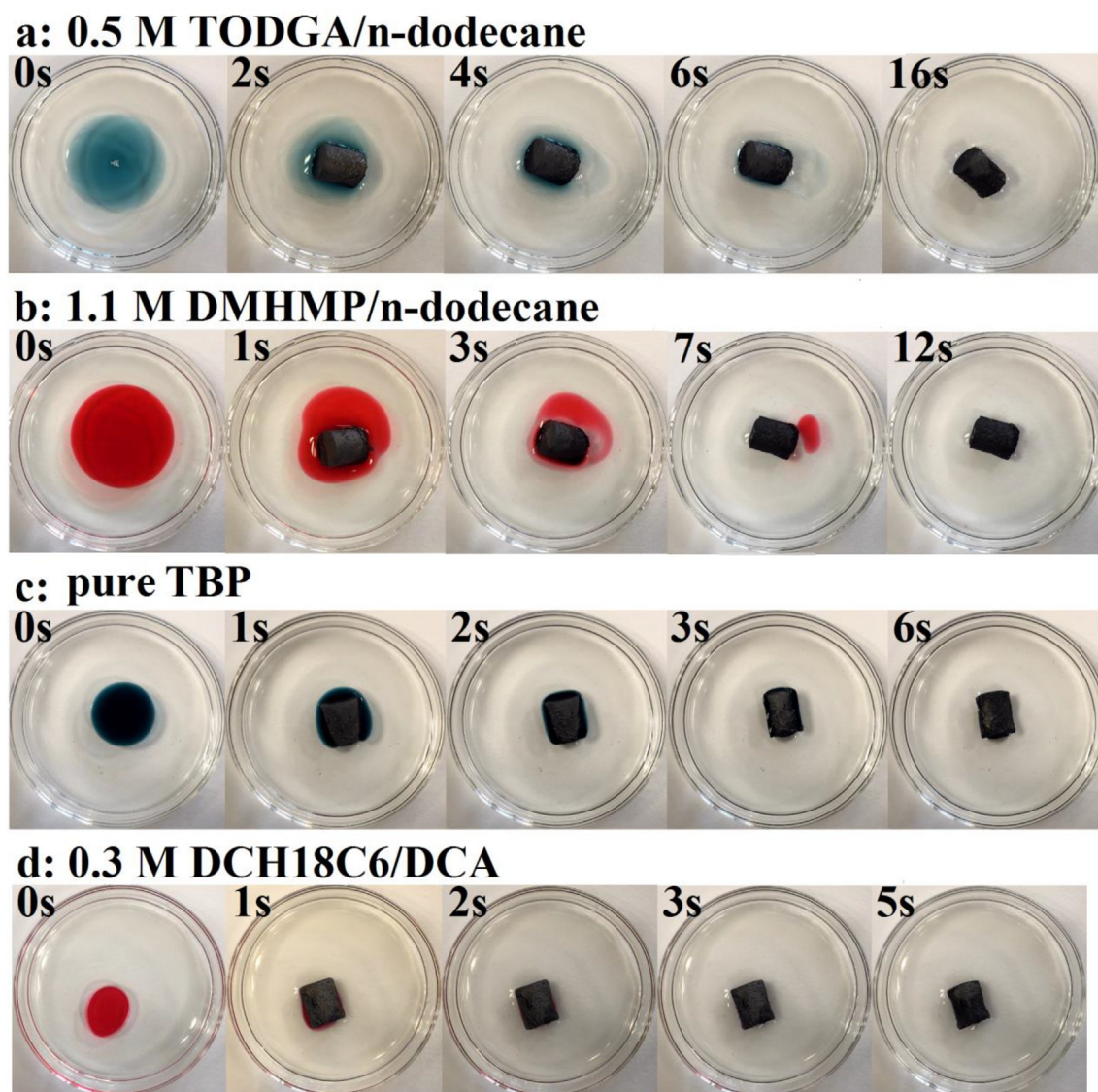


Fig. 1. Snapshots of the adsorption process of organic solution at different time interval (0.5 mL of organic solution was adsorbed by 3.0 mg of GA). (a) 0.5 mol·L⁻¹ TODGA/n-dodecane dyed with Sudan Black B, (b) 1.1 mol·L⁻¹ DMHMP/n-dodecane dyed with Oil Red O, (c) pure TBP dyed with Sudan Black B and (d) 0.3 mol·L⁻¹ DCH18C6/DCA dyed with Oil Red O. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 1

Comparison of the immobilization capacity for various impregnated adsorbents (Q).

Impregnated adsorbent	Matrix material	Functional extractant	Q (g·g ⁻¹)	Reference
GA-DCH18C6/DCA(0.3 M)	GA	DCH18C6/DCA(0.3 M)	286	Present work
GA-DMHMP/n-dodecane(1.1 M)	GA	DMHMP/n-dodecane(1.1 M)	201	Present work
GA-TODGA/n-dodecane(0.5 M)	GA	TODGA/n-dodecane(0.5 M)	206	Present work
GA-TBP	GA	TBP	221	Present work
Amberchrom® CG-71C-TODGA	Amberchrom® CG-71C	TODGA	0.13	Van Hecke and Modolo, 2004
Amberchrom® CG-71C-DMDOHEMA	Amberchrom® CG-71C	DMDOHEMA	0.33	Van Hecke and Modolo, 2004
SiO ₂ -P-TODGA	SiO ₂ -P	TODGA	0.33	Zhang et al., 2005
SiO ₂ -P/TBP	SiO ₂ -P	TBP	0.30	Zhang, 2007
SiO ₂ -P-Calix[4]arene-R14	SiO ₂ -P	Calix[4]arene-R14	0.24	Zhang, 2007
Egy-sorb-TBP	Egy-sorb	TBP	2.4	Hussein, 2011
XAD-16HP-D2EHFA	XAD-16HP	D2EHFA	0.54	Tang et al., 2017
AC-KCuCF	AC	KCuCF	12.5	Wang et al., 2009

common shortcomings: (1) low immobilization capacity of extractant on matrix due to the high density and limited porosity of the matrix materials; (2) complex preparation process, and some others. Therefore, it is necessary to find a more appropriate matrix material to achieve the simple production of adsorbents and high immobilization capacity of

extractants.

Graphene aerogel (GA) is a kind of three-dimensional graphene architecture material. As a special carbonaceous material, GA has the unique properties of superhydrophobic, high porosity, extremely high specific surface area and low density (Chen et al., 2017, 2018; Han

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