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ABSTRACT

Hydrogen is considered to be one of the renewable and cleanest energy source and most probable successor of conventional petroleum fuels. Hydrogen storage in nanoporous materials has been attracting a great deal of attention in recent years. The addition of carbon materials such as graphene oxides (GOs) and carbon nanotubes (CNTs) into MOFs can improve the physicochemical properties of parent MOFs with excellent chemical, mechanical and distinguished electronic thermal robustness. The decoration of the surface of graphene by metal could greatly facilitate the hydrogen storage. In the current study, the parent materials and their composites synthesized have been characterized by powder x-ray diffraction (XRD), scanning electron microscopy (SEM), transmission electron microscopy (TEM), and gas adsorption isotherms. The composite systems, GO/Cu-BTC/Ni@f-MWCNTs and GO/Cu-BTC/Pd@f-MWCNTs reached a hydrogen storage capacity of 3.91 and 4.21 wt. % at 77 K and 1.23 and 1.71% at 298 K.

KEYWORDS

Composites, Graphene Oxide, Hydrogen Adsorption, Hydrogen Storage, Metal-Organic Frameworks, Sustainable Energy

1. INTRODUCTION

Due to the on-going urbanization and growth of the world's population, there will be about 2.5 billion more people added to the urban population by 2050, mainly in Asia. These results increase stress on the natural resources which are available today (Sachs, 2008). Transport infrastructure is one of the most important factors for a country's progress. Although India has a large and diverse transport sector with its own share of challenges, like air pollution, depletion of relic fuels, etc. these can be overcome

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by energy-efficient technologies (Cigu et al., 2019). Sustainable transport systems are one amongst them which are majorly required for our country to progress and to compete with other countries. The sustainable transport system will decrease the stress on natural resources, pollution and help in reducing global temperatures (Aldakhil et al., 2019). As we also know that energy shortage is globally rampant due to fossil fuel level depletion which has prompted researchers to quest for alternative energy sources (Ansari & Holz, 2019). So, there is a need for other energy sources which replenish the gasoline products and at the same time remove problems corresponding to enormous emissions of greenhouse gases which are leading to climate change. In order to mitigate these greenhouse gases and to produce clean energy sources such as hydrogen came into light due to their efficacy towards substitution of relic fuels (Attari et al., 2019). Hydrogen is considered to be one of the renewable and cleanest energy sources and most probable successor for the conventional petroleum fuels. The most challenging aspect is the storage of hydrogen for hydrogen-powered vehicles (Zarbo et al., 2019). Until now, full implementation of a hydrogen-based energy system has been hindered in part by the challenge of storing hydrogen gas (Zoolfakar et al., 2014). The goal of a new hydrogen economy is to develop a suitable hydrogen storage medium to meet the U.S. Department of Energy (DOE) target of 1.5 kWh/kg system (5.5 wt % hydrogen) by 2020 for onboard light-duty vehicles, material-handling equipment, and portable power applications (USDOE). Metal-Organic frameworks (MOFs) are rapidly emerging as a promising material for gas storage applications (Panella et al., 2004, Thomas, 2009, Züttel et al., 2004, Langmi et al., 2003). Their exceptionally large surface area with tunable pore size and volume make them a good storage medium for clean energy applications especially for hydrogen (Hirscher et al., 2010). Among various MOFs, Cu-BTC [Cu_3(benzene-1,3,5-tricarboxylate)$_2$(H_2O)$_3$], is one of the most studied open framework materials for gas adsorption and storage applications (Chui et al., 1999). A paddle-wheel structure is constructed in CU-BTC framework by coordinating carboxylate moieties from 1,3,5-tricarboylate linker and Cu^{2+} ions. After removing the water from the framework of Cu-BTC, it will become an open three-dimensional porous structure. The exceptionally high surface is (1721 cm^2/g) and microporosity (0.74 cm^3/g) makes Cu-BTC one of the potential candidates for physisorption of hydrogen (Schüth et al., 2002, Lee et al., 2007, Yang et al., 2018,). So, this is the reason we have chosen Cu-BTC as MOF component for our studies. Physical adsorption has the great advantage of being completely reversible process with rapid kinetics. Further, due to the low adsorption enthalpy involved in physisorption, high storage capacities are reached only at lower temperatures. So, MOFs tend to exhibit high adsorption capacity only at low temperatures; however, their properties can be further improved by tuning the sorption behaviour by changing the chelating organic linkers, adding metals and making composites. A MOF composite is a multi-component material with multiple phases, undergoing tremendous improvement in gas adsorption (Panella et al., 2006). Extensive research is explored for the development of composites from MOFs, various carbon-based materials & metals with better hydrogen uptake capacity at room temperature are complying with the requirements of DOE for onboard vehicle applications. In this direction, Ardelan et al. (2012) had reported the preparation of composite material IMROF-8 and Pt on activated carbon catalyst which did not seem to exhibit this significant enhancement of hydrogen uptake compared to pristine materials (Ardelean et al., 2012). Another composite from IMROF-1 and AC/Pt had exhibit similar hydrogen uptake behaviour at room temperature (Costentin et al., 2010). However, improved hydrogen uptake was reported in case of Cu-BTC and activated carbon (Levasseur et al., 2010). Further, higher amount of hydrogen adsorption was observed for the composite GO@Cu-BTC compared to Cu-BTC at temperature of 77 K (2.81 wt % to 3.58 wt %) (Wang & Yang, 2008). Furthermore, at low pressures, Cu-BTC shows higher uptake of hydrogen compared to other MOFs (MOF-5) which is desirable for onboard applications (Panella et al., 2006). On the other hand, metal & metal oxide developed MWCNTs show higher uptake behaviour at room temperature. (Reyhani et al., 2011). Our previous results indicate that the MWCNTs decorated with nanoparticles of Mg, Fe, Cu, Zn exhibit improved uptake capacities for hydrogen at room temperature and moderate pressure (Konni et al., 2015, Konni et al., 2017, Konni et al., 2019, Chowdhury et al., 2012). Moreover, a strong