

## Efficient Iodine Capture of Cu<sup>0</sup>-Decorated Hexagonal Boron Nitride in Dry and Humid Environments

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### 1. Introduction

In the past few decades, silver-based sorbents have been widely used for removing radioactive iodine due to their high affinity with iodine and low solubility of AgI [1,2]. However, their popularity has been hindered by their high toxicity and exorbitant price, limiting their practical application. To address these issues, researchers have been progressively working to develop novel materials that are environmentally friendly and cost-effective, serving as alternatives to Ag-based sorbents.

Recent studies [3–5] have shown promising results with other metals like Bi, Cu, Fe, and Mg, which possess economic, non-toxic, and efficient properties, making them potential candidates to replace silver for capturing iodine. Compared with Ag, copper (Cu) has garnered significant attention as promising for iodine gas sorption. Moreover, Cu demonstrates significant superiority due to the presence of thermodynamically stable CuI, which boasts a lower Gibbs free energy of formation ( $\Delta G_f = -69.4 \text{ kJ mol}^{-1}$ ) compared to AgI ( $-66.3 \text{ kJ mol}^{-1}$ ). This difference in energy values reinforces Cu's distinct advantage over Ag.

In this study, we synthesized Cu@h-BN sorbents through a facile solvothermal treatment process using h-BN as the substrate. The sorption properties of Cu@h-BN were thoroughly explored in dry and humid environments. The characterization of the obtained composites is then investigated with XRD to elucidate their compositions and structures.

### 2. Methods and Results

In this section, we delve into the experimental conditions for both dry and humid iodine sorption.

#### 2.1 I<sub>2</sub> capture in dry and humid environments

The iodine sorption of Cu@h-BN was carried out under dry and humid environments with a 700 ml desiccator as reported in our previous literature. Briefly, 0.7 g of non-radioactive iodine crystals are placed into the bottom of the desiccator. Then, 0.07 g Cu@h-BN was placed on the stainless-steel plate above the iodine crystal, covered with a knobbed lid, and reacted at 200 °C for several times under static air.

To investigate the impact of water on I<sub>2</sub> sorption, experiments were conducted under controlled humidity conditions. To maintain a constant relative humidity

level (18 % and 75 % RH at 75 °C) within the desiccator, we carefully prepared and placed saturated solutions of CaCl and NaCl in an oven set at 75 °C for ~ 24 h. Subsequently, the I<sub>2</sub> uptake experiments were carried out using the same amount of powder and iodine as in the dry environment, while maintaining a temperature of 75°C.

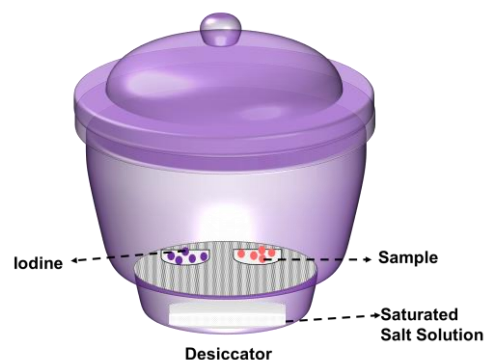


Fig. 1. Schematic of desiccator containing saturated salt solution.

### 3. Result and Discussion

Fig. 2 displays the XRD patterns of h-BN, Cu@h-BN, and I-Cu@h-BN. The distinct peaks associated with the (002), (100), (101), (102), (004), (103), (104), and (110) planes represent the characteristic diffraction patterns of commercial h-BN (PDF No. 00-034-0421). After the loading of Cu, the diffraction pattern of the synthesized Cu<sup>0</sup>@h-BN revealed three distinct peaks at  $2\theta = 43.3^\circ$ ,  $50.5^\circ$ , and  $74.1^\circ$ . These peaks can be attributed to the (111), (200), and (220) crystal planes of cubic metallic copper (Cu<sup>0</sup>, PDF No. 00-004-0836), signifying the presence of pure Cu<sup>0</sup> in the composite. After capturing I<sub>2</sub> at 200 °C, the observed diffraction peaks were identified as characteristic of the dominant phase of cubic CuI (PDF No. 01-075-0832). These results strongly suggest that Cu<sup>0</sup> underwent a reaction with I<sub>2</sub>, transforming into a stable form of CuI.

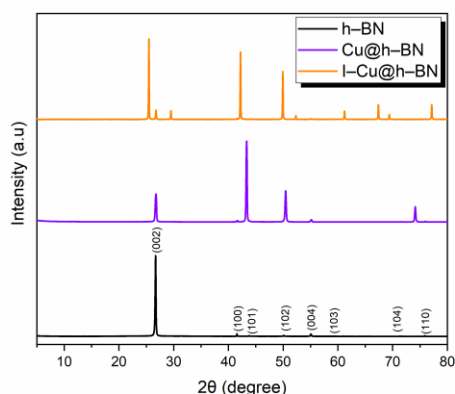


Fig. 2. XRD patterns of h-BN, Cu@h-BN and I-Cu@h-BN.

#### 4. Conclusions

In this study, we successfully synthesized metallic copper-decorated hexagonal boron nitride ( $\text{Cu}^0\text{@h-BN}$ ) using a simple solvothermal treatment. Subsequently, we conducted iodine sorption experiments on  $\text{Cu}^0\text{@h-BN}$  under varying conditions: two different temperatures (75 °C and 200 °C) and two different environments (ambient air and controlled humidities). Further research is essential to explore the immobilization of iodine waste forms, aiming to achieve stable storage of  $^{129}\text{I}$ .

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