

## Kinetics and mechanism of adsorptive removal of copper from aqueous solution with poly(vinyl alcohol) hydrogel



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**Abstract:** Recently, a renewed interest in techniques for heavy metal removal of wastewater has been growing because of embarking opportunities for industrial applications. We investigated the adsorption capacity of the copper on the poly (vinyl alcohol) hydrogel from the aqueous solution. Chemical structure and water absorption of the hydrogel were studied using FTIR and water uptake measurement, respectively. The results showed that the poly (vinyl alcohol) was crosslinked with glutaraldehyde, and the hydrogel highly exhibited the equilibrium swelling ratio because of its hydrophilicity property. Additionally, it was found that the adsorption process followed the pseudo-second-order kinetics and the mechanism diffusion was controlled by particle and film diffusions.

**Key words:** heavy metals removal; hydrogel; wastewater; adsorption; diffusion mechanism

### 1 Introduction

Heavy metals are serious causes of environmental pollutions affecting living microorganism and human with regards to their non-biodegradation and then accumulation in environmental compartments (air–water–soil–biota). Particularly, water is very important to organisms and is a dissolving medium of heavy metals. Many toxic heavy metals to human, such as copper (Cu), lead (Pb), cadmium (Cd), and chromium (Cr) [1,2], should be removed from wastewater before discharging to environments. Especially, Cu is an essential element in trace amount but at a higher level it is toxic to plants, animals, and humans [3]. Recently, Cu is widely used in various important industrial applications, so its removal and recovery from wastewater are significant for the environmental protection and human health.

Nowadays, numerous processes exist for removing dissolved heavy metals, including chemical precipitation [4,5], photocatalytic degradation [6], membrane filtration [7], electrochemical technology [8], reverse osmosis [9,10], and adsorption [11]. Among these processes, adsorption is one of the most effective and economical

methods for removing the heavy metals from aqueous solutions [12–15].

Considering environmental concerns, poly (vinyl alcohol) (PVA) is a water soluble polymer with characteristics of non-toxicity, biocompatibility and biodegradability [16,17]. Another crucial factor enhancing the suitability of PVA is hydrophilicity owing to its regular linear structure with a large number of side hydroxyls on the molecular chain [18,19]. Hence, PVA has been manufactured into a variety of adsorbent types, such as ion-exchange film [20] and hydrogel [21,22]. PVA is a semi-crystal hydrophilic polymer consisting of one hydroxyl group in each repeat unit with being crosslinkable representing desirable adsorbent structure. Its mechanical strength and chemical stability can be enhanced through crosslinking by chemical reaction, which can be defined as three-dimensional polymeric network [23]. Like other hydrogels, the PVA hydrogel shows faster adsorption kinetics for removing heavy metals from aqueous solution [24]. Recently, several research groups have investigated the removal of Cu with different types of adsorbent materials such as PVA/chitosan [22,25], PVA semi-IPN poly (acrylic acid)/tourmaline [26], and PVA/SiO<sub>2</sub> [27].

Surprisingly, there is less report about treatment of Cu(II) from aqueous solution using only PVA so far. Therefore, in the present study, we investigated the kinetics of Cu(II) ion adsorption with the neat PVA hydrogel as a novel adsorbent. The PVA hydrogel was prepared from PVA with glutaraldehyde as a crosslinking agent. The synthesized PVA hydrogel was investigated by Fourier transform infrared spectroscopy and water uptake capacity test, as well as batch adsorption equilibrium in order to study the effect of pH, contact time and temperature. Additionally, the kinetics and mechanism of adsorption were studied by using pseudo-first-order, pseudo-second-order and intraparticle diffusion models.

## 2 Experimental

### 2.1 Materials

Poly(vinyl alcohol) ( $M_w=140000$  g/mol,  $M_w/M_n=3.0$ , 87%–89% hydrolyzed) was purchased from Sigma–Aldrich (Germany). Glutaraldehyde was obtained from Fluka (Switzerland) and  $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$  was from Merck (Germany). All other chemicals were of analytical grade, and were used in as-received state without further purification.

### 2.2 Preparation of PVA hydrogel

PVA aqueous solution (10% in mass fraction) was prepared by dissolving PVA powder in distilled water at 90 °C with constant stirring for 2 h, and then cooled to room temperature. Crosslinking solution (5.71 mL) prepared from 50% (mass fraction) methanol, 10% acetic acid, 1.25% glutaraldehyde and 10% sulfuric acid with solution mass ratio of 3:2:1:1 was added into PVA solution (10.0 mL) under constant stirring. After that, the mixture was poured into a Petri disk and the gel was then formed within 30 min. The prepared PVA hydrogel was dried at 40 °C in a vacuum oven overnight.

### 2.3 FTIR spectroscopy

The PVA hydrogel was characterized by a Fourier transform infrared (FTIR) spectrophotometer (FT–710, Horiba FTIR) to verify its composition and chemical structure from functional groups. The samples were prepared by film casting techniques, and then the spectrum for each sample was recorded in the wavenumber range of 4000–400  $\text{cm}^{-1}$  at room temperature.

### 2.4 Water uptake capacity of PVA hydrogel

The ability of the PVA hydrogel to absorb medium was studied to understand the diffusion of medium and heavy metals into the PVA hydrogel, which is essential for heavy metal removal application. The water uptake

capacity ( $W_c$ ) of the PVA hydrogel was evaluated using the following equation.

$$W_c = \frac{m_t - m_0}{m_0} \times 100\% \quad (1)$$

where  $m_t$  and  $m_0$  refer to the mass of swollen adsorbent hydrogel at time  $t$  and dry state, respectively. Pre-weighed dry PVA hydrogel sample, defined as  $m_0$ , was immersed into a certain amount of deionized water. After immersion in water, the samples were taken out from water at certain time intervals (every 10 min), gently rapidly dried with filter paper to remove the excess water, and subsequently weighed to define  $m_t$ . All the tests were performed in triplicate ( $n=3$ ) under identical conditions.

### 2.5 Adsorption capacity

The adsorption capacity of Cu(II) ions onto the PVA hydrogel was carried out at ambient condition. Dry PVA hydrogel (1 g) was introduced into the Erlenmeyer flask with solution (30 mL) containing 50 mg/L Cu(II) ions. The flasks were placed in a thermostatic stirrer and agitated at the fixed speed (100 r/min). Samples were taken periodically for measurement of aqueous phase at each time interval. The concentration of remaining Cu(II) ions in solution was estimated by an atomic absorption spectrophotometer (AAS) (AAnalyst 200, Perkin Elmer) [28]. The effect of contact time, pH and temperature was studied. For each condition, three samples ( $n=3$ ) were investigated. The adsorption capacity ( $q_e$ ) of the Cu(II) ions onto the PVA hydrogel was calculated using the following equation:

$$q_e = \frac{(C_0 - C_e)V}{m} \times 100\% \quad (2)$$

where  $C_0$  and  $C_e$  refer respectively to the initial and the equilibrium concentrations of Cu(II) ions solution;  $V$  is the volume of the Cu(II) ions solution; and  $m$  is the mass of the PVA hydrogel. The adsorption of Cu(II) ions was determined by

$$\eta = \frac{(C_0 - C_t)}{C_0} \times 100\% \quad (3)$$

where  $C_0$  and  $C_t$  are the concentrations at initial and time  $t$  of Cu(II) ions solution, respectively.

### 2.6 Kinetics of adsorption and mechanism

The kinetic parameters are useful in predicting the adsorption rate used as important information in designing and modeling of the adsorption process. Therefore, we investigated the kinetic rate of the Cu(II) ion adsorption on the PVA hydrogel using pseudo-first-order and pseudo-second-order models. Additionally, the intraparticle diffusion or Webber and Morris model

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