

## ACRYLATE-MODIFIED LINSEED HYDROGEL FOR THE REMOVAL OF COBALT IONS FROM CONTAMINATED WATER

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**Abstract**

Copolymerization of mucilage extracted from the flax seeds was done to design an efficient adsorbent for the removal of Co(II) ions from contaminated water. The copolymerized flax seed mucilage was used as adsorbent for Co(II) removal from contaminated water. It was evaluated for its ability to remove the Co(II) from the aqueous solution with the aim of imparting good water holding capacity. Batch adsorption tests were performed to study the effect of initial concentration of Co(II) ions, copolymer dosage, time to contact, temperature and the pH. The best conditions for the highest removal of Co(II) were as follows: concentration of Co(II) ions = 150 mg/L, dose of copolymer = 35 mg/g, time to contact = 40 min, temperature = 298K and the pH = 6. Therefore, in this case, the adsorption isotherm and adsorption kinetic could be determined.

**1. Introduction**

Heavy metals are very persistent, bioaccumulate and are toxic and form an important category of environmental contaminants. The planet's ecological equilibrium is seriously threatened by their endurance in ecosystems, which also puts humans and other species that feed them at danger [1]. Cobalt (Co<sup>2+</sup>) is one of the many heavy metals that is commonly a concern. Cobalt (Co(II)) is an integral part of vitamin B12, or cobalamin, for both humans and animals. It is involved in metabolism, it enhances the immune system and possesses antimicrobial properties [2]. Unfortunately, the quantity of Co(II) released into the environment exceeds the allowable norms, both in the nature and manmade

processes, which put the health of living creatures in danger. High levels of Co(II) exposure may cause diarrhea, vomiting, nausea, blood loss, thyroid damage, and heart disease. Low dose exposure to Co(II) may lead to anemia, anorexia and chronic edema [3].

The environmental hazards of Co(II) can be multiplied, since it can come in many forms, which makes it more bioavailable, and consequently more dangerous. Permitted levels for this metal in drinking water have been quickly adopted by agencies such as the US EPA and the World Health Organisation. The more that the concentrations of co-exposures and increasing concentrations in environment exceeds both allowables, they are directly related

to anthropogenic activities such as waste, production of products, and industrial emissions. The combination of several physicochemical methods has been used to remove heavy metals from polluted water. Examples are osmosis [4], ultrafiltration membrane system [5] such as reverse osmosis, chemical precipitation [6], precipitation [7] and ion exchange [8]. But the technique that is attracting more interest is adsorption, which offers a number of advantages [9].

It is one of the most preferred methods over other methods owing to its ability to achieve a high degree of enrichment of adsorbents, simple separation methods, and the use of environmentally friendly materials as sorbents [10]. Some biosorbents contain activated carbon, biological materials or agricultural waste. However, the low sorption properties of many traditional materials are a serious problem and is driving research to develop new, inexpensive and environmentally friendly water treatment sorbents [11].

Hydrogels based on polysaccharides have become of main interest in recent years. These materials have very good potential to be used as sorbents for the sequestration of Co(II) ions from groundwater leachates and wastewaters as they can be chemically functionalized with carboxylic acid anhydrides (e.g. succinic anhydride) to increase the metal-binding capacity of the material. Mucilage and polysaccharide rich materials are found to be stimuli-responsive, super-porous, superabsorbent and non-toxic, which has led to successful research on using them to develop efficient sustained-release drug delivery systems [12-27].

*Linum usitatissimum* L. The seeds of flax (Flax) were extruded a hydrogel known as Flax seed hydrogel (FSH). FSH has been used effectively as a sustained-release agent for the encapsulation of the pH-responsive drug delivery systems [28, 29]. This study aims to develop a chemically more enhanced and eco-friendly FSH-based super-sorbent which can remove  $\text{Co}^{2+}$  from the aqueous environment. The study aims to enhance the metal binding capacity of the FSH by converting it to its copolymeric form in which

the number of functional groups are more, which increases the availability of functional groups. In order to determine their significance in improving the sorption efficiency, the effects of the important operational variable such as initial metal concentration, the sorbent dose, contact time, pH and temperature will also be statistically analysed.

## 2. Materials and methods

### 2.1. Materials

The seeds collected from a local store were physically cleaned by removing dust and garbage, and separating the seeds, before testing. The flaxseed was washed before the extraction process and all the experimental procedure was conducted in DW water. No further purification was required as all of the reagents and solvents used are of analytical purity or higher. All of the glassware used prior to performing any experiments was thoroughly cleaned with nitric acid.

### 2.2. Isolation of hydrogel from Linseed and synthesis of sorbent

The isolation of the hydrogel composite from the flaxseed (FSH) was carried out as reported by [28] and the fabrication of the adsorbent was carried out as reported by [24].

### 2.3. Sorption studies

Batch adsorption experiments have been used to identify the optimal conditions for the adsorption of Co(II) ions removal onto the copolymer as well as the impact of numerous significant critical factors. The initial metal concentration, the amount of sorbent employed, the pH of the solution, the contact duration, and the temperature were the five main factors that were examined. Metal stock solution of 1000 mg/L of Co(II) was made by dissolving Cobalt salt in 1 L of DW. Different working solutions at various concentrations of the stock solution were prepared and the pH of each solution was finalized. 100 mL of a metal ion solution (usually 150 mg/L) was placed in an Erlenmeyer flask, and shaken for 40 min on a shaking thermostat at a specified temperature for each experiment.

The solid sorbent was filtered to remove any coagulated Co(II) ions in the solution. The amount of Co(II) ions removed from each sorbent material was calculated using the standard formulas listed in Equations 1 and 2, where  $q_e$  equals to the amount of Co(II) ions adsorbed by each sorbent. The concentration of both ions still present in the filtrate was determined using Flame Atomic Absorption Spectroscopy (FAAS).

$$q_e = \frac{C_i - C_e}{m} \times V \quad (1)$$

$$\text{Percentage uptake} = \frac{C_i - C_e}{C_i} \times 100 \quad (2)$$

Here  $q_e$  ( $\text{mg g}^{-1}$ ) indicates the equilibrium sorption capacity, while  $C_i$  and  $C_e$  are the initial and final concentrations of the metal in the solution respectively ( $\text{mg L}^{-1}$ ) and  $V$  is the volume of the solution (L) and  $m$  is the mass of the adsorbent (g).

### 3. Results and discussions

#### 3.1. Effect of initial metal ion Concentration

The ability of copolymer to sorb Co(II) ions as a function of the initial concentration. The

amount of metal stored by copolymer increased with the increasing concentration of metal ions in the solution, until it reached close to saturation level, as shown in Figure 1, with increasing concentrations of metal ions ( $\text{mg metal per gram of sorbent}$ , or  $q_e$ ). Co(II) was the most efficient sorbed ion whereas increasing concentration of either ion did not give any additional sorption capacity to copolymer at the starting condition of  $150 \text{ mg L}^{-1}$ . This indicates a need for additional research to determine the sorption efficiency of copolymer at higher starting metal ionic concentration. The enhanced sorption capacity was believed to be caused by an increase in sorbed metal ion concentration and concentration gradient which would drive the movement of the metal ions to copolymer. The plateau observed at Co(II) ion initial values above  $150 \text{ mg L}^{-1}$  indicates that remaining cation sorption sites on the surface of copolymer were saturated with Co(II) ions and no further sorption occurred [30, 31].

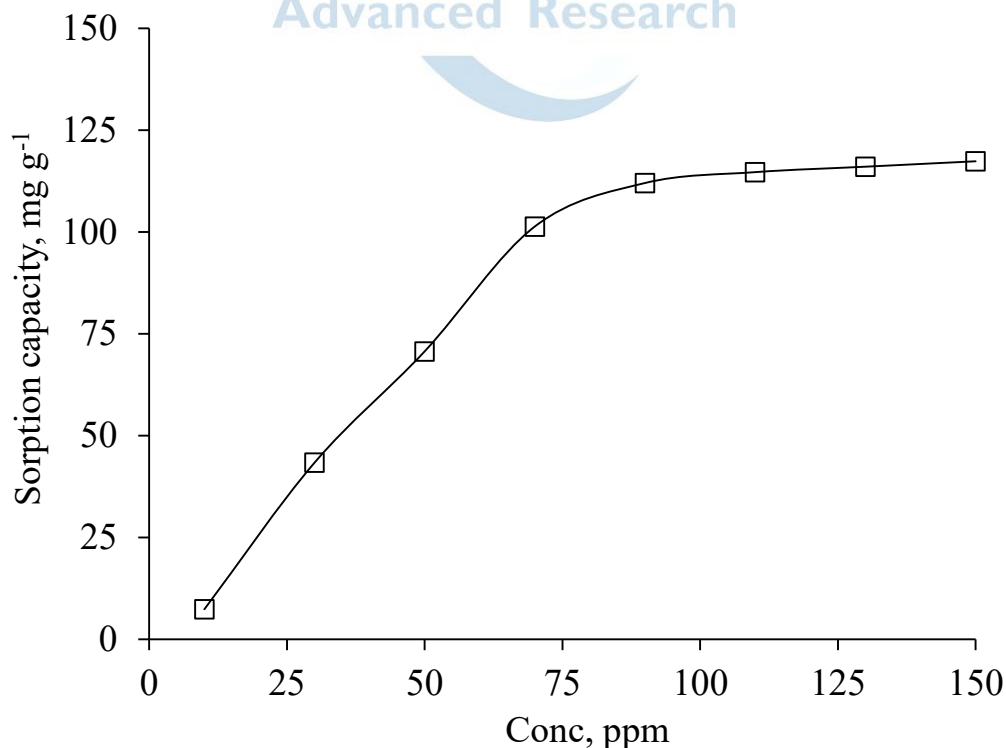


Figure 1: Effect of initial concentration.

### 3.2. Effect of sorbent dosage

Experimental testing was performed to determine the optimum amount of sorbent needed to maximise metals removal. The results demonstrated that the adsorption of both Co(II) with the increasing dose of mass of copolymer increased as the number of binding sites increased in the system with the increasing amount of sorbent. After a correct mass of 35

mg, however, the rate of uptake of the metals decreased slightly. This drop can be attributed to the metal ion concentration in the solution; if the amount of metal ion in the solution is not enough to fill up the sorbent, the excess amount of sorbent will have vacant binding sites [27, 32]. From this data it was found that 35 mg mass of sorbent is an ideal mass to be used for further experiments as shown in Figure 2.

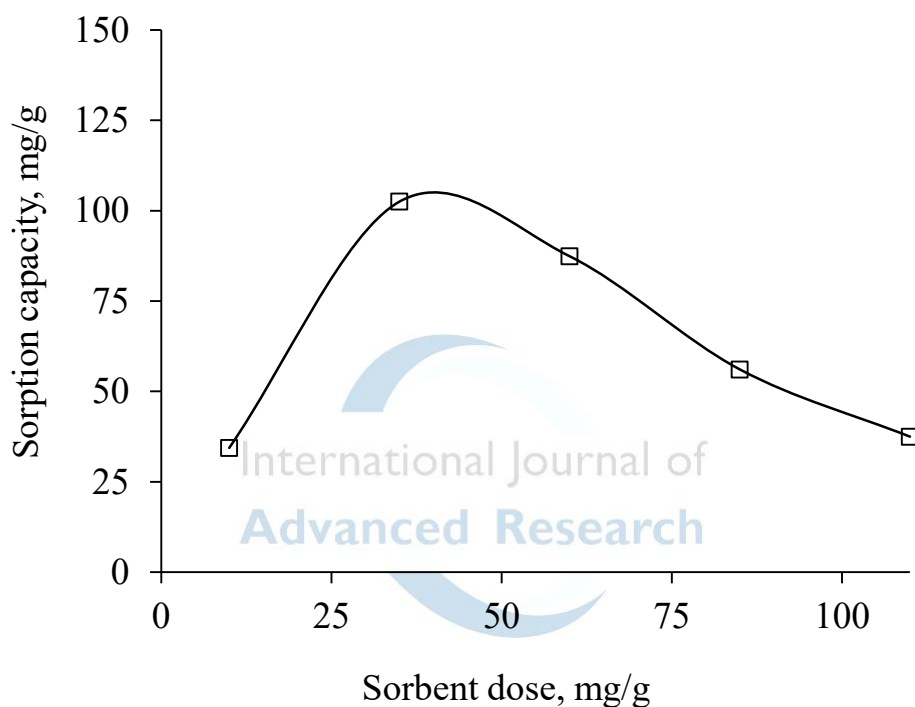


Figure 2: Effect of Na-FSH dose.

### 3.3. Effect of pH

The sorption capacity of copolymer is greatly influenced by the pH because it affects the surface charge and the ionization state of functional groups. The Co(II) sorption studies were performed using copolymer materials with a range of pH (2.0-10.0) as shown in Figure 3. The protonation of copolymer is thought to be the source of the low adsorption values seen in acidic media. This situation does not allow metals to be taken up by the cation exchange sites of the vault as it is. The sorption capacity of copolymer increased with increasing pH levels, which

increased to the peak at pH 6.0. At this higher range, the metal sorption is very fast as different functional groups are deprotonated and the sorbent acquires a negative charge at pH higher than its point of zero charge ( $pH_{ZPC}$ ). Outside this zone, precipitation of  $Co(OH)_2$  leads to a decrease in the amount of metal ions present in solution and thus the effectiveness of the exchange reactions [35]. The area around the  $pH_{ZPC}$  is positive and has a limited ability to bind to metal ions. The ideal pH for carrying out the remaining sorption experiments was thus found to be 6.

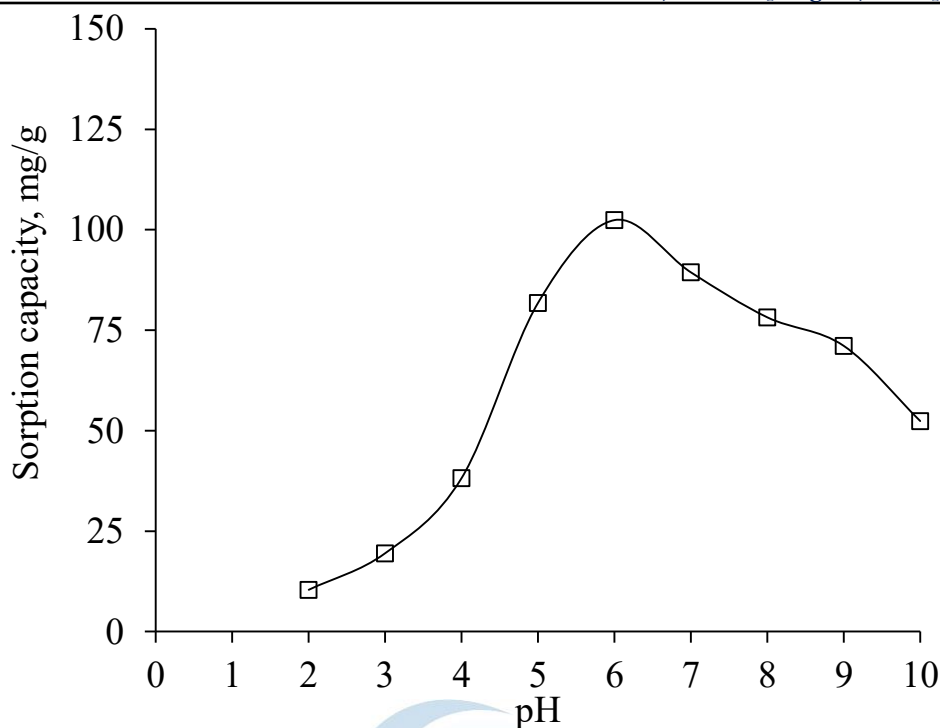


Figure 3: Effect of pH.

### 3.4. Effect of contact time

The amount of metal ions sorbed by the total sorption capacity depends greatly on the interaction time between the active sites of the sorbent and the metal ions present in solution. The determination of contact duration allows for an evaluation of the most suitable kinetic model for the material and insight into the adsorption mechanism. For the case of copolymer, the contact time was calculated for both Co(II) ions. After 40 min of contact, the majority of the metals were adsorbed and resulted in 85% removal of metal ions (Figure 4). An abundance of sodium-succinate functional groups on copolymer and the quick kinetics of the ion-

exchange mechanism, which exchanges  $\text{Na}^+$  ions with Co(II) ions, were the causes of this initial fast metal absorption. Following 40 min, the metal adsorption rate significantly declined due to the fact that the repulsive forces of the incoming cations, with the already adsorbed metal cations, prevented the advancement of additional metal ions to the available active sites for adsorption. The concentration of metal ions adsorbed did not change significantly after 40 min, thus, the longer the contact time, the more copolymer will not be able to increase its sorption capacity [23, 36]. This shows that soon copolymer will get into equilibrium and it will be a beneficial and useful substance.

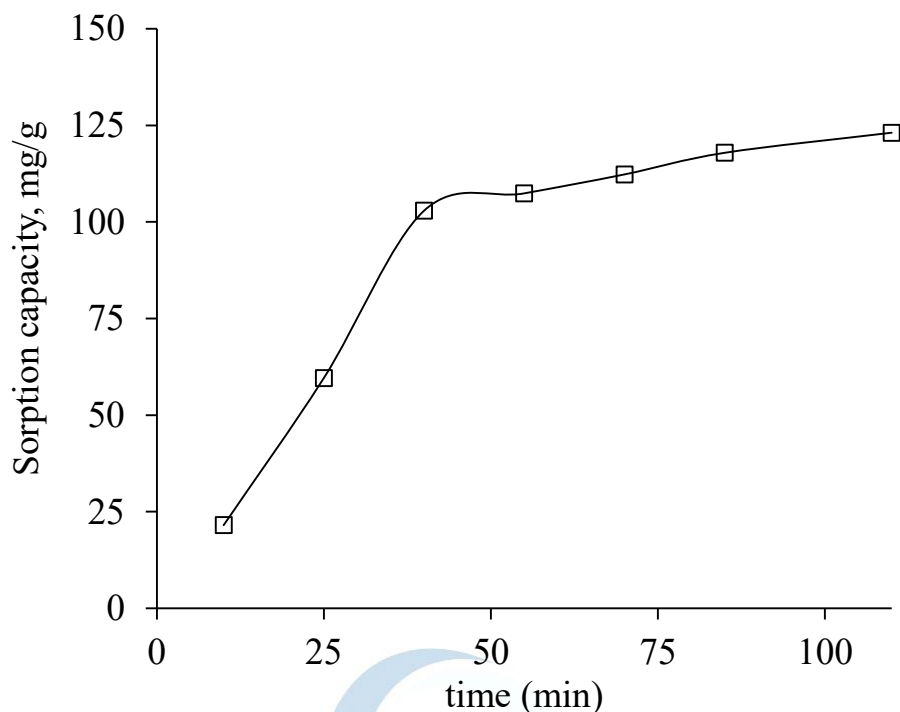


Figure 4: Effect of time.

### 3.5. Effect of temperature

The effect of temperature on the sorption performance of copolymer in the presence of hazardous metals is investigated in this work to gain more insight into the thermodynamics of the sorption process more specifically, to determine if the sorption process is exothermic or endothermic. The temperature range at which the experiments were carried out was 298 K to 343 K. As seen from Figure 5, the sorption capacity of the adsorbent decreased with increase of temperature and it is an exothermic process. A

lower temperature (less than 298 K) would not be sufficient to allow the mobility of Co(II) ions to fill all of the active sites on the surface of the sorbent, because the mobility of the  $\text{Na}^+$  ions in the sorbent at this lower temperature would be much faster. The number of metal cations that can be absorbed into the sorbent will decrease as the temperature increases, because at higher temperatures, the activity of  $\text{Na}^+$  ions would be higher [23, 24]. These results indicate that the optimum temperature for the sorbent to absorb Co(II) ions is 298 K.

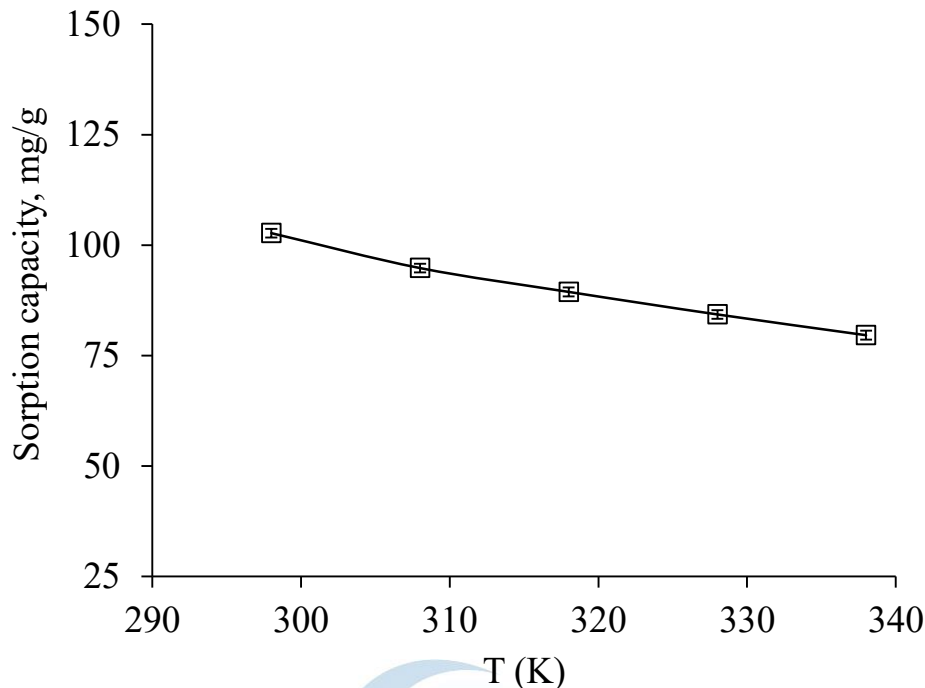


Figure 5: Effect of temperature.

#### 4. Conclusions

In this study, the novel biosorbent as copolymeric flax seed hydrogel was successfully developed by esterification. A number of factors affecting the ability to absorb Co(II) ions from aqueous solutions of copolymer were found to have significant effects. The sorption kinetics of both metals were very fast, and more than 85 % of both were absorbed in the first half hour of the operation. The high sorption capacities and good kinetics measured for the selective sorption of Co(II) in polluted water confirmed the very promising potential of copolymer for this purpose, which can now be used repeatedly.

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