# 알지네이트 혼합비드를 이용한 구리와 페놀의 이성분계 흡착평형 특성

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# **Adsorption Equilibrium Characteristics of Copper Ions and Phenol Binary System Using Composite Alginate Beads**

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## **Introduction**

Heavy metal solutions are widely used in industrial activities such as metal finishing, electroplating, painting, dying, photography, surface treatment, printed circuit board manufacture, etc. Most of the heavy metal ions are well-known toxic and carcinogenic agents, while metal residues in the environment pose a threat not only for human health, but also have serious detrimental effects for the aquatic ecosystem [1].

Phenolic compounds are produced in industrial processes such as gasification, coking, oil refining, phenolic resin manufacturing and paint stripping. Biological treatment of phenol is generally favoured over other processes in terms of lower cost as well as the possibility of complete mineralization. However, there is a limitation associated with the biological treatment due to the toxicity exerted by phenol itself at higher concentrations.

Many biopolymers are known to have a strong affinity for metal ions and the use of alginic acid as adsorbent for the recovery of valuable metal ions as well as removal of toxic metal ions has been studied [2]. Alginic acid is a biopolymer carrying carboxyl groups capable of forming complexes with metal ion.

Activated carbon is used widely to remove organic substances, which are evaluated as BOD (biochemical oxygen demand) or COD (chemical oxygen demand), and halogen from tap water and waste water, as well as dyes, phosphate and nitrogen from colored textile waste water and inorganic nutrient rich waste water [3,4]. Activated carbon can be used for the removal of both inorganic and organic materials, as well as low molecular weight and polymer substances.

In this study, alginate bead and activated carbon-alginate composite bead (AAC) were prepared by reaction with calcium chloride. And the adsorption of single and binary components for heavy metal ion and organic onto alginate bead and AAC beads was studied experimentally and theoretically.

#### **Materials and Methods**

 Alginate and powdered activated carbon (PAC) were obtained as powdered from Junsei and Yacuri chemical Co., Japan, without further purification. All of the reagents including CuCl22H2O (Junsei chemical Co., Japan), phenol (Sigma Co., USA), calcium chloride anhydrate (Duksan Pharm. Co., Korea) and phosphate buffered saline solution were reagent grade. The powdered alginate was dissolved in a phosphate buffer saline solution to produce a viscous solution with approximately 3 wt% alginate. And then, biopolymer alginate solution was cast into bead by a phase-inversion technique using calcium chloride solution. After mixing 3g of activated carbon with 100ml of 3 wt% alginate solution, the solution was casting in the calcium chloride solution in the same manner as used in the preparation of alginate bead. The alginate bead and AAC beads were characterized to determine particle size, pore size distribution and porosity after drying using direct drying at room temperature, vacuum drying, oven drying and freeze drying.

# **Results**

The pore structure of the AAC beads by freeze drying is shown in Fig. 1. As can be seen in this Figure, the highly reticular, three dimensional porous matrix of the AAC beads cross section is particularly evident at 1,500×. The physical properties of AAC bead are listed in table 1. The uniform pore structure distribution of AAC bead is an important criterion for the proper adsorption of heavy metal ions and organics on the whole surface area of the AAC beads. Fig. 2 shows the adsorption amount of copper ions onto three different adsorbents. The adsorption capacity of copper ions onto different adsorbents was in the following order : alginate bead > AAC bead > activated carbon. In case of alginate bead, the bead has many negative charges due to the dissociation of the carboxyl groups and the copper ions species is mostly cationic. So, attraction between the surface layer of the bead and the cationic copper results in increased adsorption. Fig. 3 shows the adsorption amount of phenol onto AAC bead at different initial pH of the solution. Adsorption capacity of phenol onto AAC bead increased with decreasing initial pH of the solution. The adsorption isotherm of phenol onto AAC bead was very similar to that of PAC. Since both adsorption isotherms are the Langmuir type. Fig. 4 shows that adsorption isotherms of copper ions and phenol binary system onto AAC bead at different initial pH of the solution. As can be seen in this figure, adsorption amount of copper ions onto AAC bead was greater than that of phenol in the binary system. Since adsorption affinity of copper ions onto AAC bead was larger than that of phenol. In case of copper ions, adsorption capacity increased with increasing pH of the solution. On the other hand, adsorption capacity of phenol onto AAC bead decreased with increasing pH. The adsorption equilibrium constants of copper ions and phenol binary system onto AAC beads listed in table 2.

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Table 1. Physical properties of AAC bead.

Property	Drying Sieve No.	Mean diameter BET surface		Pore	Pore volume
	type	[cm]	area $\lceil m^2/g \rceil$ radius $\lceil A \rceil$		$\lceil$ cm <sup>3</sup> /g]
AAC bead	Freeze $10 \sim 12$	0.21	193.8	26.50	0.26

Table 2. Adsorption equilibrium constants for binary component of  $Cu^{2+}$  and phenol onto AAC bead (1:1) at different pH.

	parameter	pH				
Isotherm type		copper ions		phenol		
		3	5	3	5	
Langmuir	$q_m$	2.37	2.55	0.74	0.72	
	b	2.09	2.58	18.2	5.96	
	$error(\% )$	3.12	6.54	9.13	9.93	
Freundlich	$\mathbf k$	1.59	1.78	0.86	0.83	
	n	2.26	2.93	2.75	1.90	
	$error(\% )$	4.27	8.27	9.15	10.1	
<b>Sips</b>	$q_m$	2.40	2.23	0.74	0.72	
	b	2.05	4.09	10.1	6.32	
	$\mathbf n$	0.99	0.95	1.237	0.98	
	$error(\% )$	3.16	7.97	10.3	10.6	



Fig. 1. SEM photograph of cross sectional view of AAC bead (×15,000**)** 



Fig. 2. Adsorption isotherms of copper ions onto different adsorbents



Fig. 3. Adsorption isotherms of phenol onto AAC bead at different pH.



Fig. 4. Adsorption isotherms of copper ions and phenol binary system onto AAC bead