



# Solvent-assisted synthesis of potassium copper hexacyanoferrate embedded 3D-interconnected porous hydrogel for highly selective and rapid cesium ion removal



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## ABSTRACT

Potassium copper hexacyanoferrate-embedded poly(vinyl alcohol)-citric acid hydrogel film (HPC) was prepared via a two-step method of Cu immobilization, followed by the diffusion of potassium hexacyanoferrate accelerated by acetone evaporation. The diffusion-derived KCuHCF formation in the preformed hydrogel facilitated the preservation of the 3D-interconnected hydrogel structure and dispersion of the KCuHCF nanoparticles. Using acetone as a non-solvent, reverse diffusion of the incorporated Cu in the hydrogel matrix was hindered; hence a large amount of KCuHCF was loaded in the matrix. The HPC exhibited substantially enhanced Cs<sup>+</sup> removal properties in terms of adsorption capacity, kinetics and selectivity. From the adsorption isotherm, the HPC showed a very high Cs<sup>+</sup> uptake of 667 mg/g KCuHCF. Moreover, the adsorbent revealed stable and high Cs<sup>+</sup> removal efficiency of 99.9% across a wide pH range from 2 to 10. The kinetics of Cs<sup>+</sup> removal was remarkably rapid with 99.5% removal achieved within 30 min from a dilute Cs<sup>+</sup> solution (9.18 ppm). When using seawater, the HPC exhibited almost unaltered Cs<sup>+</sup> removal efficiency above 99.5%, and high distribution coefficient  $K_d$  value of  $7.7 \times 10^5$  ml/g at an extremely low Cs<sup>+</sup> concentration (0.67 ppm,  $V/m = 1000$  ml/g), which highlighted the tremendous affinity for Cs<sup>+</sup>.

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## 1. Introduction

To meet our energy needs in the 21st century, nuclear power has emerged as one of the most promising alternatives to fossil fuels, providing approximately 9.7% of the world's energy [1]. However, from an environmental perspective, nuclear energy can pose a threat due to the release of approximately 2000–2300 t of radioactive waste into the environment each year [1]. Thus, for the continuous and long-term operation of nuclear facilities, environmental remediation and confinement of generated wastes remain a key priority [2]. Aside from the issues of operational radioactive effluent, nuclear incidents such as those witnessed at Fukushima Daiichi, and Chernobyl have resulted in the long-term and catastrophic contamination of the biosphere, hence issues around accident tolerant power plants are also being addressed [3].

Among all the radioactive elements, gamma ray emitters <sup>134</sup>Cs and <sup>137</sup>Cs are the richest and the most hazardous nuclear fission

products of uranium [4–6]. Both isotopes are considerably harmful to humans and can easily destroy the environmental habitat since they can quickly migrate through ground and sea waters, accumulating in the biosphere for hundreds of years [5].

Ion exchange with an inorganic adsorbent is a promising technology for the recovery of radioactive elements from nuclear waste streams due to its convenience (availability), effectiveness and high adsorptive capacity. Zeolites [7,8], crystalline silicotitanates [9,10], chalcogenides [11–13], and clay minerals [14,15], are common inorganic ion exchangers, however, their low selectivity for Cs over other competitive elements such as Na, K, Mg, Co, Ni or actinides, inhibits their practical application for large scale processes [5].

In the last few decades, the transition metal based hexacyanoferrate (HCF) has demonstrated excellent ion exchange properties for the selective separation of Cs<sup>+</sup>, even in a large excess of co-existing competing cations (Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>, . . .), which are often present in contaminated effluents [1,4,5,16–18]. Since metal HCF has a well-known perovskite-like face-centered cubic crystal structure, with a channel diameter of around 3.2 Å, small hydrated cations such as Cs<sup>+</sup> can be inserted, whereas the larger hydrated

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ions such as  $\text{Na}^+$  are selectively rejected [4,19]. Particularly, copper hexacyanoferrate has been widely used due to its simple preparation, low cost, superior  $\text{Cs}^+$  capture properties, and chemical stability over a wide pH range [3,17,18,20]. In spite of

the advantageous structural properties for the selective removal of  $\text{Cs}^+$ , one of the main drawbacks of the adsorbent is related to its ultra-fine particle size, increasing the difficulty to remove the occupied ion exchangers following adsorption [21]. Furthermore,

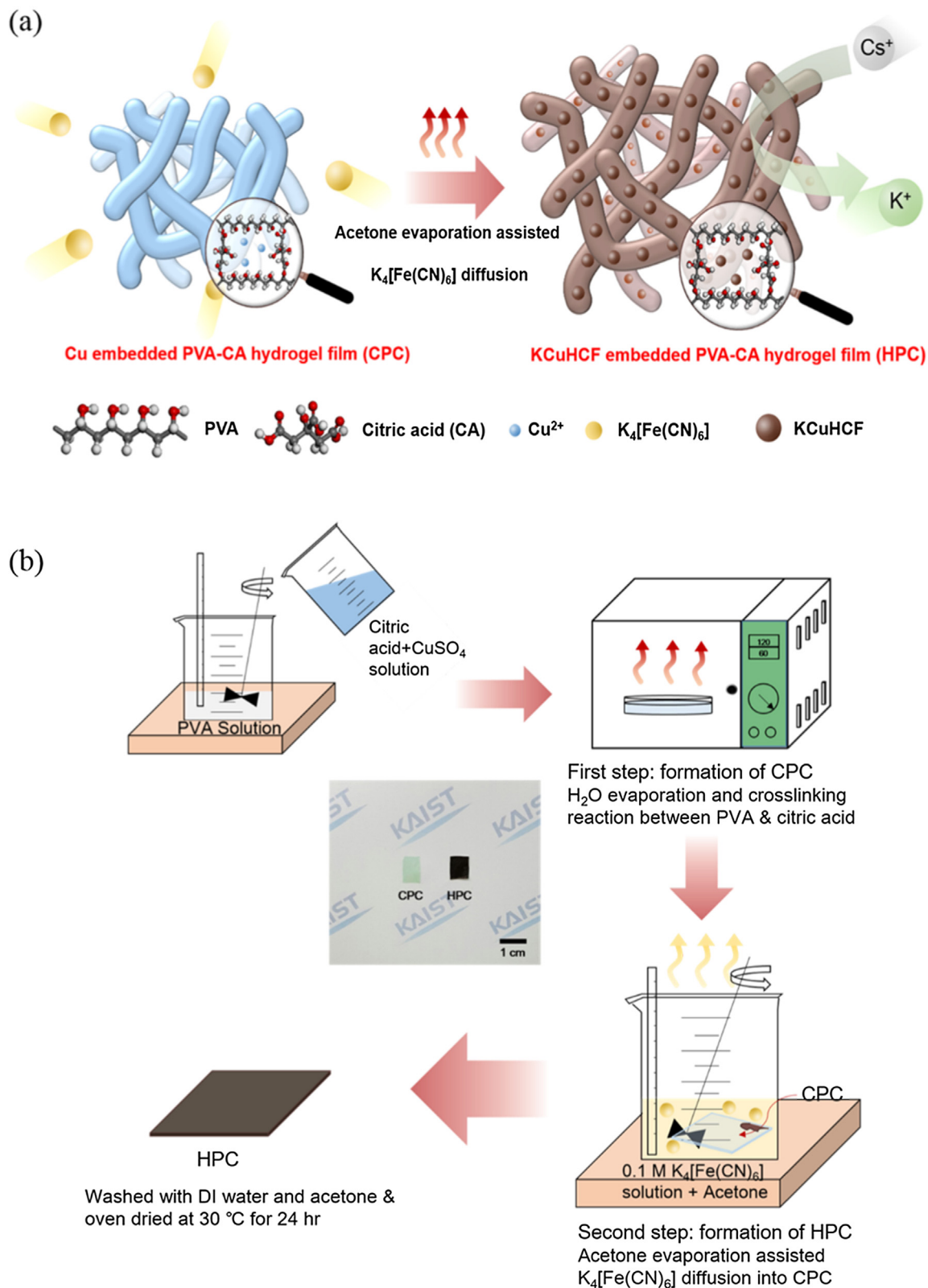


Fig. 1. (a) Schematic representation of CPC and HPC. (b) Two-step synthesis procedure for HPC.

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