PhD Thesis

Name: Abhijit Das (18CH92P01)

Department of Chemical Engineering

Indian Institute of Technology, Kharagpur

Title: Effective and selective adsorption of Uranium (VI) using emerging and novel adsorbents from real-life alkaline leach liquor

Abstract

India has a vast reserve for ore of Uranium (U) in Tummalapalle region, Andhra Pradesh, in the form of carbonate. Very low concentration of U in the ore makes its extraction difficult. Alkaline leaching is the only technique for extraction of U from carbonate ore. However, the major problem for recovery of U is its low concentration in the alkaline leach liquor in presence of large quantity of other salts, like, carbonate, bicarbonate and sulfate. Therefore, innovative and selective adsorbents were developed in this work to address the above issue. Incorporation of APTES functionalized GO in the interlayer space of Ni-Al-LDH was developed to enhance the selective and effective adsorption of uranium. A hyper branched cross-linked polymeric adsorbent (HCPA) was developed using free radical polymerization for selective recovery of uranium(VI) (U) from real-life alkaline leach liquor. Further, zeolitic imidazolate framework-67 (ZIF-67) was used to examine the removal of U from simulated and actual alkaline leach liquor. Moreover, a diethylenetriamine (DETA) functionalized metal organic framework (ZIF-67) incorporated polyacrylonitrile (PAN) bead (MFB) was developed to capture U efficiently from simulated medium and alkaline leach liquor. The porous framework of MIL-100(Fe) was functionalized using malononitrile (MN), through an in-situ Knoevenagel condensation reaction for the selective and effective recovery of U from alkaline leach liquor. The adsorption mechanism between uranium and adsorbents was determined by different methods, including microscopic, spectroscopic, computational, and macroscopic techniques. Overall, the Langmuir capacity of 3-aminopropyl triethoxysilane (APTES) modified sodium dodecyl sulfate (SDS) functionalized graphene oxide (GO) intercalated nickel-aluminium layered double hydroxide (Ni-Al-LDH) was found to be 948, 1,026, and 1162 mg/g at 323, 313, and 303 K, at pH 8.5. The maximum Langmuir adsorption capacity of

hyper branched cross-linked polymeric adsorbent (HCPA) for U was 1012 mg/g at 303 K, pH 8.5, and 98 % of U was removed from alkaline leach liquor. It was also found that with 100 kg of adsorbent and 925 mg/L feed U concentration, the breakthrough time was about 400 h, and this time was reduced to 180 h if the flow rate was increased to 5000 L/day. The synthesized ZIF-67 reveals a significant amount of U uptake capacity (396 mg/g) and remarkable removal percentage for U in both synthetic and alkaline leach liquor (>98 % and 97.4 %, respectively). The Langmuir sorption capability of diethylenetriamine (DETA) functionalized metal organic framework (ZIF-67) incorporated polyacrylonitrile (PAN) bead (MFB) was 835 mg/g, at pH 7 and, 298 K. For real life alkaline leach liquor with U concentration 825 mg/L and 100 to 500 kg of adsorbent, the adsorption column had a breakthrough life of 46 to 108 days with constant flow rate of 500 L/day. The resultant malononitrile (MN) functionalized MIL-100(Fe) exhibited excellent Uranium (U(VI)) removal capacity (i.e., 270 mg/g) at highly alkaline pH (~ 10). Overall, this study provides a detailed description of the design, synthesis and application of various novel, functional adsorbents towards efficient removal of U from real life alkaline leach liquor.

Keywords: *Uranium adsorption*; *Alkaline leach liquor*; *Coordination interaction*; *Layered double hydroxides (LDHs)*; *Polymeric adsorbent*; *Metal organic frameworks (MOFs)*; *Malononitrile*; *Diaminomaleonitrile*; *Desorption*; *Continuous column study*.