A novel hybrid adsorbent composite matrix, polymethacrylic acid-grafted chitosan/bentonite (PMAA-g-CTS/B) was prepared through graft copolymerization reaction of methacrylic acid and chitosan in the presence of bentonite and N,N'-methylenebisacrylamide as cross linker. The composite was characterized using FTIR, XRD, XPS, surface area analyzer and Zeta potential measurements. Batch experiments were conducted to evaluate the efficiency of PMAA-g-CTS/B for the removal of U(VI) and Th(IV) from aqueous solutions. The adsorption behavior of the composite towards U(VI) and Th(IV) from simulated nuclear industry wastewater and seawater was studied using batch process. The adsorption behavior of PMAA-g-CTS/B towards U(VI) and Th(IV) from water and seawater was studied under varying operating conditions of pH, concentration of U(VI) and Th(IV), contact time, and temperature. The effective range of pH for the removal of U(VI) and Th(IV) was 5.0−6.0. Kinetic data followed a pseudo-second-order model. The equilibrium data were correlated with the Langmuir isotherm model. The equilibrium sorption capacity was estimated to be 117.2 for U(VI) and 110.5 mg/g for Th(IV) at 30°C. Adsorption-desorption experiments over four cycles illustrate the feasibility of the repeated uses of this composite for the extraction of U(VI) and Th(IV) from aqueous solutions. The finding of the investigation showed that the composite exhibits high adsorption capacity for U(VI) as well as Th(IV) ions.

**Keywords:** Chitosan; Bentonite; Composite; Graft copolymerization; Adsorption; Desorption; Uranium(VI) and Thorium(IV)