

Uranium Adsorption Mechanism By X-Ray Spectroscopy Technique



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Introduction and Importance

Developing techniques for extracting uranium from seawater is attracting considerable current interest because land-based uranium sources would be depleted by the end of this century. Our ocean contains a very large quantity of uranium (about 1000 times more than terrestrial ores) which is sufficient to support nuclear power production in the next few centuries. Uranium exists in seawater at a low concentration (~3 ppb) and as the very stable uranyl tris-carbonato complex, $UO_2(CO_3)_3^{4-}$. Screening studies conducted in the 1980s with more than 200 functionalized adsorbents showed that sorbent materials with the amidoxime group $RC(NH_2)(NOH)$ were most effective for uranium adsorption from seawater. Recent research efforts in Japan and in other countries are focused on using amidoxime-based adsorbents for extracting uranium from seawater. The amidoxime-based fiber can be prepared by a radiation-induced graft polymerization method which involves electron beam irradiation of polyethylene and acrylonitrile ($CH_2=CH-CN$) grafting onto the polyethylene fabrics. The cyano groups of the grafted polymer are then converted to the amidoxime groups (Figure 1). This type of sorbents show good mechanical strength and high capacity for uranium sorption from seawater in both laboratory and marine experiments..

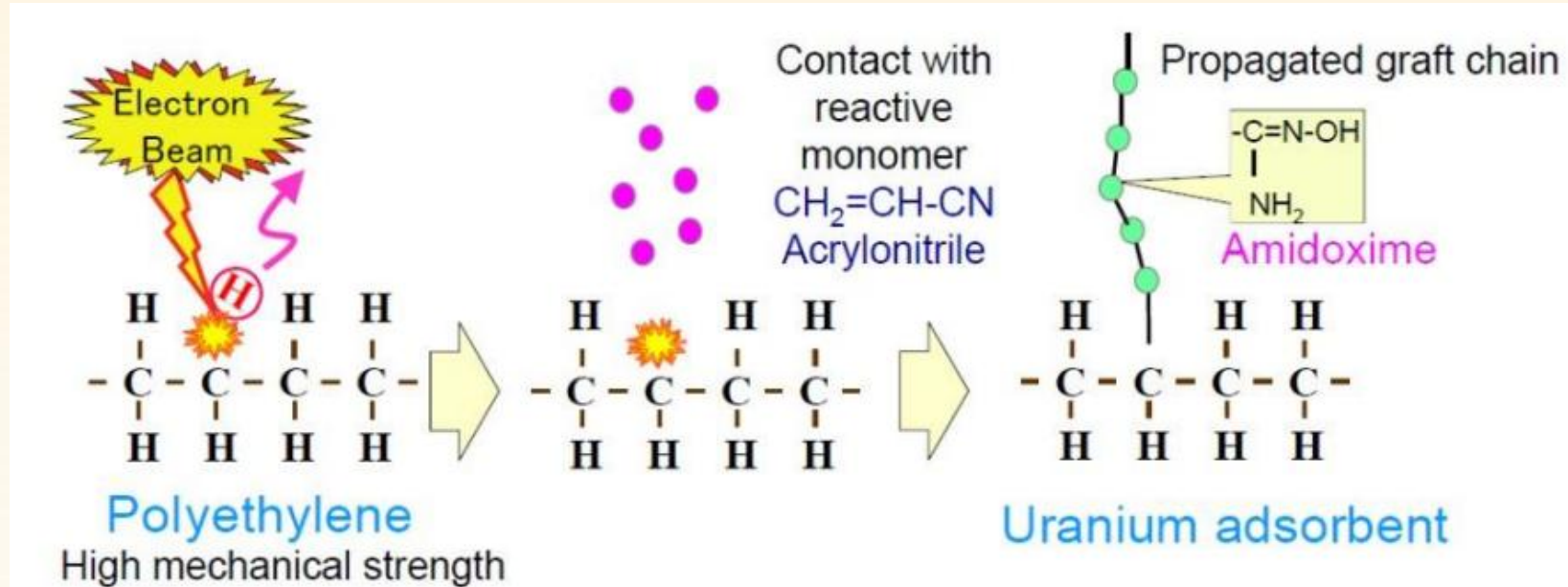


Figure 1. Amidoxime-based sorbents prepared by radiation-induced graft polymerization

Preliminary FTIR Spectra

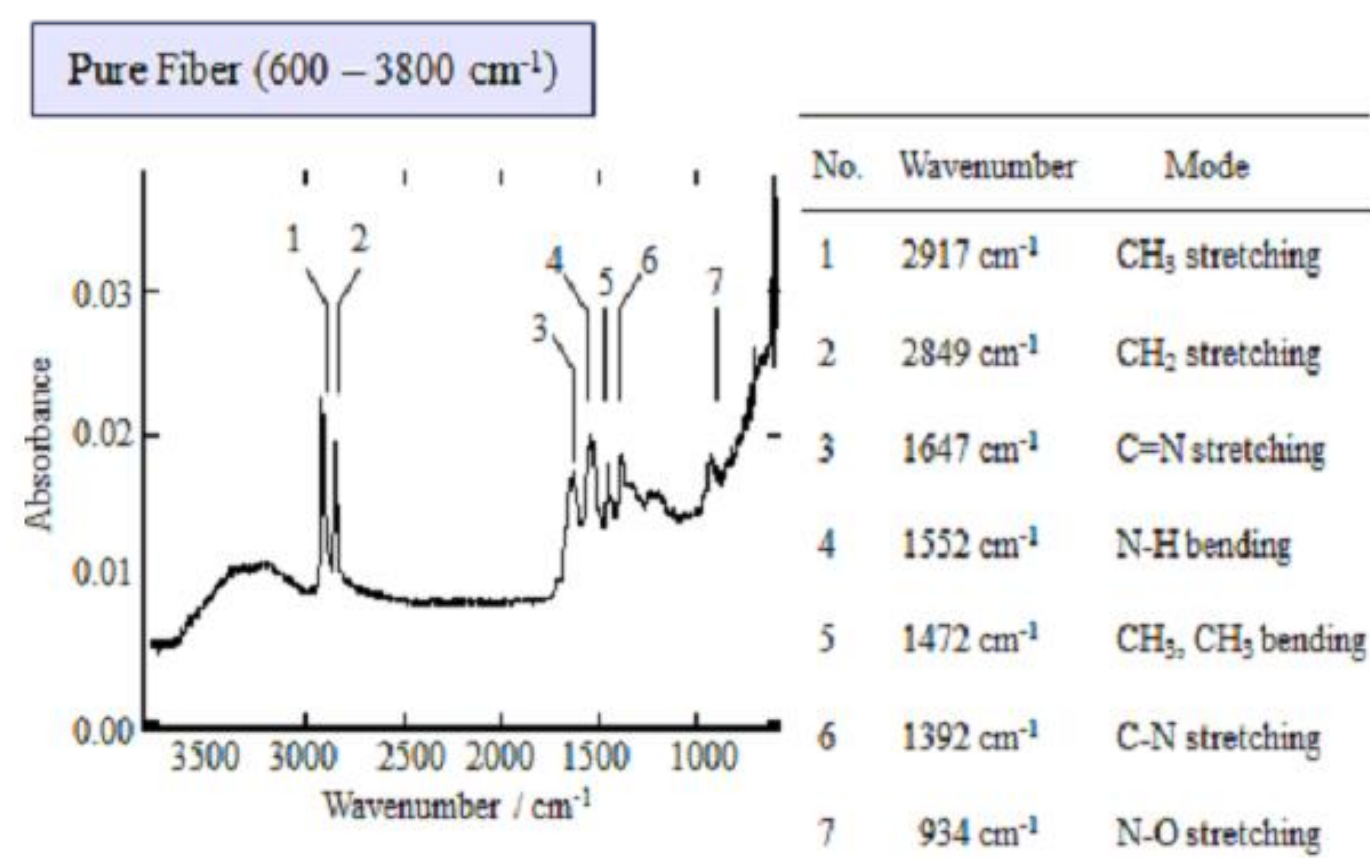


Figure 2. FT-IR spectrum of the original AO sorbent and peak assignments

Figure 3. Ultraviolet spectrum of the standard uranium solution

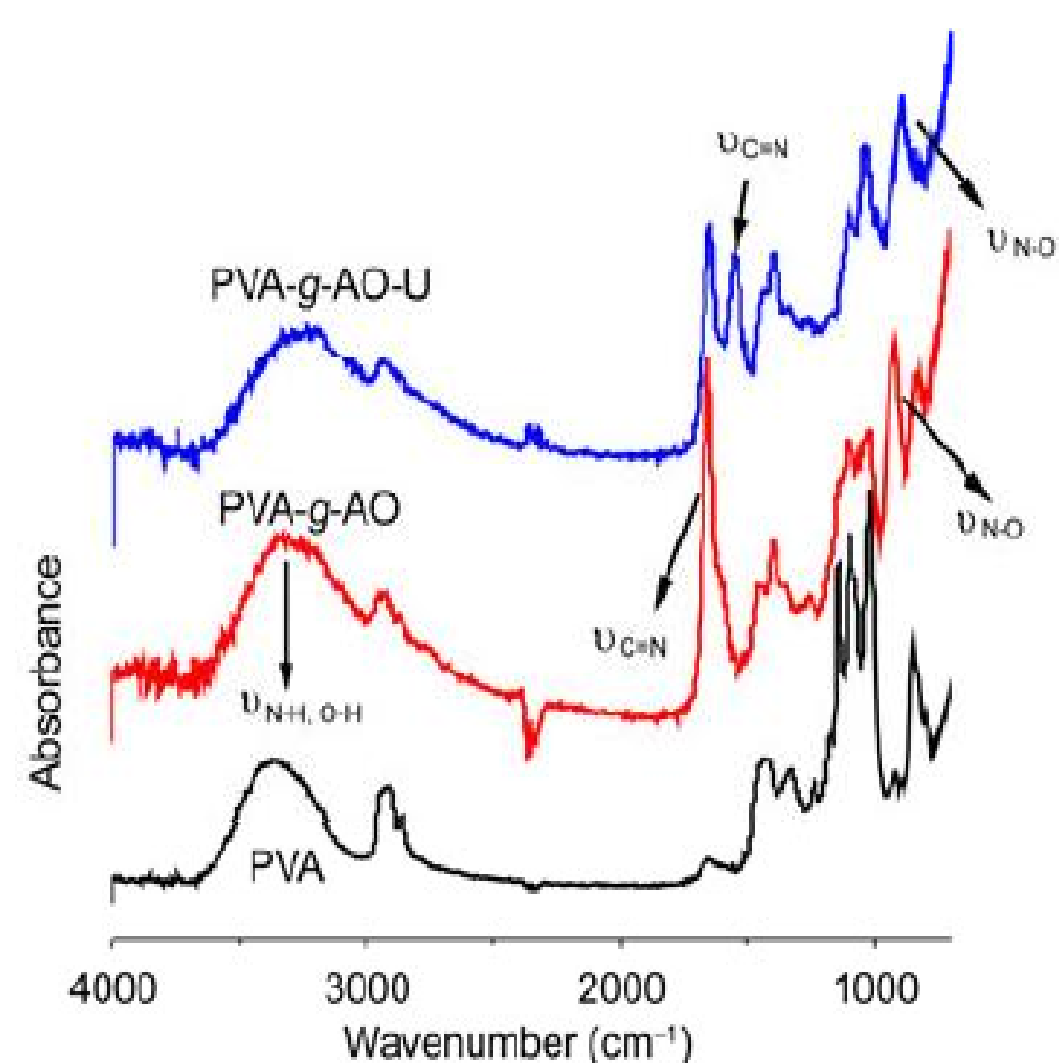
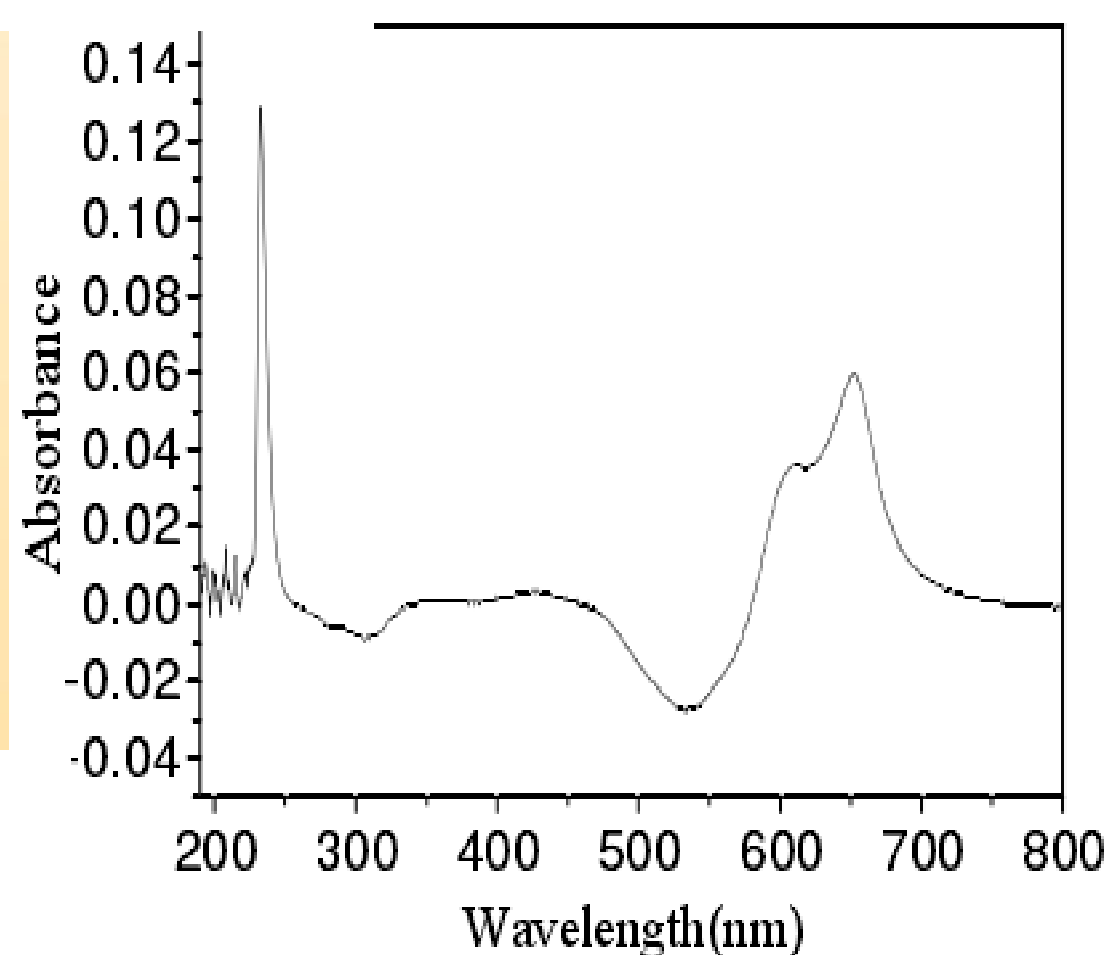


Figure 4. IR spectra of PVA, PVA-g-AO and PVA-g-AO-U

Theoretical Proposed U Complexes with Amidoxime Based Adsorbent

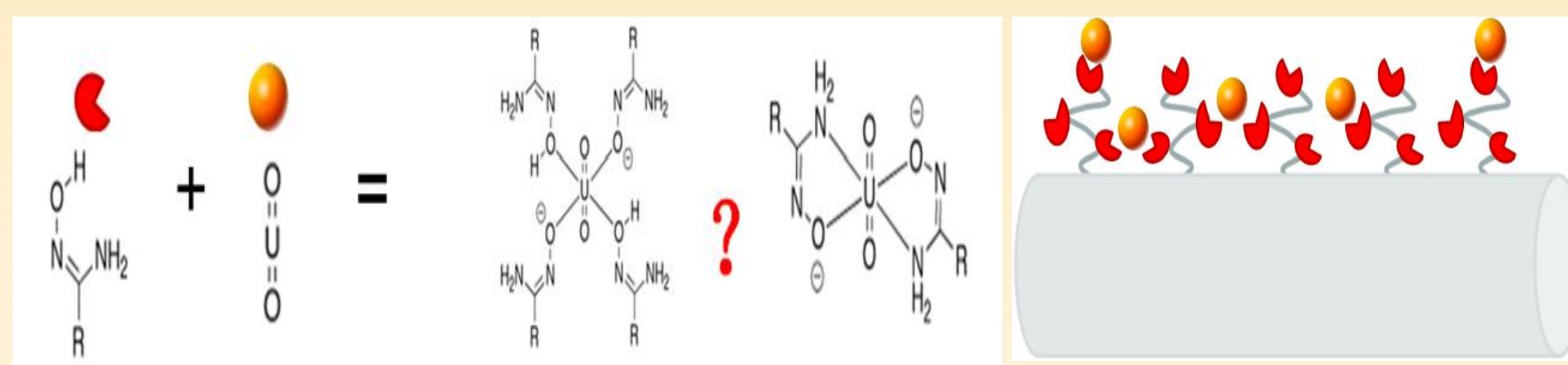


Figure 5. Structure of the amidoxime and amidoximate ligands and UO_2^{2+} binding motifs proposed to occur in amidoxime-based polymers. Solvent molecules complementing fifth and/or sixth coordination sites on the uranyl cation are not shown

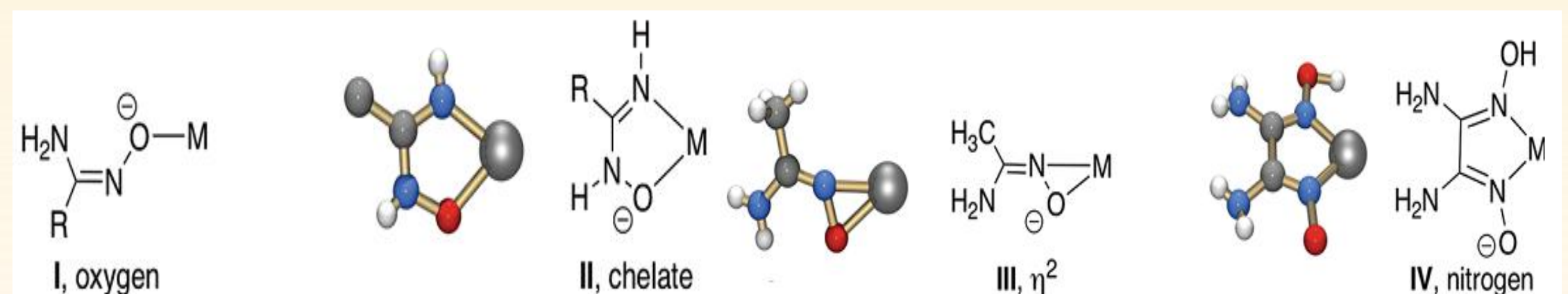


Figure 6. Four possible binding motifs for the amidoximate anion: (I) oxygen bound, (II) chelate, (III) η^2 binding with N-O bond, and (IV) nitrogen bound.

Mechanism and X-Ray Characterization

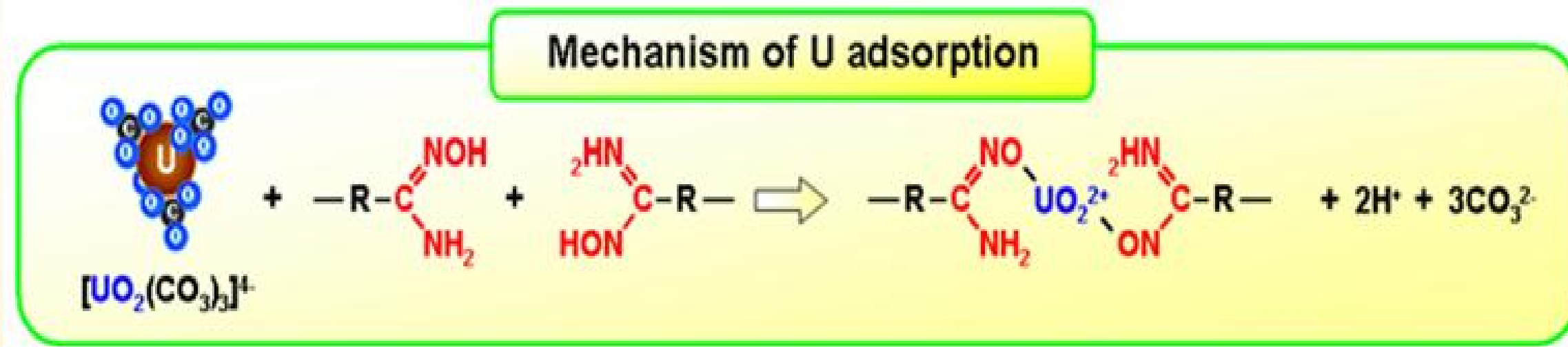


Figure 7. Mechanism of Uranium Adsorption

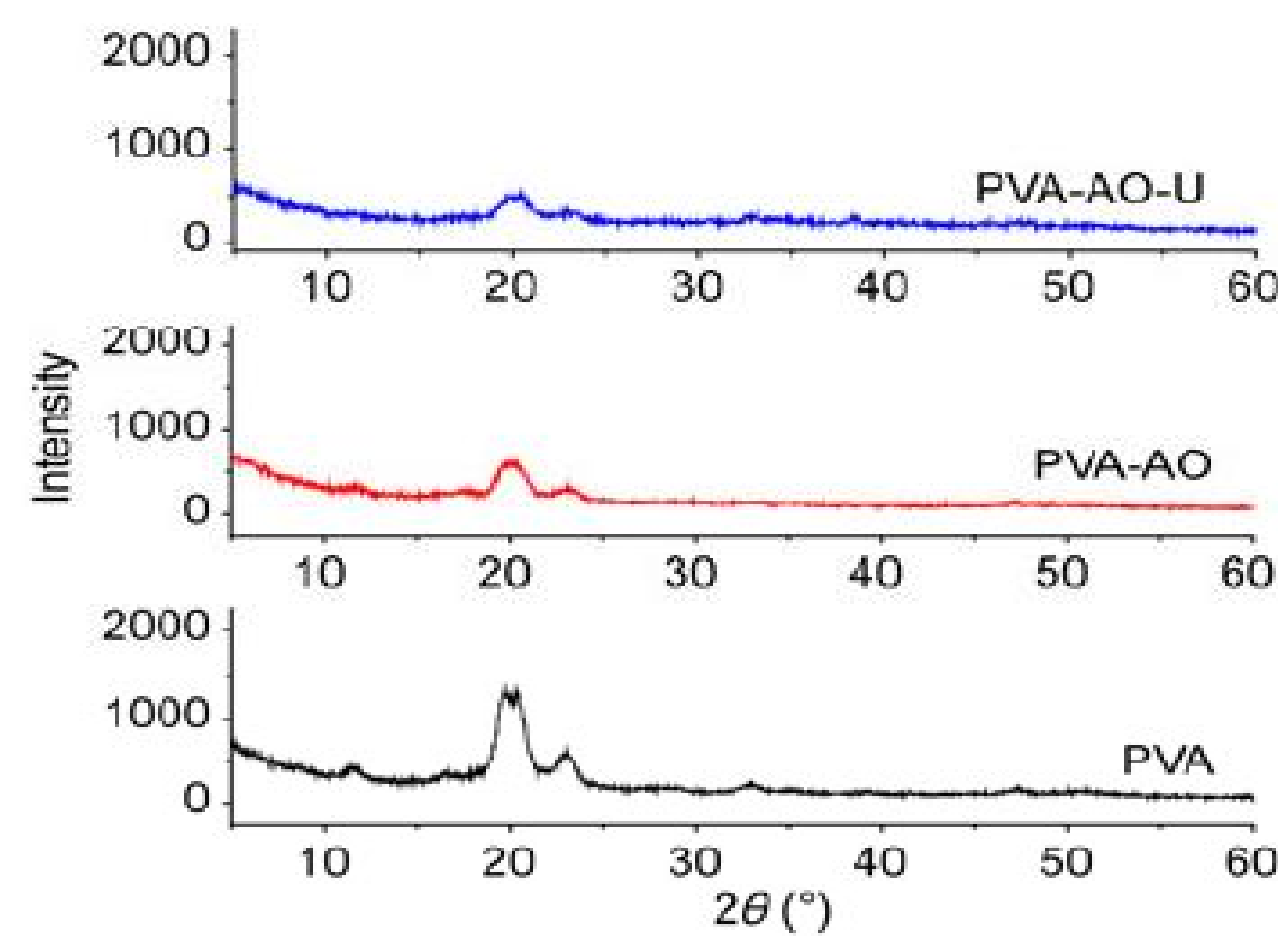


Figure 8. XRD spectra of PVA, PVA-g-AO and PVA-g-AO-U

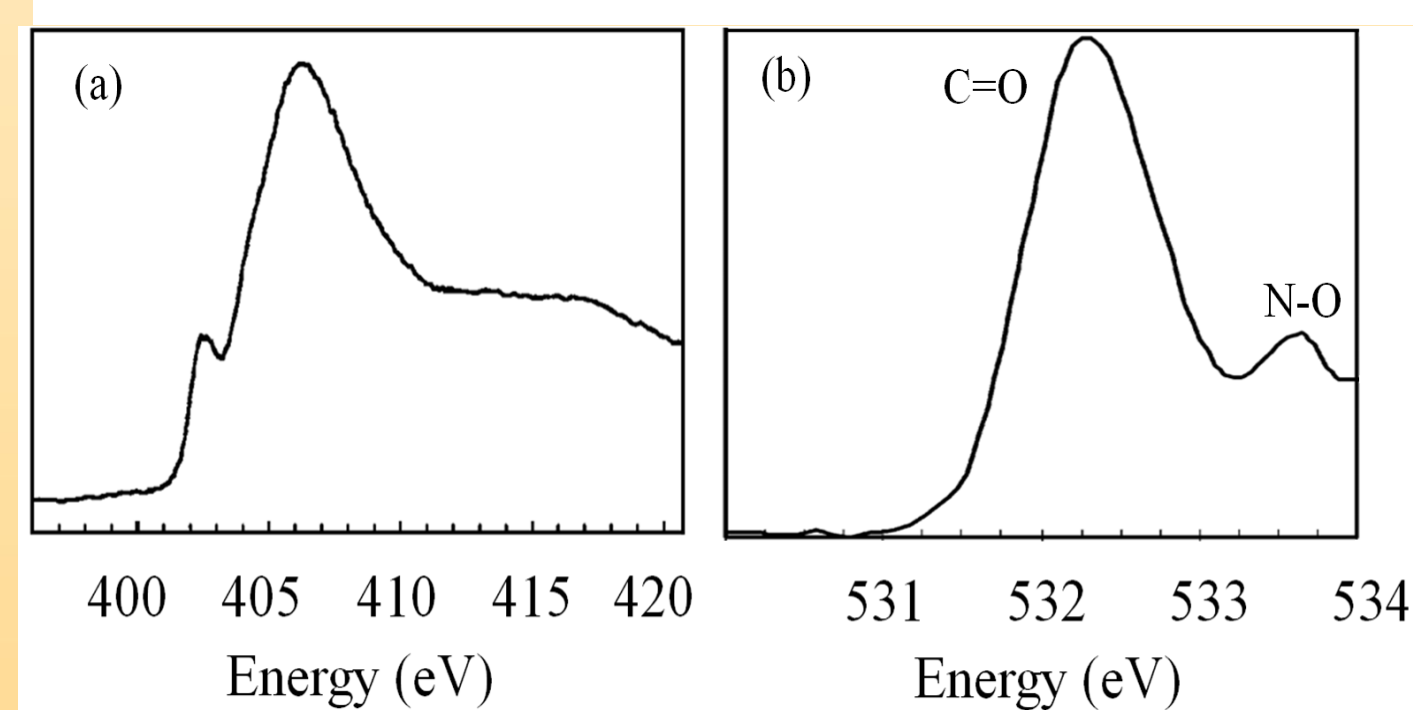


Figure 9. (a) $R-NH_2$ nitrogen edge X-ray absorption spectrum; (b) Molecular oxygen edge absorption spectrum containing N-O bond and C=O bond, the former peak is between 532-533eV, and 533-534eV respectively.

Future Outlook

- Understanding the Coordination Modes, the Sorption Mechanism and Kinetics at the Molecular Level.
- To find the structure between uranyl and AO complex.
- Check the performance of Hyperbranched polymer fibers functionalized with AO group

Acknowledgement: This work is supported by the National Natural Science Foundation of China (11204285 and 51325301) and the experimental fund of Shanghai Synchrotron Radiation Facility (SSRF).

