Molecular size distribution of gamma-ray irradiated humic acid complexed with metal ions

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Abstract: Physicochemical properties of Aldrich humic acid after Co-60 gamma-ray irradiation up to 100 kGy, and then after sequential ultrafiltration to their fractions (<10 kDa, 10-50 kDa, 50-100 kDa, >100 kDa) were analyzed. The metal complexation ability of humic acid in each fraction with Cs^{+}, Sr^{2+}, Eu^{3+} and Zr^{4+} was also discussed.

Keywords: Humic acid, Gamma-ray irradiation, Size distribution, Complexation

1. Introduction

Naturally occurring humic acid (HA) exists in dissolved and suspended forms in natural water systems. In a deep geological repository of HLW, when the HA in underground water reaches to radioactive waste packages, the physicochemical property of HA and its complexation ability with radionuclides leached from the waste might be altered. Quantification of the capacities of carboxylic and phenolic groups in HA, the degree of acid dissociation, and their impacts on metal complexation have been determined [1]. In the present study, we investigate the effect of gamma irradiation on the relationship between molecular size distribution of HA and the complexation ability with metal ions.

2. Experimental

Gamma-ray irradiation of Aldrich HA solution (0.05 mol/dm^3 NaCl) was performed at Co-60 gamma-ray irradiation facility in KURNS. The applied doses at 0.69 kGy/h were 0 (non-irradiation), 1, 5, 10, 50, 100 kGy by controlling the irradiation period. After irradiation, Cs(I), Sr(II), Eu(III) or Zr(IV) was spiked in HA irradiated solutions, then aged for 7 days at room temperature under atmospheric condition. These solutions were separated into four fractions (>100 kDa, 50-100 kDa, 10-50 kDa, and <10 kDa) by a sequential ultrafiltration method, and the concentration of metal ion in each fraction was determined by ICP-MS.

3. Results and discussion

No precipitation was observed in the HA solutions after gamma-ray irradiation while the brownish color of solution gradually turned to lighter with increasing the dose from 0 to 100 kGy. The negative correlation of total organic carbon (TOC) with the dose was clearly observed, suggesting the 0.25 g_organic-C were decomposed by 1 kGy. More than 90% of monovalent Cs ion were mainly present in a fraction of <10 kDa, and the distribution of Cs was independent of the applied gamma irradiation dose. Meanwhile, polyvalent metal ions, Sr, Eu, Zr were found in larger fractions than Cs. And, the major fraction of them shifted to smaller one with dose.

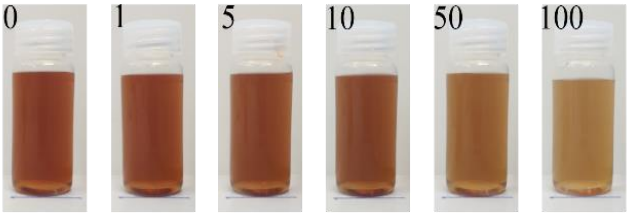

Fig.1 Appearance of irradiated humic acid solutions (0 kGy - 100kGy).

References

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