Contribution ID: **211** Type: **Poster**

Characterization of Czech bentonites B75 and S65 and montmorillonite SAz-1: determination of acid/base titration curves and CEC

Thursday, 15 May 2014 17:30 (1h 15m)

Bentonite clay is proposed to be used as buffer material in final disposal of radioactive waste mainly because of its favorable swelling properties and significant sorption capacity for both cationic and anionic forms of many critical radionuclides present in spent fuel. The sorption properties of clay materials used to be described with two types of sorption sites: layer- and edge-sites. The layer-sites are characterized by their concentration and constants describing ion exchange on them, the edge-sites are characterized also by their concentration, further by two protonation constants.. These parameters, which knowledge will help for evaluation and modelling of interaction of radioactive contaminants with the clay surface, could be obtained by evaluation of acid-base titration curves. Our titration curves were obtained: (i) by semi-continual titration method on the assumption that the equilibrium is reached when the change of electrochemical potential, or pH, in given time-interval (between two sequential additions of titrant solution) is less than 2 mV between the readings after 5 and 10 minutes, (ii) by batch method. The semi-continual method was carried out by automatic titrator TIM 845 and as for the batch method, the influence of contact time (1 to 3 weeks) on the consumption of titrant was studied. In this case, the consumptions of titrant could be influenced by dissolution of the clay material, and therefore, this effect was checked up by back titrations.

Our experiments were carried out in suspension of bentonite in 0.1 M $NaNO_3$ with the initial phase ratio V/m = 250 ml/g. For example, the pretreated bentonite B75, characterized by the producer as a non-activated, was titrated with 0.1M HNO_3 and 0.1M NaOH. The pretreatment of bentonite was done mainly with the aim to eliminate the influence of carbonates on the consumption of titrants. For evaluation of titration curves we develop an original method based on the combination of codes UCODE_2005 and PHREEQCv3. The debugging of this new evaluation method was facilitated by comparison with former approach based on a code prepared in the FAMULUS environment. On the basis of discussion of experimental and calculated data, the recommendations, comprising the optimal experimental conditions for characterization of clay materials by acid-base titration, were formulated.

Dependences of total sorption capacity of bentonites B75, S65 and montmorillonite SAz-1 on pH were compared with values of cation-exchange capacity (CEC) obtained using standardized Cu-triethylenetetramine method. Generally, the CEC corresponds to the summation of two types of charges arising from: (i) isomorphous substitution of cations in the clay crystal structure (permanent charge of layer-sites), (ii) coordination of the cations at the edges of silicate layers (pH-dependent charge of edge-sites). In case of smectites, the pH-dependent charge varies between 10 and 20 % of the total charge.

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Session Classification: Poster Session - Chemistry of Nuclear Fuel Cycle / 1st ASGARD International Workshop

Track Classification: Chemistry of Nuclear Fuel Cycle / 1st ASGARD International Workshop