Accepted Manuscript

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V.L. Vinograd, D.A. Kulik, F. Brandt, M. Klinkenberg, J. Weber, B. Winkler, D. Bosbach

PII: S0883-2927(17)30359-1

DOI: 10.1016/j.apgeochem.2017.11.009

Reference: AG 3991

To appear in: *Applied Geochemistry*

Received Date: 10 August 2017

Revised Date: 27 October 2017

Accepted Date: 23 November 2017

Please cite this article as: Vinograd, V.L., Kulik, D.A., Brandt, F., Klinkenberg, M., Weber, J., Winkler, B., Bosbach, D., Thermodynamics of the solid solution - Aqueous solution system $(Ba,Sr,Ra)SO_4$ + H_2O: I. The effect of strontium content on radium uptake by barite, *Applied Geochemistry* (2017), doi: 10.1016/j.apgeochem.2017.11.009.

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Vinograd, V.L. [1], Kulik, D.A.[2], Brandt, F.[1], Klinkenberg, M.[1], Weber, J. [3], Winkler, B.[4], Bosbach, D.[1]

[1]*Institute of Energy and Climate Research (IEK-6) – Nuclear Waste Management and Reactor Safety, Research Centre Jülich GmbH, 52425 Jülich, Germany*

[2]*Paul Scherrer Institut, Laboratory for Waste Management, 5232 Villigen PSI, Switzerland*

[3]*Chemical Science Division, Oak Ridge National Laboratory,TN 37830, USA*

[4]*Goethe University, Frankfurt, Germany*

Abstract

Thermodynamic properties of mixing in the ternary (Ba,Sr,Ra)SO_4 solid solution are determined using first principles based total energy calculations and Monte Carlo simulations. Two levels of theory, which correspond to the regular mixing and the generalized Ising model, are considered. The results show that the regular mixing parameters increase along the row of Ba-Ra, Ba-Sr and Sr-Ra binary systems proportionally to the squared difference of molar volumes of the end-members. The magnitudes of pairwise interactions similarly increase along the same row, manifesting a tendency to short-range ordering (SRO). In the (Ba,Sr)SO_4 system the SRO effect is approximately equivalent to a 40% decrease in the value of the regular mixing parameter. The ternary solid solution is well described as a regular mixture with the binary parameters W_{BaRa} =