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3 **Solubility Constants of Hydroxyl Sodalite at Elevated**
4 **Temperatures Evaluated from Hydrothermal**
5 **Experiments: Applications to Nuclear Waste Isolation**
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Abstract

In this study, solubility constants of hydroxyl sodalite (ideal formula, $\text{Na}_8[\text{Al}_6\text{Si}_6\text{O}_{24}][\text{OH}]_2 \cdot 3\text{H}_2\text{O}$) from 25°C to 100°C are obtained by applying a high temperature Al-Si Pitzer model to evaluate solubility data on hydroxyl sodalite in high ionic strength solutions at elevated temperatures. A validation test comparing model-independent experimental data to model predictions demonstrates that the solubility values produced by the model are in excellent agreement with the experimental data.

The equilibrium constants obtained in this study have a wide range of applications, including synthesis of hydroxyl sodalite, de-silication in the Bayer process for extraction of alumina, and the performance of proposed sodalite waste forms in geological repositories in various lithologies including salt formations. The thermodynamic calculations based on the equilibrium constants obtained in this work indicate that the solubility products in terms of $m_{\Sigma\text{Al}} \times m_{\Sigma\text{Si}}$ for hydroxyl sodalite are very low (e.g., $\sim 10^{-13} [\text{mol} \cdot \text{kg}^{-1}]^2$ at 100°C) in brines characteristic of salt formations, implying that sodalite waste forms would perform very well in repositories located in salt. The information regarding the solubility behavior of hydroxyl sodalite obtained in this study provides guidance to investigate the performance of other pure end-members of sodalite such as chloride- and iodide- sodalite, which may be of interest for geological repositories in various media.

1. Introduction

Sodalite, with a chemical formula of $\text{Na}_8[\text{Al}_6\text{Si}_6\text{O}_{24}][\text{Cl}, \text{OH}, \text{I}]_2 \cdot x\text{H}_2\text{O}$, is one of the important minerals that precipitates during the Bayer process in production of alumina when bauxite ores are reacted with caustic liquor. The flexible structure of this mineral allows for immobilization of elements of concern. While Na bearing compounds are normally very soluble (Vance and Adl, 1981), sodalite-type compounds including hydroxyl sodalite have relatively low solubilities among the variety of Na compounds. As hydroxyl sodalite can be easily made at relatively low cost, it appears to be poised as a lower-cost alternative for immobilization of radionuclides when compared to other alternatives like vitrification. For example, radioactive iodine (^{129}I) which has a half-life of 15.7 million years can potentially be immobilized in a sodalite waste form. Also, for contaminated electrolytes produced when electrorefining spent nuclear fuel (e.g., Sheppard et al., 2006; Hirabayashi et al., 2012; Lepry et al., 2013; Capone et al., 2014; Maddrell et al., 2014; Gilbert, 2013, 2015), sodalite is a candidate waste form. The reprocessing of spent nuclear fuel for disposition using an electrometallurgical method results in a high-level waste salt with chloride (Priebe and Bateman, 2006, 2008). Zeolite is added to the salt waste which is converted to sodalite at high temperatures ($\sim 900^\circ\text{C}$) (Priebe and Bateman, 2006, 2008). Finally, disposal of sodium from desalination plants is also a huge issue. Since hydroxyl sodalite can be made from the materials considered as industrial wastes such as fly ash and paper-sludge ash (e.g., Zeng et al., 2002; Wajima et al., 2007), hydroxyl sodalite can also be considered to be a viable alternative for disposal of sodium from desalination plants.

80 Additionally, lithium-substituted sodalite ($\text{Li}_2\text{Na}_6[\text{Al}_6\text{Si}_6\text{O}_{24}][\text{Cl}, \text{OH}, \text{I}]_2 \cdot x\text{H}_2\text{O}$) is
81 a candidate waste form for the waste generated by the LiCl-Li₂O oxide reduction (Park et
82 al., 2013) electrochemical process of spent nuclear fuel (UNF) (Lepry et al., 2013):

87
88 It should also be mentioned that because of flexible structure sodalite, a number of
89 sodalite derivatives have been tailored specifically to other wastes such as technetium
90 (and rhenium as an analog to technetium) (e.g., Mattigod et al., 2006; Missmer and
91 Rutherford, 2013; Pierce et al., 2014; Dickson et al., 2015a and 2015b; Luksic et al.,
92 2015), cesium and silver (Borhade and Wakchaure, 2015), and uranium and plutonium
93 (Morss et al., 2000). In most cases, the anion structural site in sodalite would be
94 occupied by the nuclides of interest such as TcO_4^- . In some cases, the cation structural
95 site would be substituted by the nuclides of interest such as contaminated Li(I) and
96 radioactive Cs(I).

97 Finally, hydroxyl sodalite membranes can be used for treatment of waste water
98 (Scott et al., 2002; Zeng et al., 2002; Harja et al., 2013), for production of ultra pure
99 water (Khajabi et al., 2010), and for separation of H₂ and CH₄ (Nabavi et al., 2014). It is
100 also widely used in separation and refinery industries as catalysts, adsorbents, and ion
101 exchangers (Borhade and Wakchaure, 2015).

Salt formations are the ideal medium for disposal of contaminated electrolytes from electrorefining of spent nuclear fuel, and of sodalite-based waste forms. The solutions associated with salt formations are of high ionic strength, and similar solutions are also employed in the Bayer process for extraction of alumina and the process for synthesizing hydroxyl sodalite at elevated temperatures. Therefore, accurate knowledge of solubilities of sodalite in a wide range of ionic strength solutions at elevated temperatures has important bearings on many fields including nuclear waste management.

The objective of this work is to derive solubility constants for the pure hydroxyl end member of sodalite, hydroxyl sodalite (ideal formula, $\text{Na}_8[\text{Al}_6\text{Si}_6\text{O}_{24}][\text{OH}]_2 \cdot 3\text{H}_2\text{O}$), from the solubility data in a wide range of ionic strength solutions at elevated temperatures, by using the high temperature Al-Si Pitzer model (Xiong, 2013, 2014).

2. Evaluating Solubility Constants of Hydroxyl Sodalite at Elevated Temperatures

In this study, the standard state for a solid phase is defined as its pure end-member with unit activity at temperatures and pressures of interest. The standard state of the solvent in aqueous solutions is pure solvent at temperatures and pressures of interest. The standard state for an aqueous solute is a hypothetical 1 molal (m) solution of infinite dilution at temperatures and pressures of interest.

The computer code EQ3/6 Version 8.0a (Wolery et al., 2010; Xiong, 2011) is employed as the modeling platform. The goal of the modeling is to minimize the difference between experimental and model predicted values. A similar modeling approach can be found in Xiong (2013, 2014).

In the modeling, experimental data are used to generate EQ3/6 input files in the first step. In the second step, the values of the targeted parameter (e.g., $\log K$) are changed until the difference between experimental values (e.g., $m_{\Sigma Al}$ and $m_{\Sigma Si}$) and model-produced values predicted by the computer code is minimized. In the third step, the final selected value for the targeted parameter is verified to see how it reproduces experimental data.

The uncertainties reported in this study are two standard deviations (2σ). Error propagations are calculated based on uncertainties associated with regressions and equilibrium constants in the model.

In Table 1 are listed the Al-Si equilibrium constants for temperature of 25 to 250°C. Data were adopted from Xiong (2013, 2014).. Similarly, the Pitzer parameters for the respective species for the high temperature Al-Si Pitzer model are tabulated in Table 2, which is valid to 100°C.

Zeng and Li (2012) conducted systematic solubility studies of hydroxyl sodalite in high ionic strength solutions at temperatures ranging from 30°C to 75°C. The stoichiometry for hydroxyl sodalite is $\text{Na}_{7.26}[\text{Al}_6\text{Si}_{6.12}\text{O}_{24}][\text{OH}]_{1.68} \cdot 2.91\text{H}_2\text{O}$. This stoichiometry slightly deviates from the ideal one. The solubility reaction for the above stoichiometry can be written as follows,

Using EQ3/6 Version 8.0a as the modeling platform, the equilibrium constants for Reaction (3) were derived from the solubility data of Zeng and Li (2012) (see Table 3).

3. Model Verification and Validation

In Figures 1 and 2, solubilities of hydroxyl sodalite as a function of ionic strength at 30°C, 45°C, 60°C and 75°C predicted by the model are compared with the experimental values. As observed the model-predicted values compared favorably well with experimental data, demonstrating the robustness of the model.

In Figure 3, the equilibrium constants for hydroxyl sodalite in the temperature range from 30°C to 75°C are plotted as a function of reciprocal temperatures. It is clear that the relation between log K and reciprocal temperatures is highly linear. According to this linear relation, the equilibrium constants at 25°C, 90°C, and 100°C, are extrapolated (Table 3).

Palmer et al. (2001) conducted solubility experiments on hydroxyl sodalite in 0.5 mol•kg⁻¹ NaOH with 3.0 mol•kg⁻¹ NaNO₃ at 30°C. In Figure 1, their model-independent, experimental data are compared with the model-predicted values. Figure 1 shows that the model-predicted values are in excellent agreement with their experimental values. Palmer et al. (2001) also presented experimental data in 3.0 mol•kg⁻¹ NaOH with 3.0 mol•kg⁻¹ NaNO₃ at 63°C. Their model-independent, experimental data at 63°C are compared with the model-predicted values at 60°C (Figure 2). Figure 2 indicates that the model-predicted values at 60°C are also in excellent agreement with their experimental data at 63°C.

Wajima et al. (2007) obtained solution chemistry for hydroxyl sodalite in high ionic strength NaOH solutions up to 4.5 mol•kg⁻¹ at 90°C. Therefore, their experimental data are used for the validation test for the predicted log K at 90°C and for the model. In the work of Wajima et al. (2007), concentrations were given on the molarity scale. In the validation test, all of their concentrations on the molar scale were converted to molal scale according to the following equation, based on densities of supporting solutions used in experiments at respective temperatures, which are calculated from density equations of Söhnel and Novotný (1985),

$$m_i = \frac{1000 \times M_i}{1000\rho - \sum_i M_i E_i} \quad (4)$$

where m_i is concentration of i species on molality scale, ρ density of solution, M_i is concentration of i species on molarity scale, and E_i the molecular weight of i species.

Figure 4 demonstrates that the solubility data produced by the model are in good agreement with the model-independent values from Wajima et al. (2007), especially in the high ionic strength range. Notice that the hydroxyl sodalite in Wajima et al. (2007) was synthesized from the paper-sludge ash, and hence there were other impurities such as K, Mg, Ca, Fe and Ti, although they were not quantified in solution chemistry in the original work. Therefore, considering the effect of other impurities as they may have different specific interaction effects with Al and Si, the agreement between the model-predicted values and the experimental values is satisfactory in the relatively low ionic strength range.

4 Applications

In this section, the solubility constants obtained in this work are applied to generic repositories in salt formations and granite to evaluate the performance of hydroxyl sodalite up to 100°C. For a generic repository in salt formations, it is assumed that the brine is dominated by sodium and chloride, and is saturated with halite. For instance, the Waste Isolation Pilot Plant (WIPP) brines, GWB and ERDA-6, are dominated by sodium and chloride (Xiong and Lord, 2008). The Na⁺ and Cl⁻ concentrations are 4.04 mol•kg⁻¹ and 6.40 mol•kg⁻¹ for GWB, and 5.35 mol•kg⁻¹ and 5.27 mol•kg⁻¹ for ERDA-6, respectively (Xiong and Lord, 2008). It is also assumed that the hydrogen ion concentration of the brine on a molal scale (i.e., pH_m) is controlled by equilibrium with brucite, as brucite has been proposed as the engineered barrier for the geological repository in a domal salt formation at Asse, Germany (Schuessler et al., 2002), and MgO has been emplaced in the WIPP as the engineered barrier (Xiong and Lord, 2008).

For the calculations performed herein, the equilibrium constants for dissolution of halite at temperatures above 25°C,

were taken from the EQ3/6 database (Wolery, 1992). The equilibrium constants for dissolution of brucite,

the value at 25°C is from Xiong (2008). The values above 25°C are from Brown et al. (1996).

For the first hydrolysis reaction of Mg^{2+} ,

the equilibrium constants above 25°C were from McGee and Hostetler (1975).

In Figure 5, displayed are the predicted solubility products of hydroxyl sodalite in terms of $m_{\Sigma\text{Al}} \times m_{\Sigma\text{Si}}$ as a function of reciprocal temperatures for a generic repository in salt formations. As indicated by Figure 5, the performance of hydroxyl sodalite in such a generic repository is expected to be excellent because of its low solubilities. Its solubility products at 25°C and 100°C are in the range of $\sim 2 \times 10^{-11}$ and $\sim 10^{-13} (\text{mol} \cdot \text{kg}^{-1})^2$, respectively, i.e., its solubility on the order of $\sim 10^{-5.5}$ and $\sim 10^{-6.5} \text{ mol} \cdot \text{kg}^{-1}$, indicating that it is very stable in salt formations (Figure 5).

In Figure 5, the predicted solubility products of hydroxyl sodalite for a generic repository in granite are also shown. The chemical compositions for the solution in such a generic repository are assumed to be similar to those at Olkio, Finland (Table 4) (Pitkänen et al., 2007; POSIVA, 2010), a geological repository in granite. For simplicity, pH_m is also assumed to be controlled by dissolution of brucite in the calculations. This assumption is reasonable, as the reaction of the groundwater with the corrosion product(s) of low carbon steel canisters such as Fe(OH)_2 would make the near-field environments mildly alkaline, favoring formation of brucite. Notice that the groundwater already contains magnesium concentrations in milli-molal range (Table 4). The predicted solubility products of hydroxyl sodalite in terms of $m_{\Sigma\text{Al}} \times m_{\Sigma\text{Si}}$ in a generic repository in granite are higher than those in salt formations (Figure 5). The solubility products at

25°C and 100°C are in the orders of $\sim 5 \times 10^{-8}$ and $\sim 2 \times 10^{-10} \text{ (mol} \cdot \text{kg}^{-1})^2$, respectively, indicating that its solubility is in the order of $\sim 2 \times 10^{-4}$ and $\sim 10^{-5} \text{ mol} \cdot \text{kg}^{-1}$ at 25°C and 100°C, respectively (Figure 5). This would suggest that the performance of hydroxyl sodalite in a generic repository in granite is still good.

The above assessment concerns only the pure end member of hydroxyl sodalite. As the solubilities of other sodalites such as iodide-sodalite are expected to behave similarly to that of hydroxyl sodalite, it can be anticipated that the disposal of them in a generic repository in salt formations would also exhibit excellent performance. However, further studies on other sodalites are an important subject. When the systematic data for other sodalites become available, further model development will be followed.

5 Conclusions

In this study, equilibrium constants for hydroxyl sodalite were obtained up to 100°C by applying the high temperature Al-Si Pitzer model to evaluate the solubility data for high ionic strength solutions, up to $\sim 11 \text{ mol} \cdot \text{kg}^{-1}$. The solubility constants at 30°C, 60°C, and 90°C are validated by the model-independent, experimental data. It is expected that the model equilibrium constants can be applicable in such fields such as synthesis of hydroxyl sodalite for treatment of waste water, the evaluation of the performance of the sodalite waste forms for various nuclear wastes, and de-silication in the Bayer process for extraction of alumina.

Although there are no direct studies for comparing the durability of natural sodalite with that of synthetic sodalite, it would be expected that the durability of synthetic sodalite would be similar to that of natural sodalite. This is because the solubility of synthetic sodalite in terms of Al and Si is close to, or below, the background of the groundwater. For instance, the solution product of $m_{\Sigma\text{Al}} \times m_{\Sigma\text{Si}}$ at 80°C in granite

rock is on the order of $10^{-7} (\text{mol} \cdot \text{kg}^{-1})^2$ (Savage et al., 1992). In comparison, the solubility product of $m_{\Sigma \text{Al}} \times m_{\Sigma \text{Si}}$ for hydroxyl sodalite at 80°C in a generic granite repository is on the order of $10^{-9} (\text{mol} \cdot \text{kg}^{-1})^2$, as indicated by Figure 5.

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Table 1. Equilibrium constants for silica and aluminum species considered in the Al-Si Pitzer model of Xiong (2013, 2014, references therein) up to 250 °C

Reaction	T, °C	log K
$\text{H}_4\text{SiO}_4 = \text{H}^+ + \text{H}_3\text{SiO}_4^-$	25	-9.68 ± 0.14
	50	-9.34 ± 0.16
	75	-9.10 ± 0.17
	100	-8.94 ± 0.16
$\text{H}_3\text{SiO}_4^- = \text{H}^+ + \text{H}_2\text{SiO}_4^{2-}$	25	-13.45 ± 0.07
	50	-12.95 ± 0.25
	75	-12.56 ± 0.25
	100	-12.28 ± 0.25
$2\text{H}_4\text{SiO}_4 = \text{H}^+ + \text{H}_5\text{Si}_2\text{O}_7^- + \text{H}_2\text{O}$	25	-8.50
	50	-8.14 ± 0.25
	75	-7.86 ± 0.25
	100	-7.65 ± 0.25
$\text{H}_5\text{Si}_2\text{O}_7^- = \text{H}^+ + \text{H}_4\text{Si}_2\text{O}_7^{2-}$	25	-10.90
	50	-10.59 ± 0.25
	75	-10.38 ± 0.25
	100	-10.24 ± 0.25
$3\text{H}_4\text{SiO}_4 = 3\text{H}^+ + \text{H}_5\text{Si}_3\text{O}_{10}^{3-} + 2\text{H}_2\text{O}$	25	-29.40
	50	-28.75 ± 0.25
	75	-28.34 ± 0.25
	100	-28.11 ± 0.25
$\text{Al}(\text{OH})_4^- = \text{Al}^{3+} + 4\text{OH}^-$	25	-34.05 ± 0.05
	50	-33.44 ± 0.05
	75	-33.11 ± 0.05
	100	-32.99 ± 0.05
$\text{AlOOH}(\text{cr}) + 2\text{H}_2\text{O} = \text{H}^+ + \text{Al}(\text{OH})_4^-$	100	-12.89 ± 0.04
	150	-12.03 ± 0.05
	200	-11.34 ± 0.05
	250	-10.98 ± 0.06
$\text{Al}(\text{OH})_4^- + \text{H}_2\text{SiO}_4^{2-} = \text{Al}(\text{OH})_3\text{HSiO}_4^{3-} + \text{H}_2\text{O}(\text{l})$	20	-0.42 ± 0.10
	25	-0.42 ± 0.15
	50	-0.38 ± 0.25
	75	-0.35 ± 0.25
	100	-0.33 ± 0.25

Table 2. Pitzer interaction parameters employed in this study

Binary Interaction Parameters				
Interaction Pair	$\beta^{(0)}$	$\beta^{(1)}$	C^ϕ	References
$\text{Na}^+ - \text{H}_3\text{SiO}_4^-$	0.043 ± 0.019	0.24 ± 0.11		Hershey and Millero (1986)
$\text{Na}^+ - \text{H}_2\text{SiO}_4^{2-}$	0.32 ± 0.08	0.13 ± 0.50		Hershey and Millero (1986)
$\text{Na}^+ - \text{H}_5\text{Si}_2\text{O}_7^-$	-0.0571 ± 0.04	0.34 ± 0.13		Xiong (2013)
$\text{Na}^+ - \text{H}_4\text{Si}_2\text{O}_7^{2-}$	-0.0227 ± 0.06	1.56 ± 0.40		Xiong (2013)
$\text{Na}^+ - \text{H}_5\text{Si}_3\text{O}_{10}^{3-}$	0.078 ± 0.03	4.29 ± 0.80		Xiong (2013)
$\text{Na}^+ - \text{Al}(\text{OH})_3\text{HSiO}_4^{3-}$	0.078 ± 0.03	4.29 ± 0.80		Xiong (2013)
$\text{Na}^+ - \text{Al}(\text{OH})_4^-$	0.051	0.25	-0.00090	Wesolowski (1992)
Interaction Involving Neutral Species and Mixing Parameters				
Interaction Pair	λ_{ij}	θ_{ij}	Ψ_{ijk}	References
$\text{Na}^+ - \text{H}_4\text{SiO}_4^0$	0.0925			Azaroual et al. (1997)
$\text{OH}^- - \text{H}_2\text{SiO}_4^{2-}$		-0.0812 ± 0.003 $(\frac{\partial \theta_{ij}}{\partial T})_P =$ $-9.35 \pm 0.44 \times 10^{-5}$		Xiong (2013)
$\text{OH}^- - \text{H}_2\text{SiO}_4^{2-} - \text{Na}^+$			-0.017 ± 0.02	Xiong (2013)
$\text{OH}^- - \text{Al}(\text{OH})_4^- - \text{Na}^+$			-0.04800	Wesolowski (1992)
$\text{Cl}^- - \text{Al}(\text{OH})_4^- - \text{Na}^+$			-0.04857	Xiong (2014)

Table 3. Equilibrium constants of hydroxyl sodalite retrieved from solubility experiments in this study

Reaction	T, °C	log K ⁰
$\text{Na}_{7.26}\text{Al}_6\text{Si}_{6.21}\text{O}_{24.03}(\text{OH})_{1.68} \cdot 2.91\text{H}_2\text{O}(\text{cr}) + 21.12\text{H}_2\text{O}(\text{l}) =$ $7.26\text{Na}^+ + 6\text{Al}(\text{OH})_4^- + 1.26\text{OH}^- + 6.12\text{H}_4\text{SiO}_4(\text{aq})$	25	-60.34 ^A
	30	-61.18
	45	-64.20
	60	-66.69
	75	-68.89
	90	-71.26 ^A
	100	-72.68 ^A

^A Extrapolated values, based on the linear relation between log *K* and reciprocal temperature in Kelvin for hydroxyl sodalite.

Table 4. The chemical compositions of the groundwater for the geological repository in granite at Olkiluoto, Finland*

Total Dissolved Salts (TDS), mg/L	Ionic Strength, mol•kg ⁻¹	pH**	Na ⁺ mol•kg ⁻¹	K ⁺ mol•kg ⁻¹	Mg ²⁺ mol•kg ⁻¹	Ca ²⁺ mol•kg ⁻¹
49,483	1.36	8.2	0.3672	5.0×10 ⁻⁴	0.0015	0.2590
Sr ²⁺ ^A mol•kg ⁻¹	Mn ²⁺ ^A mol•kg ⁻¹	Cl ⁻ mol•kg ⁻¹	SO ₄ ²⁻ mol•kg ⁻¹	ΣH ₄ SiO ₄ mol•kg ⁻¹	ΣCO ₃ ²⁻ mol•kg ⁻¹	ΣAl ³⁺ ^B mol•kg ⁻¹
0.00116	9.5×10 ⁻⁵	0.8783	5×10 ⁻⁵	2.1×10 ⁻⁴	4×10 ⁻⁵	1×10 ⁻²⁰

* The chemical compositions refer to the groundwater from the OL-KR12 borehole at the depth of 708 m taken from Pitkanen et al. (2007) and POSIVA (2010). The original concentrations on molar scale (mol•dm⁻³) are converted to those on molal scale (mol•kg⁻¹) based on the solution density (1.0323 g/mL) calculated from TDS, according to the density model of NaCl solutions in the Fracture Matrix Transport (FMT) code (Novak, 1996).

** In POSIVA (2010), the pH is 8.2. In the model calculations, the pH is assumed to be controlled by brucite.

^A Those components are not inputted for the calculation of hydroxyl sodalite solubility, as they are not supported by the database and do not affect hydroxyl sodalite solubility.

^B Aluminum concentrations are not presented in Pitkanen et al. (2007) and POSIVA (2010). For the purpose of reaction path calculations, an initial concentration of 10⁻²⁰ mol•kg⁻¹ is assigned.

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510 **Figure Captions**

511

512 Figure 1. A plot showing the model predicted solubility of hydroxyl sodalite as a
513 function of ionic strengths at 30°C and 45°C in comparison with experimental values.
514 The size of error bars is equal to or smaller than the symbol size. For the reaction
515 equation, please see Eq. 3 in text.

516

517 Figure 2. A plot showing the model predicted solubility of hydroxyl sodalite as a
518 function of ionic strength at 60°C and 75°C in comparison with experimental values. The
519 size of error bars is equal to or smaller than the symbol size. For the reaction equation,
520 please see Eq. 3 in text.

521

522 Figure 3. A plot showing equilibrium constants of hydroxyl sodalite as a function of
523 reciprocal temperatures in Kelvin.

524

525 Figure 4. A plot showing comparison of model predicted solubilities of hydroxyl sodalite
526 as a function ionic strengths with experimental values, which are independent from the
527 model development. The size of error bars is equal to or smaller than the symbol size.

528

529 Figure 5. A plot showing predicted solubilities of hydroxyl sodalite as a function of
530 reciprocal temperatures for a generic repository in salt formations assuming that brine is
531 dominated by Na^+ and Cl^- and hydrogen ion concentrations (pH_m) are controlled by
532 dissolution of brucite, and for a generic repository in granite assuming that the
533 groundwater compositions are similar to those at Olkiluoto, Finland (see text for details),
534 and pH_m are also controlled by dissolution of brucite.

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Figure 1

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Figure 2

Figure 3

Figure 4

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Figure 5