

The Density of Aqueous Sodium Hydroxide-Sodium Aluminate Solutions: Data Review and Model Development

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ABSTRACT

The density of Hanford tank waste supernatants affects the design and performance of waste treatment processes. The density of aluminate ion $[Al(OH)_4^-]$ bearing sodium hydroxide solutions is important for describing the caustic leaching processes in the Hanford Waste Treatment Plant Flowsheet. The dissolved aluminate has a particularly large impact on the density of supernatants, but this ion is absent from most density estimation algorithms because of its rarity in most industrial processes. Fortunately, there is a large amount of published data on the density of aqueous sodium-hydroxide-sodium aluminate solutions, which can be used to develop density models. This study reviewed the available data and determined the partial molar volume of sodium hydroxide and sodium aluminate for mixtures of these salts in water by regression. This study determined that much of the published data suffered from a strong correlation between the sodium hydroxide and sodium aluminate concentrations in solution. Nonetheless, there was sufficient un-correlated data to identify and quantify the affect of both hydroxide and aluminate concentration on solution density. The density was found to increase linearly with both hydroxide and aluminate concentrations over a wide composition range. The effect of temperature on the density of aqueous sodium hydroxide-sodium aluminate solutions was found to be statistically significant but small in magnitude.

INTRODUCTION

The Hanford Waste Treatment Plant (WTP) is being designed and constructed to treat high-level and low-activity nuclear waste currently stored in underground tanks. The high-level waste is in the form of (hydr)oxide sludges in the Hanford tanks and frequently contains high concentrations of the mineral gibbsite $[\gamma-Al(OH)_3]$ [1]. Gibbsite, not being radioactive, is currently expected to be leached from sludges that contain high aluminum concentrations using sodium hydroxide, to minimize the mass of waste that must be vitrified as high-level waste [2]. The reaction in the leach process is [2]:



where $Al(OH)_3$ is gibbsite, OH^- is the hydroxide ion and $Al(OH)_4^-$ is the dissolved aluminate ion. The resulting solution, containing the dissolved aluminum and remaining sodium hydroxide, is processed as low-activity waste. The density of the solution changes during gibbsite dissolution because aluminum has a large impact on the density of supernatants [3-4]. This density change

needs to be accounted for to appropriately utilize the waste treatment plant vessel volumes. The purpose of this paper is to develop models to predict the density of aqueous sodium hydroxide sodium aluminate solutions. This work was performed for the United States Department of Energy under contract DE-AC27-01RV14136, to support the Hanford Waste Treatment Plant Project.

There are currently many correlations available to predict density as a function of composition and temperature. These correlations are empirical in nature [5]. Therefore, experimentally determined coefficients need to be available to use the models. Unfortunately, most of these correlations do not contain the aluminate ion, which makes them useless for modeling the WTP leaching process. A number of correlations have been developed to model the density of Hanford waste supernatants [3-4]. These models have not been tested for simpler solutions of sodium hydroxide and sodium aluminate expected in the WTP leaching process.

In this paper, simple models of solution density as a function of the concentration of sodium hydroxide, sodium aluminate, and water will be developed by fitting polynomial functions to published density data. The available data will be reviewed for appropriateness for use in these models.

DATA REVIEW

All of the published data on the density of aqueous sodium hydroxide-sodium aluminate solutions that the authors could find was collected and evaluated. The studies evaluated are listed in Table I. Several other reports, not shown in Table I, were also reviewed but discarded without an in-depth evaluation. These discarded reports either lacked the complete composition data, or the concentrations were rounded to two or less significant figures, which is an unacceptable number of significant figures for the purpose here. This section evaluates the statistical design of the datasets in Table I to determine if they are adequate for delineating the effects of composition on density.

Table I. Correlation Coefficients and Temperature Ranges for the Datasets Evaluated.

Source, Reference Number	Correlation Coefficient for NaOH and NaAl(OH) ₄ Concentration (unitless)	Temperature Range (°C)
Baron et al., [6]	-0.222	25-90
Hovey et al., [7]	0.971	10-55
Konigsberger et al., [8]	0.046	50-90
Russell et al., [9]	0.487	40-100
Sanjuan and Michard, [10]	-0.069	25
Sipos et al., [11].	0.49	25

Table I contains the temperature range of each dataset considered as well as the correlation coefficient for NaOH-NaAl(OH)₄ dissolved in water. Correlation coefficients can range from -1 to 1 [12]. A correlation coefficient close to one indicates that the concentration of NaOH increases across samples in proportion to the concentration increases of NaAl(OH)₄ in the dataset.

Similarly, a correlation coefficient close to -1 indicates that NaOH increases as the NaAl(OH)₄ concentration decreases in the dataset. Highly positive or negative correlation coefficients are troublesome when trying to separate out the effects of NaOH from NaAl(OH)₄ on the density, because it is difficult for regression software to distinguish the effect of increasing NaOH when NaAl(OH)₄ is also increasing (or decreasing) at the same rate. Regression coefficients are usually not statistically significant for highly correlated data [12]. Ideally, the correlation coefficient would be zero. For this study, datasets with correlation coefficients between -0.25 and 0.25 were deemed acceptable, and as will be shown below, adequate statistical significance of the model coefficients were achieved using the datasets that meet these criteria. Based on these criteria, the datasets of Baron et al. [6], Sanjuan and Michard [10], and Konigsberger et al. [8] were used in this study. The temperature range covered across these three datasets is 25°C to 90°C, which bounds the temperature range expected in the caustic leach process.

Inspecting Table II, which contains more detailed descriptive statistics for the three selected studies, provides insight into the breadth of the data used in model development and defines the scope of inference for the model. The Sanjuan and Michard [10] dataset contains only low-density liquids. The Konigsberger et al. [8] dataset has some higher density solutions than the Sanjuan and Michard [10] data, but has solution densities that are consistently lower than the solution densities in Baron et al. [6]. Baron et al. [6] reports less significant digits for the measured densities because less precise density measurement techniques were available in 1965. Nonetheless, the Baron et al. [6] data is precise to three decimal places (versus six decimal places for the other two studies), which is enough precision for WTP purposes. Sanjuan and Michard [10], as well as Baron et al. [6], both report data at 25 °C. Baron et al. [6], as well as Konigsberger et al. [8], report data at 50 and 90 °C. Only Baron et al. [6] reports data at 75 °C and only Konigsberger et al. [8] reports data at 70 °C. Given that the data set of Baron et al. [6] contains higher salt concentration data than Konigsberger et al. [8], model coefficients at 70 °C and 75 °C may be somewhat biased relative to other temperatures because they contain a more narrow salt concentration range.

Table II. Characteristics of the Data in Baron et al. [6], Konigsberger et al. [8], as well as Sanjuan and Michard [10].

Source, Reference Number	Densities in g/mL (mean, minimum, maximum)	Temperatures (°C)	NaOH Mass Fraction (mean, minimum, maximum)	NaAl(OH) ₄ Mass Fraction (mean, minimum, maximum)	H ₂ O mass Fraction (mean, minimum, maximum)	Total Number of Data Points
Baron et al., [6]	1.299, 1.108, 1.492	25, 50, 75, 90	0.2188, 0.0663, 0.3534	0.1111, 0.0231, 0.2314	0.6702, 0.4858, 0.8557	110
Konigsberger et al., [8]	1.128497, 1.008470, 1.262522	50, 70, 90	0.0695, 0.0147, 0.1678	0.1087, 0.01125, 0.2793	0.9801, 0.6576, 0.9545	106
Sanjuan and Michard, [10]	1.0168872, 1.002435, 1.043318	25	0.0155, 0.0039, 0.0384	0.0044, 0.0014, 0.0069	0.9801, 0.9554, 0.9944	30

POLYNOMIAL MODEL OF SODIUM HYDROXIDE-SODIUM ALUMINATE SOLUTION DENSITIES

This section will determine if temperature and the concentration of water, sodium aluminate, and sodium hydroxide have a statistically significant impact on the density of the sodium hydroxide - sodium aluminate solutions. The data from the studies selected in Table II was combined and fit to a polynomial equation shown in Equation 2.

$$\rho = a * T + b * MF_{OH} + c * MF_{Al} + d * MF_{H_2O} \quad (\text{Eq. 2})$$

In Equation 2, ρ is the solution density in grams per mL, MF_{OH} , MF_{Al} and MF_{H_2O} equals the mass fractions of NaOH, $NaAl(OH)_4$, and H_2O , respectively, in the solution mixture. The symbol T is temperature in °C. The regression coefficient “a” is in units of grams per mL of solution per °C of the solution. The coefficients b, c, and d are in the units of grams per mL of solution per mass fraction of the component in solution. The coefficients and coefficient statistics determined by multi-linear regression are shown in Table III and the regression statistics are shown in Table IV. The P statistic is a measure of the statistical significance of the regression coefficients, and the smaller the number is, the more statistically significant the coefficient is [12]. A P statistic of less than 0.01 is commonly considered statistically significant. The P statistic of zero for the mass fractions of NaOH, $NaAl(OH)_4$ and H_2O in solution indicates that there is a 100% probability that these coefficients are significant. The P statistic for temperature is 6.6×10^{-169} , which achieves the 0.01 significance criteria by 167 orders of magnitude. Despite the highly significant nature of the temperature coefficient, the magnitude of the coefficient is very small (Table III). This indicates that temperature has a small effect on density, but this small effect is discernable. The R^2 statistic indicates the fraction of the sample variability that is accounted for in the model. The R^2 for the regression is 0.9994, which indicates that the model accounts for more than 99 % of the variability of the data. Thus, the modeling activity can be considered a success and the functional form of the model can be considered adequate.

Table III. Coefficients and Coefficient Statistics for Equation 2.

Coefficient	Value	P-Statistic	Lower 95% Confidence Interval	Upper 95% Confidence Interval
a (T)	-0.00065	4.6×10^{-169}	-0.00067	-0.00064
b (NaOH)	2.1068	0	2.1031	2.1106
c ($NaAl(OH)_4$)	1.7769	0	1.7716	1.7823
d (H_2O)	1.0134	0	1.0122	1.0146

Table IV. Regression Statistics for Equation 2.

Statistic	Value
R^2	0.9994
R^2 Adj.	0.9953
Standard Error	0.00314
Number of Observations	246
Density Range (g/mL)	1.0024 to 1.492
Temperature Range (°C)	25 to 90

ISOTHERMAL DENSITY AND PARTIAL MOLAL VOLUMES

Equation 2 is a simple equation for calculating the density of sodium hydroxide-sodium aluminate solutions. For some applications, the volume of solution for a given mass of waste is needed and the density is simply an intermediate value used in this calculation. For this task, it would be simpler to know the partial molar volume (PMV) for each solution component (NaOH, NaAl(OH)₄, and H₂O) so that the volume can be calculated from the mole fraction of each component and the total number of moles [4]. The PMV of a solute or solvent is the change in volume of the solution per change in the number of moles of the component. The total volume (V_{Total}) of the solution is the sum of the volumes of the individual components (“i’s”), as described mathematically in Equation 3.

$$V_{Total} = total\ moles * \sum PMV_i * mole\ fraction_i \quad (Eq. 3)$$

The component PMVs in salt solutions change somewhat as a function of concentration [5] but are frequently approximately constant over restricted, and sometimes large, composition ranges. Sipos et al. [11] has found that the PMV’s of sodium hydroxide-sodium aluminate solutions are only weakly dependent on solution concentration at 25 °C. Therefore, the PMVs were approximated as constants over the composition range in this study, and this assumption was tested statistically for the precursor variable (density coefficients), as is discussed below.

Preliminary regression analysis of solution volume versus mole fraction (Eq. 3) resulted in partial molar volumes for the three components that were not statistically significant (P values > 0.01). The reason for the insignificance of the coefficients will be speculated below. Plesha [4] also found that regression analysis of Hanford tank waste supernatants resulted in statistically insignificant PMVs, and he overcame this by calculating the PMVs using a numerical technique rather than using direct regression analysis. To overcome the insignificant coefficients in this study, regression analysis was used, however the density per mass fraction of each component (as in Eq. 2) rather than the volumes versus mole fraction was regressed. The density per mass fraction was subsequently transformed into a partial molar volume. This method, as will be discussed below, was found to result in statistically significant coefficients, whereas direct regression of the solution volumes versus mole fraction did not.

The first step in determining the PMVs was to determine the change in density per mass fraction of each constituent. The density data was fit using the following equation (Eq. 4) for each temperature.

$$\rho = a * MF_{OH} + b * MF_{Al} + c * MF_{H_2O} \quad (Eq. 4)$$

Equation 4 is the same as equation 2 except there is no temperature term. The temperature effect was evaluated by fitting the equation at each temperature separately (25, 50, 70, 75 and 90 °C). The coefficients and coefficient statistics are shown in Table V. The regression statistics for the fitting activity are shown in Table VI.

Table V. Regression Coefficients for Equation 4, at 25, 50, 70, 75, and 90 °C.

Temperature (°C)	Coefficient	Value	P Statistic	Lower 95 % Confidence Interval	Upper 95 % Confidence Interval
25	a (NaOH)	2.086	1.7E-112	2.080	2.091
	b (NaAl(OH) ₄)	1.779	9.64E-97	1.770	1.789
	c (H ₂ O)	0.996	2.4E-141	0.995	0.997
50	a (NaOH)	2.079	1.6E-125	2.072	2.086
	b (NaAl(OH) ₄)	1.74	6.2E-112	1.735	1.753
	c (H ₂ O)	0.980	1.7E-143	0.978	0.982
70	a (NaOH)	2.070	2.84E-63	2.055	2.085
	b (NaAl(OH) ₄)	1.704	2.18E-70	1.697	1.71
	c (H ₂ O)	0.971	4.87E-87	0.970	0.973
75	a (NaOH)	2.049	4.81E-46	2.040	2.058
	b (NaAl(OH) ₄)	1.733	1.42E-40	1.720	1.746
	c (H ₂ O)	0.966	1.47E-47	0.962	0.969
90	a (NaOH)	2.045	3.17E-90	2.036	2.055
	b (NaAl(OH) ₄)	1.712	3.11E-77	1.697	1.727
	c (H ₂ O)	0.955	1.3E-101	0.953	0.958

Table VI. Regression Statistics for Isothermal Fitting of Equation 4 to Density Data

Temperature (°C)	25	50	70	75	90
R ²	0.999823	0.999344	0.999257	0.999443	0.998916
R ² Adj	0.98196	0.984173	0.97219	0.953938	0.978873
Standard Error	0.002298	0.003064	0.00187	0.002186	0.00414
Number of Observations	59	69	40	25	53
Density Range in Dataset (g/mL)	1.002 to 1.492	1.032 to 1.476	1.002 to 1.250	1.118 to 1.415	1.008 to 1.450

Inspecting the regression statistics indicates that the R² was greater than 0.99 for all temperatures, and the model coefficients were a function of temperature. The high R² supports the assumption that the change in density with change in composition is essentially linear for these data. The P statistics achieved the 0.01 criteria for all coefficients at all temperatures. Each of these coefficients assumed that the density changed linearly with change in concentration of the component.

In addition to the good statistical fit of the models at each temperature, the results are also consistent with the change in density with change in concentration expected from the observations of others for other salt systems [5, 13]. When one constituent is 100 % of the mass of the solution, the density per mass fraction of constituent “i” (coefficients a, b, and c) reduces to the density of the pure component. Therefore, the density coefficient should be similar to the density of the pure compound. The coefficient for water at 25 °C is 0.996 g/mL (Table V) and the density of pure water at 25 °C is 0.997 g/mL [14]. Thus, the model results are consistent with the expectation that the water coefficient is close to, but not identical to, the density of pure water [5]). Similarly, Mathias [13] indicated that the coefficients for the dissolved salts should be close in magnitude to the density of the pure solid salt. The density of pure NaOH salt at 25 °C is 2.13 g/mL [14], and the density coefficient for sodium hydroxide at 25 °C is 2.086 g/mL. Thus, this result is consistent with the observations of Mathias [13]. The density of pure NaAl(OH)₄ salt has not been reported, but it would be expected to be close to 1.78 g/mL, based on the coefficient for NaAl(OH)₄ in Table V. The small differences between the pure compounds and the density coefficients occur because of compound interactions in solution [5].

The coefficients in Table V slightly decrease with increasing temperature, consistent with the negative value for the temperature coefficient for Equation 2, and with general experience that the density of aqueous solutions decrease with increasing temperature [5]. Figure 1 plots the density coefficient for H₂O as a function of temperature. The density coefficient for water decreases slowly with increasing temperature, as does the density of pure water, but the density coefficient for water in sodium hydroxide-sodium aluminate solutions decreases at a faster rate than pure water. The change in the density coefficient with temperature can be approximated with a polynomial equation.

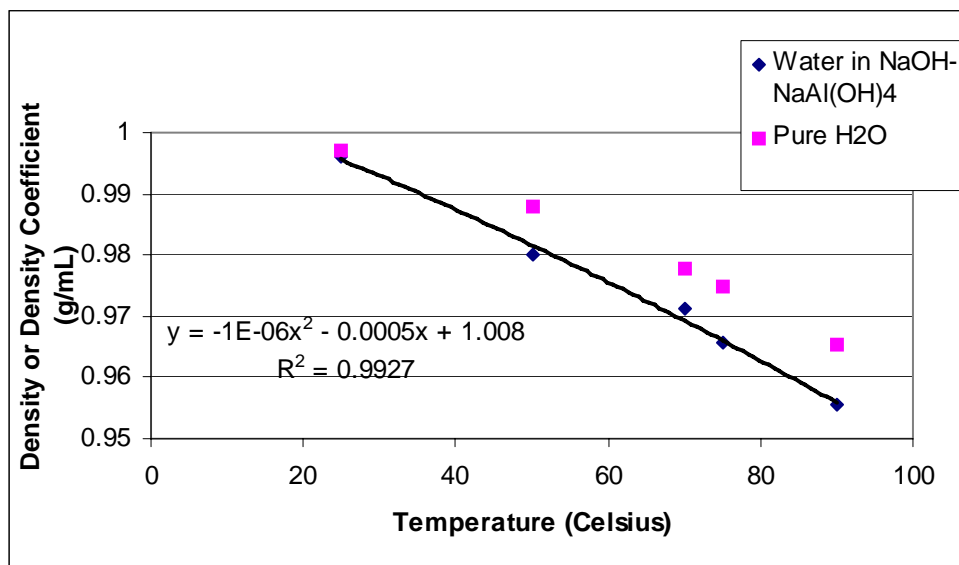


Fig. 1. The Change in the Density Coefficient for Water as a Function of Temperature.

The temperature coefficient for sodium hydroxide also decreases with increasing temperature, as shown in Fig. 2. The change in the coefficient for NaOH with temperature is approximately linear, but there is a large difference between the coefficient at 70 and 75 °C, despite the fact that there is only 5 °C temperature difference between them. This discrepancy is likely because the data at 70 and 75 degrees each came from two different studies, rather than being a true drop in the coefficient over this small temperature interval.

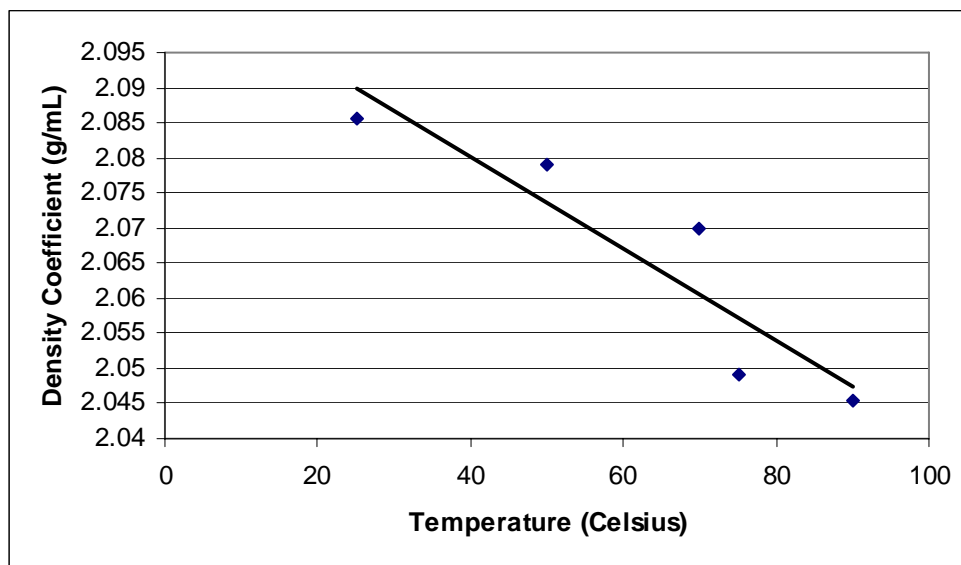


Fig. 2. Change in the Density Coefficient for NaOH with Temperature.

Figure 3 shows the change in the density coefficient for $\text{NaAl}(\text{OH})_4$ as a function of temperature. Like NaOH, the change is approximately linear, but there is a large difference between the coefficients at 70 and 75 °C, despite a small change in temperature.

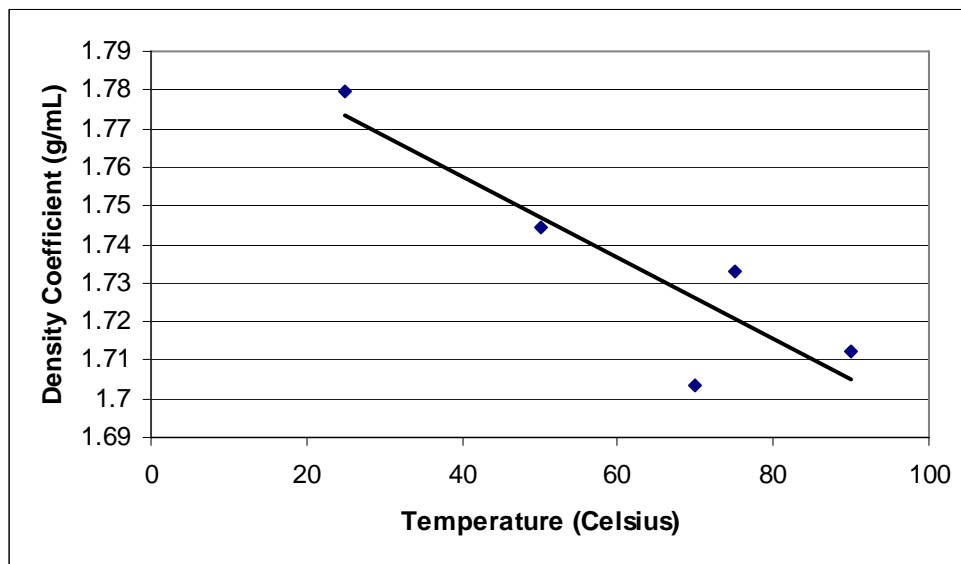


Fig. 3. Change in the Density Coefficient for $\text{NaAl}(\text{OH})_4$ with Temperature.

The density coefficients for each component can be converted into PMVs by dividing the molecular weight of the component by the coefficient (Eq. 5):

$$\left(\frac{g}{mole} \right) / \left(\frac{g}{mL} \right) = mL/mole \quad (\text{Eq. 5}).$$

Equation 5 was solved for each component at each temperature, and the result is shown in Table VII. Table VII also contains the PMV of pure water, and it can be seen that the PMV of pure water is close the PMV of water in the mixture, as observed in other salt solutions [5].

Table VII. The PMV of Pure Water and Aqueous Sodium Hydroxide-Sodium Aluminate Solution Components in Units of mL per Mole.

Temperature (°C)	NaOH	NaAl(OH) ₄	H ₂ O	Pure Water
25	19.18	66.30	18.07	18.05
50	19.24	67.64	18.36	18.22
70	19.32	69.26	18.53	18.41
75	19.52	68.08	18.64	18.46
90	19.56	68.91	18.84	18.65

From inspecting Table VII, it can be speculated why the component coefficients were statistically insignificant when the PMVs were determined by direct regression of Equation 3. The PMV of NaOH is very close the to the PMV of water (Table VII), and NaOH and water make up the majority of the moles of solution. Likely, the small difference in PMV made it difficult to distinguish the effects of NaOH from H₂O. Fortunately, NaOH and H₂O have very different molecular weights; so transforming the data to mass fraction (from mole fractions in Equation 3) emphasized the difference between the two components, allowing the effect of NaOH and H₂O on density to be discerned.

The PMVs in Table VII can be compared to the PMVs calculated by Plesha [4] at approximately 25 °C for Hanford tank waste supernatants that contain NaOH and NaAl(OH)₄ along with many other dissolved sodium salts. Plesha [4] calculated the PMVs for individual ions and water rather than for the salts and water, so the PMVs of the ions in the salts need to be summed together to make them equivalent to the PMVs derived in this study. The PMV of Na⁺, OH⁻, and Al³⁺ derived by Plesha [4] are 11.2, 10.2, and 17.1 mL/mole, respectively. Summing the PMV for Na⁺ and OH⁻ comes to 21.4 mL/mole, which is similar to, but a little bit higher than PMV of NaOH in Table VII (19.18 mL/mol). The Plesha [4] PMV for water is 17.6 mL/mol, which is consistent with, but a little lower than the PMV of 18.07 mL/mol derived in this study for sodium hydroxide-sodium aluminate solutions. Summing the Plesha [4] PMVs for Na⁺, Al³⁺ and four OH⁻'s to make NaAl(OH)₄ comes to 69.2 mL/mol, which is only a little bit higher than the NaAl(OH)₄ PMV in sodium hydroxide-sodium aluminate solutions shown in Table VII (66.30 mL/mole). These comparisons indicate the PMV for mixture components in the sodium hydroxide-sodium aluminate solutions is similar to, but not identical to, the more compositionally diverse Hanford tank waste supernatant solutions.

Hovey et al. [7] reported a PMV of 45 mL/mol for $\text{NaAl}(\text{OH})_4$, which is inconsistent with both this study and Plesha [4]. As noted in Table I, $\text{NaAl}(\text{OH})_4$ and NaOH were significantly correlated in the Hovey et al. [7] dataset, which would have made it difficult to obtain an accurate PMV using that dataset. Indeed, they did not report the statistical significance of their regression coefficients, so it is unclear how accurate their analysis is. Therefore, it is not surprising that the PMV reported by Hovey et al. [7] is in disagreement with the one reported in Table VII.

The PMV of water is usually smaller in sodium salt solutions than in pure water because sodium binds six waters per mole, and the sodium-bound water is more compact than free water [15]. This is consistent with the PMV of water in Hanford Supernatants reported by Plesha [4], who reported a PMV for water of 17.6 mL/mole, which is slightly lower than the PMV of pure water. This is inconsistent, however, with the PMV of water in sodium hydroxide-sodium aluminate solutions, which is larger than the PMV of pure water (Table VII). Buchner et al. [16] reported that the aluminate ion inhibits other ions in solution from binding water molecules. The impact of aluminate on the ability of other ions in solution to bind water is likely a contributing factor to the large PMV for water in sodium hydroxide-sodium aluminate solutions reported in Table VII.

CONCLUSION

Published density data for aqueous sodium hydroxide-sodium aluminate solutions was evaluated. Data was selected for use that had adequate statistical designs for determining component effects on solution density. A density model was fit to this data, and it was found that the density of the solutions changed roughly linearly with the concentration of water, sodium hydroxide, and sodium aluminate. The temperature was also found to have a small but discernable effect on density. The PMV of water, sodium hydroxide, and sodium aluminate were determined. These PMVs were found to be comparable to PMVs reported for Hanford tank waste.

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