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Isopiestic determination of the osmotic and activity coefficients of the  $\{yMg(NO_3)_2 + (1-y)MgSO_4\}$  (aq) system at T = 298.15 K

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## **ACCEPTED MANUSCRIPT**

Isopiestic determination of the osmotic and activity coefficients of the  $\{yMg(NO_3)_2 + (1-y)MgSO_4\}$  (aq) system at T = 298.15 K

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Abstract Isopiestic (vapor pressure) measurements were made for aqueous mixtures of  $Mg(NO_3)_2$  and  $MgSO_4$  at  $T = (298.15 \pm 0.01)$  K with  $Mg(NO_3)_2$  ionic strength fractions y =(0.19691, 0.42542, 0.60113, 0.79583, and 1), along with a separate series of measurements for  $MgSO_4(aq)$ , y = 0, all using KCl(aq) as the reference standard. The resulting 44 molality-based osmotic coefficients for the ternary mixtures, ionic strength range  $I_{\rm m} = (2.5924 \text{ to } 8.4583)$ mol·kg<sup>-1</sup>, were modeled with an extended form of Pitzer's ion-interaction model, both with the usual Pitzer mixing terms and also with Scatchard's neutral-electrolyte model mixing terms, and also with the Clegg-Pitzer-Brimblecombe model based on the mole-fraction-composition scale. The molality-based osmotic coefficients  $\phi$  of these mixtures at each fixed ionic strength fraction fall in a regular order between those of the corresponding limiting binary solutions with no crossovers in this ionic strength range. Model parameters for  $Mg(NO_3)_2(aq)$  and  $MgSO_4(aq)$  at T = 298.15 K needed for these calculations were evaluated by using the present isopiestic results along with other sets of reliable osmotic coefficients gleaned from the published literature. Our osmotic coefficients for the ternary system are compared with those from a previous isopiestic study and a hygrometric study and the present results were judged to be more accurate. For this ternary system, both mixing parameters are needed for the extended Pitzer model with Pitzer mixing terms {standard uncertainty of fit  $u(\phi) = 7.0 \times 10^{-3}$ } whereas three parameters gave a significantly better fit when the Scatchard mixing terms are used {standard uncertainty of fit  $u(\phi)$ =  $4.0 \cdot 10^{-3}$ ; the fits with two Scatchard mixing parameters did slightly better than the fit with both Pitzer mixing terms}, as did the Clegg–Pitzer–Brimblecombe model with three mixing parameters {standard uncertainty of fit  $u(\phi) = 5.9 \times 10^{-3}$ }, with use of the three Scatchard mixing parameters giving the most precise representation of the experimental results. Including higherorder electrostatic mixing terms did not improve the representations but had the opposite effect.

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