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## Tracking nitrite's deviation from Stokes–Einstein predictions with pulsed field gradient <sup>15</sup>N NMR spectroscopy†

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**Predicting the behavior of oxyanions in radioactive waste stored at the Department of Energy legacy nuclear sites requires the development of novel analytical methods. This work demonstrates <sup>15</sup>N pulsed field gradient nuclear magnetic resonance spectroscopy to quantify the diffusivity of nitrite. Experimental results, supported by molecular dynamics simulations, indicate that the diffusivity of free hydrated nitrite exceeds that of free hydrated sodium despite the greater hydrodynamic radius of nitrite. Investigations are underway to understand how the compositional and dynamical heterogeneities of the ion networks at high concentrations affect rheological and transport properties.**

Radioactive wastes stored in underground tanks at the Department of Energy legacy nuclear sites, such as the Hanford Site in Washington State, are comprised in part of solutions of sodium salts of nitrite (NaNO<sub>2</sub>) and nitrate oxyanions, as well as sodium salts of hydroxide (NaOH) and aluminate.<sup>1–4</sup> Studying the structure, stability, and dynamics of these oxyanion solutions is of benefit to ongoing activities to transport the waste out of the tanks for safe processing and long-term storage.<sup>5</sup> Determining the intermolecular interactions of oxyanions with other constituents within concentrated alkaline electrolytes has proved challenging due to the disparate sensitivities of experimental techniques to those interactions.<sup>6</sup> These ambiguities challenge the inclusion or omission of terms defining ion-pair formation in solubility models, such as those used to predict the stability of oxyanion-bearing electrolyte solutions.<sup>7–9</sup> The difficulty is accentuated when attempting to predict the solubility and stability of multicomponent electrolytes approaching the complexity of radioactive waste.<sup>3</sup>

Recent research has sought to address those ambiguities through the application of hydration models to correlate the activity of water with the solubility of NaNO<sub>2</sub> in single- and multi-component aqueous solutions.<sup>10,11</sup> The solubility of concentrated NaNO<sub>2</sub> in aqueous solutions of NaOH has also been measured and the solutions were characterized to assess the extent of OH<sup>−</sup> mediated ion-pair formation.<sup>12</sup> A subsequent study correlated the vibrational properties of NO<sub>2</sub><sup>−</sup> to the rheological and thermodynamic properties of the solution.<sup>13</sup> However, a critical knowledge gap in understanding the properties of NO<sub>2</sub><sup>−</sup> in aqueous solutions is information about the diffusivity of NO<sub>2</sub><sup>−</sup>.

This work quantifies the diffusivity of dissolved, <sup>15</sup>N-enriched NaNO<sub>2</sub> with <sup>15</sup>N pulsed field gradient stimulated echo nuclear magnetic resonance (PFGSTE-NMR) spectroscopy. With the addition of <sup>23</sup>Na and <sup>1</sup>H PFGSTE-NMR experiments, the diffusivity of NO<sub>2</sub><sup>−</sup>, sodium ions (Na<sup>+</sup>), and water (H<sub>2</sub>O) can all be described to compare the relative diffusion coefficients of these species. The diffusion coefficient of NO<sub>2</sub><sup>−</sup> exceeded the diffusion coefficient of Na<sup>+</sup> across a wide range of concentrations, despite the greater hydrodynamic radius and molecular weight of NO<sub>2</sub><sup>−</sup>. To gain insight into the experimental observation, molecular dynamics (MD) simulations of NaNO<sub>2</sub> solutions were performed. Analysis of the trajectories yielded the pair-wise radial distribution functions (RDF), enabling estimation of the hydrodynamic radii in the absence of ion pairing, as well as mean squared displacement-derived diffusion coefficients, and PageRank-derived<sup>14</sup> ion-ion cluster compositions. Lastly, <sup>15</sup>N PFGSTE-NMR spectroscopy was applied to an alkaline electrolyte solution of NaOH and NaNO<sub>2</sub> at a composition relevant to radioactive waste processing.

PFGSTE-NMR spectroscopy implements pairs of spatially dependent magnetic field gradients to label the position of NMR-active nuclei in molecules and track their translational diffusion over millisecond timescales.<sup>15–18</sup> While analysis of NMR-active nuclei such as <sup>1</sup>H, <sup>13</sup>C, and <sup>31</sup>P (among others) is routine due to the nuclei's favorable NMR properties, many quadrupolar nuclei and spin ½ nuclei, such as <sup>15</sup>N, are less

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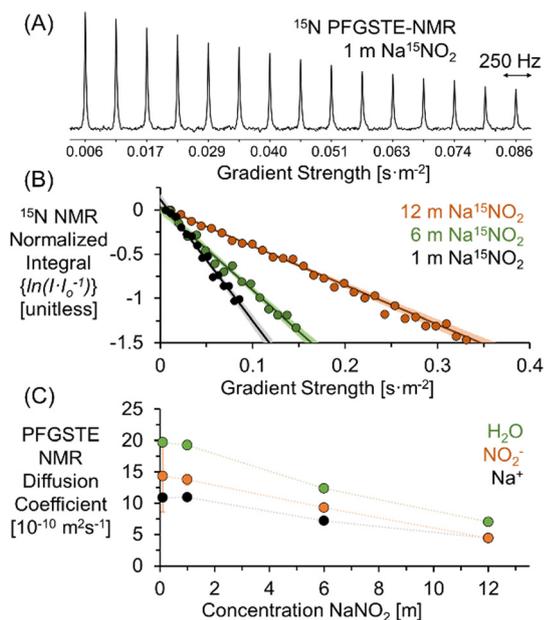
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frequently investigated due to their relatively low gyromagnetic ratio compared to  $^1\text{H}$ , poor natural abundance, or unfavorable spin–lattice and spin–spin relaxation properties.<sup>19–21</sup> Building on others' use of  $^{15}\text{N}$  PFGSTE-NMR to study isotopically enriched  $^{15}\text{N}_2$  diffusion in zeolites,<sup>22</sup> this work overcomes these limitations by using aqueous solutions of isotopically enriched  $\text{Na}^{15}\text{NO}_2$ .

The  $^{15}\text{N}$  NMR spectrum of 98%  $^{15}\text{N}$  enriched  $\text{Na}^{15}\text{NO}_2$  dissolved in  $\text{H}_2\text{O}$  exhibits a single, ensemble resonance across the concentration range of 0.1 to 12 m  $\text{Na}^{15}\text{NO}_2$ . As shown in the ESI,† the  $^{15}\text{N}$  NMR chemical shift of  $\text{NO}_2^-$  increases linearly with a slope of 0.1 ppm  $\text{m}^{-1}$  between 0.1 and 12 m  $\text{Na}^{15}\text{NO}_2$ . As discussed previously,<sup>12</sup> the increase in the  $^{15}\text{N}$  chemical shift of  $\text{NO}_2^-$  can be attributed to a combination of changes in the dielectric constant of the solution, along with perturbation of the local environment of the nitrogen nucleus on  $\text{NO}_2^-$  through changes in the hydration shell or interionic interactions underlying dynamic exchange equilibria at multiple timescales.

Fig. 1A shows the signal attenuation as a function of applied gradient strength in the  $^{15}\text{N}$  PFGSTE-NMR spectra of a solution of 1 m  $\text{Na}^{15}\text{NO}_2$ . For this series of spectra, the total experimental measurement time is approximately 1 day. To quantify the diffusion coefficient, the dependence of the progressive signal decoherence of the NMR resonance is related to the strength of the applied pulsed field gradients. As shown in Fig. 1B, by constructing a Stejskal–Tanner plot, the slope of the log-normal signal intensity is mathematically related *via* the



**Fig. 1** (A)  $^{15}\text{N}$  PFGSTE-NMR spectra of 1 m  $\text{Na}^{15}\text{NO}_2$  in  $\text{H}_2\text{O}$  at 20 °C. (B) Stejskal–Tanner plots of the  $^{15}\text{N}$  PFGSTE-NMR spectra of 1, 6, and 12 m  $\text{Na}^{15}\text{NO}_2$  solutions. The 95% confidence interval of the linear fit is shown in lighter hues. Note that for visual clarity, the Stejskal–Tanner analysis of the  $^{15}\text{N}$  PFGSTE-NMR spectra of the 0.1 m  $\text{Na}^{15}\text{NO}_2$  solution is shown in the ESI.† (C) PFGSTE-NMR results of  $^1\text{H}$ ,  $^{23}\text{Na}$ , and  $^{15}\text{N}$  diffusion coefficients quantifying the diffusion coefficient of  $\text{H}_2\text{O}$ ,  $\text{Na}^+$ , and  $\text{NO}_2^-$  for  $\text{Na}^{15}\text{NO}_2$  solutions at 20 °C.

Stejskal–Tanner equation (ESI†) to the diffusion coefficient of  $^{15}\text{NO}_2$ .<sup>23–25</sup> The data at all concentrations is well fit by a single linear function, facilitating the extraction of an averaged diffusion coefficient<sup>26</sup> attributable to  $\text{NO}_2^-$ .

As reported in Fig. 1C, the diffusivity of  $\text{H}_2\text{O}$  and  $\text{Na}^+$  can be quantitatively and specifically measured with  $^1\text{H}$  and  $^{23}\text{Na}$  PFGSTE-NMR spectroscopy, respectively. In combination with  $^{15}\text{N}$  PFGSTE-NMR spectroscopy, this allows measurement of the diffusion coefficients of the ensembles of  $\text{Na}^+$ ,  $\text{NO}_2^-$  and  $\text{H}_2\text{O}$  species. These diffusivity coefficients were measured from 0.1 to 12 m  $\text{Na}^{15}\text{NO}_2$  in  $\text{H}_2\text{O}$  at 20 °C. For all concentrations, the diffusion coefficients of  $^{23}\text{Na}$  and  $^1\text{H}$  in solutions of  $\text{Na}^{15}\text{NO}_2$  were compared to those measured in solutions of natural abundance  $\text{NaNO}_2$ . As shown in the ESI,† across the entire measured concentration range, the  $^1\text{H}$  and  $^{23}\text{Na}$  diffusion coefficients were in good agreement between the enriched and unenriched samples, indicating the substitution of  $^{15}\text{N}$  for  $^{14}\text{N}$  in  $\text{NO}_2^-$  had little impact on the diffusivity of  $\text{H}_2\text{O}$  and  $\text{Na}^+$ .

As shown in Fig. 1C, a comparison of the experimentally acquired diffusion coefficients for  $\text{NO}_2^-$  and  $\text{Na}^+$  indicates that the diffusion coefficient of  $\text{NO}_2^-$  is greater than  $\text{Na}^+$  at low concentrations and that the relative diffusivities approach unity as the concentration of  $\text{NaNO}_2$  approaches saturation ( $\sim 12$  m). Note that from the MD simulation partial RDF of nitrite in 1 m  $\text{NaNO}_2$ , the hydration number of nitrite is about 11, which is in good agreement with the upper range found in literature (between 6<sup>27</sup> and 11.9).<sup>28</sup> The upper bound of the range far exceeds the number of water molecules (6) solvating  $\text{Na}^+$ .<sup>29</sup>

MD simulations were performed to verify that the relative diffusive coefficients for  $\text{Na}^+$  and  $\text{NO}_2^-$  were in agreement with simulations performed with contemporary forcefield approximations capturing the short and long-range order in  $\text{NaNO}_2$  solutions.<sup>30</sup> The diffusion coefficients of  $\text{H}_2\text{O}$ ,  $\text{Na}^+$ , and  $\text{NO}_2^-$  determined from the calculation of the mean squared displacement of these constituents in MD trajectories of solutions of  $\text{NaNO}_2$  are shown in Fig. 2. As shown in the ESI,† the mean squared displacements reached linearity with respect to simulation time across several hundred picoseconds, facilitating the extraction of the diffusion coefficient *via* linear regression.

Using both experimental measurements and MD simulations, the ratio of the  $\text{Na}^+$  to  $\text{NO}_2^-$  diffusion coefficients can be compared to an approximation based on the hydrodynamic radii of the free hydrated ions. The pairwise RDF between  $\text{Na}^+$  or nitrogen (N) with the solvating water oxygen is shown in Fig. 2. Based on the distance ( $R$ ) to the second local minima, at 1 m  $\text{NaNO}_2$ , the hydrodynamic radii are 4.1 and 2.3 angstroms for  $\text{NO}_2^-$  and  $\text{Na}^+$ , respectively.

As shown in Fig. 3, incorporating the hydrodynamic radii determined from MD simulations into Stokes Einstein equation (ESI†) underpredicts the diffusivity of  $\text{NO}_2^-$  in solution, with both the experimental and the theoretical diffusion coefficients in good agreement at concentrations under 6 m to give a  $D(^{23}\text{Na})/D(^{15}\text{N})$  of 1.3, and converging to a ratio of 1 as the concentration approaches the solubility limit of  $\text{NaNO}_2$  in  $\text{H}_2\text{O}$ . Additional NMR experiments were collected for 1 m  $\text{NaNO}_2$



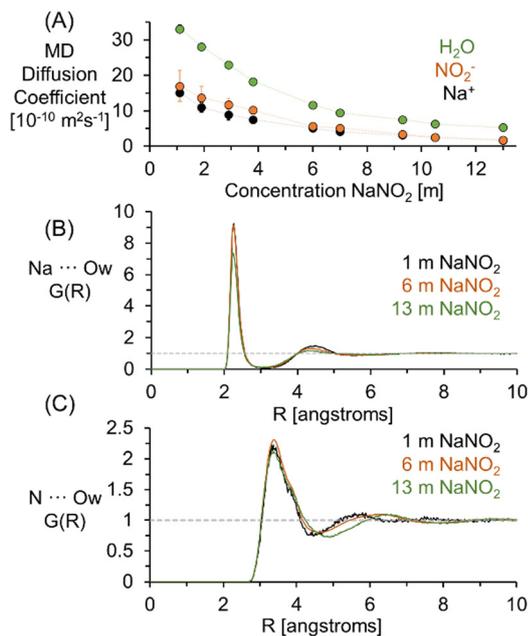


Fig. 2 (A) Diffusion coefficients of  $\text{H}_2\text{O}$ ,  $\text{Na}^+$ , and  $\text{NO}_2^-$  quantified by analysis of the MSD in MD simulations of  $\text{NaNO}_2$  solutions at a temperature of 300 K. (B) The nitrite nitrogen (N) to the oxygen of water (Ow) pairwise RDF. (C) the sodium (Na) to Ow pairwise RDF.

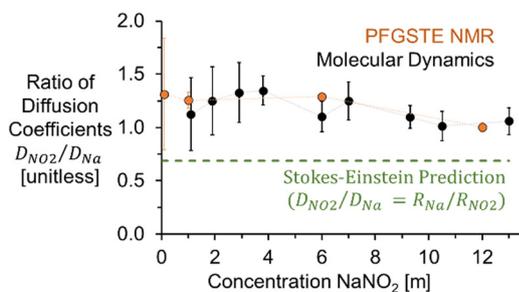


Fig. 3 Experimental and MD simulation based ratios of diffusion coefficients ( $D$ ) of  $\text{NO}_2^-$  and  $\text{Na}^+$  compared to Stokes–Einstein predictions using hydrodynamic radii of free ions.

between 20 and 80 °C. The  $^1\text{H}$ ,  $^{23}\text{Na}$ , and  $^{15}\text{N}$  diffusion coefficients exhibited exponential temperature dependence, as shown in the ESI.†

As shown in Fig. 4, PageRank analyses of the extents of ion–ion cluster formation of sodium nitrite from MD trajectories indicate that for 1 m  $\text{NaNO}_2$  in water, 95% of the nitrite is unassociated with sodium. The enhanced diffusion of the free nitrite relative to free sodium observed in 1 m  $\text{NaNO}_2$  is consistent with the results of others, who have suggested that greater than expected diffusion coefficients of  $\text{NO}_2^-$  are due to the coupling of rotational and translational diffusion modes in asymmetric polyatomic ions for both  $\text{NO}_2^-$  and nitrate, and there were similar observations when comparing the diffusivity of  $\text{NO}_2^-$  with nitrate, perchlorate, and chlorate, all of which exhibit diffusion coefficients at infinite dilution higher than predicted by Stokes–Einstein equation.<sup>31–35</sup>

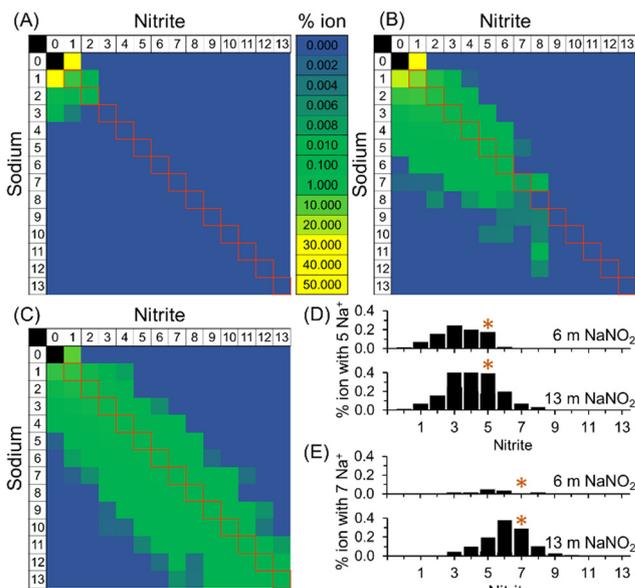


Fig. 4 Histograms derived from PageRank analysis of MD simulations of ion–ion clusters in (A) 1, (B) 6 and (C) 13 m  $\text{NaNO}_2$ . The squares highlighted in red denote the clusters where the number of anions equals the number of cations. Representative slices of the histogram are shown for (D) % ions with 5  $\text{Na}^+$  and (E) % ions with 7  $\text{Na}^+$ . The asterisks denote where the number of anions equals cations.

At high concentrations of 6 and 13 m  $\text{NaNO}_2$ , PageRank analyses of the molecular dynamics trajectories indicate that the ions form ion–ion clusters. When an ion is in ion–ion clusters, its diffusion is likely restricted, with the average observed diffusion coefficient of  $\text{Na}^+$  and  $\text{NO}_2^-$  in these species approaching equivalence. As shown in Fig. 4, particularly at 6 and 13 m  $\text{NaNO}_2$ , the relative proportions of free ions and ion–ion clusters were found to vary for cations and anions. This is highlighted in Fig. 4, where the distribution of the composition of ion–ion clusters observed with MD simulations is slightly skewed towards sodium rich ion–ion clusters. Further work is underway to develop techniques to probe the compositional and dynamical heterogeneities of these species.

Extension of  $^{15}\text{N}$  PFGSTE-NMR spectroscopy to concentrated  $\text{NaOH}$  archetypical of radioactive waste solutions at the Hanford Site in Washington State requires the application of pulsed field gradients of sufficient strength to measure diffusion in solutions with viscosities much greater than that of dissolved  $\text{NaNO}_2$  in  $\text{H}_2\text{O}$ .<sup>36,37</sup> Despite this potential complication, measurement of  $^{15}\text{N}$  diffusion in 2 M  $\text{NaOH}$  is possible, as shown in the ESI,† resulting in a Stejskal–Tanner plot well fit by a single linear function attributable to an ensemble. For this sample emulating the concentration of  $\text{NaOH}$  found in radioactive waste at the Hanford Site, the  $^{15}\text{N}$  diffusion coefficient of  $\text{NO}_2^-$  of  $10.4 \pm 0.5 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$  was also found to be greater than the  $^{23}\text{Na}$  diffusion coefficient of  $7.4 \pm 0.3 \text{ m}^2 \text{ s}^{-1}$ , coinciding with a ratio of  $1.4 \pm 0.1$  for  $D_{\text{NO}_2^-}/D_{\text{Na}}$ . The similarity between the ratio of the diffusivity coefficients in single component  $\text{NaNO}_2$  solutions and multicomponent  $\text{NaOH}/\text{NaNO}_2$  solutions may indicate that, in alkaline solutions, the composition of



the ion networks is weighted towards  $\text{OH}^-$  and  $\text{Na}^+$ . The oxyanions predominantly exist as free ions.

In conclusion, implementing PFGSTE-NMR spectroscopy to the  $^{15}\text{N}$  nucleus enables direct and isotopically specific measurement of the diffusion coefficient of  $^{15}\text{N}$ . These measurements are possible across a wide range of concentrations, with a feasible lower concentration limit established as 0.1 m of  $^{15}\text{N}$ , below which the experimental time for a single measurement exceeds two days. Quantifying the diffusion coefficient of  $\text{NO}_2^-$  enables the determination of the relative diffusion coefficient of  $\text{NO}_2^-$  to  $\text{Na}^+$  in solution. In solutions of 1 M  $\text{NaNO}_2$  and 2 M  $\text{NaOH}$  in  $\text{H}_2\text{O}$  of relevance to nuclear waste stored at the Hanford Site, the relative diffusion coefficient of  $\text{NO}_2^-$  is greater than that of  $\text{Na}^+$ , in contrast to predictions based on the hydrodynamic radius of  $\text{NO}_2^-$  and  $\text{Na}^+$ . Comparative molecular dynamics simulations of  $\text{NaNO}_2$  in  $\text{H}_2\text{O}$  also led to the extraction of a greater diffusion coefficient of  $\text{NO}_2^-$  versus  $\text{Na}^+$ . These results indicate that the coupled translational and rotational motion potentially enhance diffusion relative to  $\text{Na}^+$ . Further investigations into the structure and dynamics of  $\text{Na}^+$  and  $\text{NO}_2^-$  are underway to understand how the compositional and dynamical heterogeneities of the ion networks affect the rheological properties across multiple length and time scales. Such studies could integrate PFGSTE-NMR spectroscopy alongside other NMR techniques, such as NMR relaxometry and observations of heteronuclear Overhauser effects between amenable nuclei.

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## Conflicts of interest

There are no conflicts to declare.

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