Atomic radii of noble gas elements in condensed phases

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ABSTRACT

Neutral atomic radii of dissolved noble gas elements in condensed phases are obtained by treating the neutral atoms as "ions" of zero oxidation state and by interpolation from a plot of radius vs. oxidation state for isoelectronic ions. The major assumption is that the radius of an ion or a neutral atom having an electronic configuration of a noble gas element depends primarily on the interaction between the nucleus and the surrounding electrons and not on the interaction between the ion and its neighbors. As expected, the values of the new set of radii are slightly greater than the univalent radii and smaller than the radii of noble gas atoms in their crystals (in which the coordination number is 12). The neutral atomic radii (in ångströms) of noble gas elements are

<table>
<thead>
<tr>
<th>CN</th>
<th>He</th>
<th>Ne</th>
<th>Ar</th>
<th>Kr</th>
<th>Xe</th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>0.90 ± 0.06</td>
<td>1.18 ± 0.02</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>6</td>
<td>1.08 ± 0.06</td>
<td>1.21 ± 0.02</td>
<td>1.64 ± 0.03</td>
<td>1.78 ± 0.03</td>
<td>1.96 ± 0.02</td>
</tr>
<tr>
<td>8</td>
<td>—</td>
<td>1.32 ± 0.03</td>
<td>1.77 ± 0.06</td>
<td>1.87 ± 0.06</td>
<td>(2.03 ± 0.06 )</td>
</tr>
</tbody>
</table>

where CN = coordination number. Errors are given at the 2σ level. Values in parentheses were determined by extrapolation and have greater errors. The larger error for the He radius is due to large uncertainty in the H⁺ radius.

INTRODUCTION

The sizes of noble gas atoms partially control their solubility (e.g., Doremus, 1973, p. 121-128; Jambon et al., 1986; Carroll and Stolper, 1991, 1993) and diffusivity (e.g., Doremus, 1973, p. 128-145; Jambon, 1982; Fortier and Gilette, 1989; Carroll, 1991; Zhang et al., 1991; Carroll et al., 1993) in water and silicate melts and glasses. They are also important in understanding glass structures (e.g., Shackelford and Masaryk, 1978; Shackelford and Brown, 1981) and in molecular dynamics simulations (Angell et al., 1988). In these applications, it is often necessary to compare (1) the sizes of different noble gas atoms, such as He and Xe, (2) the sizes of noble gas atoms and those of other molecular species, and (3) the sizes of the noble gas atoms and those of holes in a condensed phase (liquid and solid) structure. Therefore, it is important to know the radius of noble gas atoms dissolved in silicate glasses and melts.

However, a review of the literature reveals many different sets of radii (Table 1). Among those listed in Table 1 are univalent radii (Pauling, 1927), radii of noble gas atoms in their low temperature FCC (face-centered cubic) crystals, van der Waals radii, three sets of covalent radii (with one set probably wrong, see reference 5 in Table 1), and atomic radii calculated from viscosity data, second virial coefficients, equations of state, and liquid volumes (for references, see Table 1 Note). Each set of radii for noble gas elements may be internally consistent and hence useful under certain conditions. However, it is not obvious whether any one of these sets applies to the case of noble gas atoms dissolved in condensed phases. Lacking such a set, authors have somewhat arbitrarily chosen different sets of radii. For example, in discussing the dependence of diffusivity and solubility on molecular size, Doremus (1973) used radii obtained from viscosity data (set 7 in Table 1); Jambon et al. (1986) used atomic radii from a handbook of chemistry and physics (which were the univalent radii; set 1); Zhang et al. (1991) used the neutral atomic radii of Forsythe (1954) for noble gas elements (also the univalent radii; set 1); Carroll (1991) used the radii of noble gases from cell constants of FCC crystals (the same as set 3); Carroll et al. (1993), however, used the univalent radii (set 1). The arbitrary selection of atomic radii in these applications casts doubt on the results. The purpose of this work is to derive a set of neutral atomic radii for noble gas elements that is consistent with ionic radii so that atomic sizes can be compared with ionic and hole sizes in condensed phases.

DERIVATION ATOMIC RADIUS OF NOBLE GASES

Numerous studies of ionic radii or bond lengths in crystal structures have been reported (e.g., Goldschmidt, 1926; Pauling, 1927, 1947; Ahrens, 1952; Shannon and Prewitt, 1969, 1970; Whittaker and Muntus, 1970; Bader et al., 1971; Shannon, 1976; Gibbs, 1982; Finger and Gibbs, 1985; Gibbs et al., 1987; Bartelmehs et al., 1989). Radii of ions in a crystal structure are readily available. However, no radii of neutral atoms dissolved in con-
Table 1. Radii (in ångströms) of noble gas elements given in the literature

<table>
<thead>
<tr>
<th>Ref.</th>
<th>Description</th>
<th>He</th>
<th>Ne</th>
<th>Ar</th>
<th>Kr</th>
<th>Xe</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>univalent radii</td>
<td>0.93</td>
<td>1.12</td>
<td>1.54</td>
<td>1.69</td>
<td>1.90</td>
</tr>
<tr>
<td>2</td>
<td>radii in crystal lattice</td>
<td>1.78</td>
<td>1.60</td>
<td>1.92</td>
<td>1.98</td>
<td>2.18</td>
</tr>
<tr>
<td>3</td>
<td>van der Waals radii</td>
<td>1.22</td>
<td>1.60</td>
<td>1.91</td>
<td>1.98</td>
<td>—</td>
</tr>
<tr>
<td>4</td>
<td>covalent radii</td>
<td>0.325</td>
<td>0.69</td>
<td>0.97</td>
<td>1.135</td>
<td>1.31</td>
</tr>
<tr>
<td>5</td>
<td>covalent radii</td>
<td></td>
<td>0.78</td>
<td>1.05</td>
<td>1.21</td>
<td>1.41</td>
</tr>
<tr>
<td>6</td>
<td>covalent radii</td>
<td></td>
<td>1.31</td>
<td>1.74</td>
<td>1.89</td>
<td>2.09</td>
</tr>
<tr>
<td>7</td>
<td>radii from viscosity (700 °C)</td>
<td>1.0</td>
<td>1.2</td>
<td>1.6</td>
<td>2.1</td>
<td>2.45</td>
</tr>
<tr>
<td>8</td>
<td>radii from viscosity (0 °C)</td>
<td>1.1</td>
<td>1.3</td>
<td>1.33</td>
<td>2.09</td>
<td>2.45</td>
</tr>
<tr>
<td>9</td>
<td>radii from viscosity (high-T)</td>
<td>0.91</td>
<td>1.12</td>
<td>1.5</td>
<td>1.6</td>
<td>1.78</td>
</tr>
<tr>
<td>10</td>
<td>radii from viscosity</td>
<td>1.35</td>
<td>1.4</td>
<td>1.71</td>
<td>1.8</td>
<td>2.02</td>
</tr>
<tr>
<td>11</td>
<td>radii from 2nd virial coeff.</td>
<td>1.32</td>
<td>1.37</td>
<td>1.70</td>
<td>1.98</td>
<td>—</td>
</tr>
<tr>
<td>12</td>
<td>radii from eq. of state</td>
<td>1.33</td>
<td>1.19</td>
<td>1.48</td>
<td>1.58</td>
<td>1.62</td>
</tr>
<tr>
<td>13</td>
<td>radii from liquid V</td>
<td>2.00</td>
<td>—</td>
<td>2.02</td>
<td>2.24</td>
<td>2.22</td>
</tr>
<tr>
<td>14</td>
<td>radius ratio</td>
<td>0.634</td>
<td>0.756</td>
<td>1.000</td>
<td>1.092</td>
<td>1.209</td>
</tr>
</tbody>
</table>

Note: references and comments: 1 = Pauling (1927). The same set of radii has been called ionic radii for +1 ions by Dean (1985, p. 3–121) and Weast (1983, p. F-170) and neutral atomic radii by Forsythe (1954, p. 643); 2 = Cook (1961, p. 13) and references therein. Radii in crystal lattice are calculated from lattice parameters of low temperature noble gas crystals (FCC structure). 3 = Dean (1985, p. 3–121). This set is essentially the same as set 2 (except for a different and more reasonable radius for He, 1.22 Å). 4 = Allen and Huheey (1980). These radii are based on extrapolated electronegativity. This set of radii is only slightly different from the radii given in Sanderson (1963, 1967). 5 = Bankowski (1966). The radii of Kr and Xe are based on bond lengths in their fluoride compounds and the radii for Ne and Ar are from extrapolation. 6 = Dean (1985, p. 3–121) and Emsley (1989). The original source is not clear but these radii are close to the bond lengths between noble gas atoms and F atoms in noble gas fluorides (Cotton and Wilkinson, 1988; Emsley, 1989) and are probably misnomers for bond lengths. 7 = Doremus (1973, p. 133). The original source is not clear, but this set of radii is not the same as any of the three sets of viscosity-based radii listed in Cook (1961, p. 13; sets 8–10 in this table). 8–13 = Cook (1961, p. 13) and references therein. 14 = Carlton (1985). The ratio of the radius of a noble gas atom over that of an Ar atom in noble gas van der Waals molecules for the case of equal forces.

Because only cation-anion ionic distances are determined directly, there are some practical difficulties in defining ionic radii. The difficulty has led to the publication of different sets of ionic radii, such as crystal radius vs. effective ionic radius (Shannon and Prewitt, 1969). Crystal radii are established with insight afforded by electron density maps and wave function calculations (Shannon and Prewitt, 1969) and thus may represent more closely the absolute values of ionic radii. However, the effective ionic radii of Shannon and Prewitt (1969), revised by Shannon (1976), are most often cited. For example, Scordari (1992) recently used effective ionic radii to discuss ionic crystal structures. We show later that the effective radii give more reasonable trends than the crystal radii when radii of isoelectronic ions are plotted against oxidation state.

To estimate atomic radii of noble gas elements, a neutral atom dissolved in a condensed phase is viewed as an "ion" with an oxidation state of zero. The effective ionic radii of isoelectronic ions from Shannon (1976) are plotted against their oxidation state for coordination numbers 4, 6, and 8 in Figure 1A–1E. For a specific coordination number (CN), the radii of isoelectronic ions vary smoothly with oxidation state, as expected. However, we note two anomalies: (1) the radius of \([{}^{14}]{\text{B}}^{3+}\) is smaller than that of \([{}^{14}]{\text{C}}^{4+}\), and (2) the radius of \([{}^{56}]{\text{Cr}}^{6+}\) is smaller than that of \([{}^{60}]{\text{Mn}}^{7+}\). (Values of noble gas atomic radii are not affected by the small anomalies.) Radii of noble gas atoms treated as ions of zero oxidation state are then obtained by interpolation (Fig. 1A–1E). For example, the radius of a Ne atom with CN = 6 was found by interpolating to zero oxidation state along the smooth curve of radii vs. oxidation state of \([{}^{15}]{\text{N}}^{-}\), \([{}^{17}]{\text{O}}^{-}\), \([{}^{19}]{\text{F}}^{-}\), \([{}^{25}]{\text{Na}}^{+}\), \([{}^{27}]{\text{Mg}}^{2+}\), \([{}^{55}]{\text{Al}}^{3+}\), \([{}^{57}]{\text{Si}}^{4+}\), \([{}^{23}]{\text{P}}^{5+}\), \([{}^{35}]{\text{S}}^{6+}\), and \([{}^{37}]{\text{Cl}}^{7+}\) with CN = 6; all these ions have the electronic configuration of 1s\(^2\) 2s\(^2\) 2p\(^6\), the same as Ne.

The approach described above assumes that the ionic radius is determined by the interaction between the nucleus and the electrons of the ion. The interaction between cations and anions is ignored. Another issue of concern is the choice of different sets of ionic radii, which is found to be unimportant. The ionic radii of cations and anions must sum to measured interatomic distances. Therefore, if the anions are assigned smaller radii, the cation radii must be increased by the same amount. For example, the ionic radii given by Whittaker and Muntus (1970) are greater by 0.08 Å for cations and smaller by 0.08 Å for the anions than the effective ionic radii of Shannon (1976). The crystal radii given by Shannon (1976) are greater than the ionic radii by 0.14 Å for cations and smaller by 0.14 Å for anions. The noble gas atomic radii obtained using the present method are almost independent of the set of radii one chooses because the interpolation point lies right between the cations and anions. For instance, the atomic radius of Ne is interpolated to be between the ionic radii of Na\(^+\) and F\(^-\). That is,

\[
r_{\text{Ne}} \approx 0.5(r_{\text{Na}^+} + r_{\text{F}^-})
\]
Fig. 1. Plot of effective ionic radii (Shannon, 1976) vs. oxidation states for isoelectronic ions (A–E) and crystal radii (Shannon, 1976) vs. oxidation state (F). CN is the coordination number. Ionic radii of noble gas atoms (zero oxidation state) are obtained by interpolation of the smooth curve fit. (A) Ions isoelectronic with He (the radius of H− is taken to be 1.4 Å for CN = 6 and 1.22 Å for CN = 4; Shannon, 1976); (B) ions isoelectronic with Ne; (C) ions isoelectronic with Ar; (D) ions isoelectronic with Kr; and (E) ions isoelectronic with Xe. (F) A plot of crystal radii (Shannon, 1976) vs. oxidation state for ions isoelectronic with Ne. Compare F with B. The trend in B is expected, whereas the trend in F is unexpected.

where \( r \) is the radius. The summation \( (r_{Na^+} + r_{F^-}) \) is the distance between the two ions in a crystal structure and is independent of the definition of ionic radii. Hence the radius of Ne obtained by interpolation is also roughly independent of the choice of ionic radii. For example, if the crystal radii of Shannon (1976) are used, the difference in noble gas atomic radii is \( \pm 0.03 \) Å. Even though crystal radii may more closely represent the actual size of ions because they are partly based on electron density mapping, we choose not to use crystal radii of Shannon (1976) because the trend in crystal radii vs. oxidation states is unexpected for isoelectronic ions. For example, the crystal radius of F− is about the same as that of Na+ (Fig. 1F). This is unexpected for the two ions with the same number of electrons because the charge of the nucleus of F− is +9e whereas that of Na+ is +11e. The
radius of Na⁺ ion is expected to be smaller because of the stronger attraction of electrons by the nucleus.

A Stineman function was used to smooth the radius data and interpolate the radius of a noble gas atom (Fig. 1A–1E). The errors were estimated in several ways. One was by moving the radius value up and down in the fitted curve and visually checking the fit. The other was by comparison of the radius obtained by the smooth fit with that obtained by simple linear interpolation using only the +1 and −1 ions. Such a linear interpolation is clearly too simplistic, and we believe that the error is slightly smaller than the radius difference between the smooth fit and the linear interpolation. The third method was by comparison of fitting results using different sets of radii (for example, effective ionic radii vs. crystal radii discussed next). Though none of these methods is sophisticated, we nevertheless report our estimate of the errors.

The neutral atomic radii for He, Ne, Ar, Kr, and Xe, with different coordination numbers, and their associated errors are given in the abstract. The ratios of these radii agree reasonably well with relative radii (i.e., radius ratios) derived by Carlson (1985) from scaling of repulsive interatomic forces in noble gas van der Waals molecules (set 14 in Table 1). The neutral atomic radii of noble gas elements with CN = 6 based on this work are greater than the covalent radii of Allen and Huheey (1980; set 4 in Table 1) by ~0.7 Å and the univalent radii of Pauling (1927; set 1) by ~0.1 Å, as expected. The radii we obtained are smaller than the van der Waals radii of noble gas atoms with CN = 12 (Dean, 1985, p. 3–121; Cook, 1961, p. 13; set 3) by ~0.25 Å. This difference is also expected since radii increase with coordination number. The radii we have derived are similar to those of Doremus (1973) for He, Ne, and Ar but not for Kr and Xe. The differences are likely due to an imperfect understanding of the relation between viscosity and atomic radius, judging from the four different sets of atomic radii that were all developed using viscosity data (sets 7–10 in Table 1).

It is concluded that the differences between this set of neutral atomic radii for dissolved noble gas elements in condensed phases and other sets of radii are consistent with current understanding of the expected trends in ionic radii. Therefore, the radius data derived from this work are reasonable and useful in several applications. First, the size effect on diffusivity and solubility of noble gas elements can be studied. Second, the atomic radii are directly comparable to the ionic radii of Shannon (1976) and thus can be used to compare the sizes of noble gas atoms dissolved in condensed phases with sizes of ions and holes in condensed phases. Third, they are useful in determining whether a molecule is greater or smaller than a noble gas atom if the size of the molecule can be cast in terms of ionic radii of Shannon (1976), such as H₂O molecules discussed later. However, caution must be exercised in comparing the sizes of different noble gas atoms with polyatomic and often nonspherical molecules such as N₂ (linear), O₂ (linear), CO₂ (linear), CH₄ (approximately spherical), etc., unless the radii of these molecules can be obtained using ionic radii. If this cannot be done, the best comparison would probably be to use the b parameters in the van der Waals equation of state for gases, except for the anomaly that the b parameter of He is greater than that of Ne (Weast, 1983, p. D-194; see radius sets 2 and 12 in Table 1).

**Effect of Molecular Size on Diffusion**

The atomic radii derived above for noble gases can be used to examine the effect of size on diffusion of neutral species in silicate glasses. The activation energy for diffusion in silicates has been suggested to follow approximately the following relation (Doremus, 1973; Jambon, 1982):

\[ E = E₁(r - r₀)² \]  

(2)

where \( r \) is the radius of the diffusing species, \( r₀ \) is the radius of the gate or doorway through which the diffusing species must jump in going from one site to the next, \( E \) is activation energy, and \( E₁ \) is the activation energy when \( r - r₀ = 1 \) Å. Equation 2 can be made into a linear relation by taking the square root of both sides; i.e., \( E^{1/2} \) is linear with \( r \). Doremus (1973, p. 133) and Bansal and Doremus (1986, p. 27) showed that the activation energies in silica glass for the diffusion of noble gases, H₂, and O₂ in silica glass vary smoothly with molecular size (except for H₂O). They also showed that the smooth trend can be adequately described by Equation 2 using molecular radii estimated from gaseous viscosities at 700 °C. However, viscosity-based radii are not unique (e.g., the four sets of viscosity-based radii in Table 1, sets 7–10, are different, and the differences are not consistent). The radius of H₂O given by Doremus (1973) and Bansal and Doremus (1986) would indicate that diffusion of molecular H₂O does not follow the trend defined by other molecular species, but it is not clear how their 1.65 Å radius for H₂O molecules was obtained. Zhang et al. (1991) examined the relation between diffusional activation energy in rhyolitic glass and radius of H₂O, He, Ne, and Ar. They used the neutral atomic radii of Forstye (1954) for noble gas elements (which are the same as the univalent radii of Pauling, 1927; set 1) and the molecular radius of H₂O in ice (1.38 Å based on the distance between neighboring O atoms given in Pauling, 1960, p. 465). They concluded that Equation 2 can adequately describe the relation between activation energies and radii, including H₂O. They also noted, however, that the relationship is not apparent if other sets of noble gas radii are used. Because the univalent radii of Pauling (1927) are not equivalent to the radius of a molecule in a crystal structure, the comparison in Zhang et al. (1991) may be questionable. For a meaningful comparison, it is necessary to use a consistent set of radii for all species under consideration.

Because effective ionic radii of Shannon (1976) were used to develop the new set of atomic radii for noble gas elements, a radius for molecular H₂O consistent with no-
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Fig. 2. Activation energy (E) for diffusion of He, Ne, molecular \( \text{H}_2\text{O} \), Ar, and Kr in rhyolitic, albic, and silica glasses (Moulson and Roberts, 1961; Zhang et al., 1991; Carroll et al., 1993, and references therein) vs. their neutral-species radii (with \( \text{CN} = 6 \)) derived in this work. The diffusional activation energy of molecular \( \text{H}_2\text{O} \) in silica glass is that of total \( \text{H}_2\text{O} \) by Moulson and Roberts (1961) without the adjustment described by Zhang et al. (1991) because the enthalpy for the reaction is not known. (A) \( E \) vs. \( r \) with straight line fits. The equations for the best-fit lines are: \( E = 193(r - 0.87) \) with \( R^2 = 0.971 \) for rhyolitic glass where \( R \) is the linear correlation coefficient, \( E \) is in kilojoules per mole and \( r \) in ångströms; \( E = 122(r - 0.81) \) with \( R^2 = 0.977 \) for silica glass; (B) \( E^0 \) vs. \( r \) with straight line fits. The best-fit lines are: \( E^0 = 10.5(r - 0.45) \) with \( R^2 = 0.934 \) for rhyolitic glass; \( E^0 = 7.46(r - 0.29) \) with \( R^2 = 0.939 \) for silica glass.

Noble gas atomic radii can be derived by interpolation from the effective ionic radii of \( \text{O}^{2-} \), \( \text{OH}^- \), and \( \text{H}_2\text{O}^+ \), ignoring the nonsphericity of \( \text{OH}^- \), \( \text{H}_2\text{O} \), and \( \text{H}_2\text{O}^+ \) species. For \( \text{CN} = 6 \), the radius of \( \text{O}^{2-} \) is 1.40 Å, that of \( \text{OH}^- \) is 1.37 Å (Shannon, 1976), and that of \( \text{H}_2\text{O}^+ \) is similar to that of \( \text{K}^+ \) (1.38 Å) based on Criste et al. (1975) and on similar cell dimensions of alunite and hydronium alunite (Ripmeester et al., 1986). Therefore, the radius of an \( \text{H}_2\text{O} \) molecule in a condensed phase structure is estimated to be 1.37 Å. (If crystal radii instead of effective ionic radii of \( \text{O}^{2-} \), \( \text{OH}^- \), and \( \text{H}_2\text{O}^+ \) are used, the interpolated radius of \( \text{H}_2\text{O} \) would be 1.31-1.38 Å if \( \text{K}^+ \) is again used as a proxy for \( \text{H}_2\text{O}^+ \).) Considering the large uncertainty in the radius of \( \text{H}_2\text{O}^+ \), a 2\( \sigma \) uncertainty in the radius of \( \text{H}_2\text{O} \) is estimated to be 0.05 Å. The radius of molecular \( \text{H}_2\text{O} \) is much smaller than that of \( \text{Ar} \), in contrast with the data quoted in Doremus (1973) (1.65 Å for \( \text{H}_2\text{O} \) vs. 1.6 Å for \( \text{Ar} \)).

\( E \) and \( E^0 \) for the diffusion of He, Ne, molecular \( \text{H}_2\text{O} \), Ar, and Kr in silica, rhyolitic, and albic glasses are plotted against their neutral atomic or molecular radii in Figure 2. For each glass, there is a smooth trend in both plots, consistent with the conclusion of Zhang et al. (1991) that diffusion of molecular \( \text{H}_2\text{O} \) behaves similarly to that of the noble gas elements. The departure of the diffusional activation energy for \( \text{H}_2\text{O} \) from the trend formed by other molecular species in Doremus (1973) and Bansal and Doremus (1986) was due to their erroneous choice of the \( \text{H}_2\text{O} \) radius.

Even though there are smooth trends in both plots (Fig. 2A and 2B), the applicability of Equation 2 may be questioned because the data seem to be slightly better described by a linear relation between \( E \) and \( r \) instead of a linear relation between \( E^0 \) and \( r \) (1 - \( R^2 \) value of the \( E \) vs. \( r \) plot is less than half that of the \( E^0 \) vs. \( r \) plot, where \( R \) is the linear correlation coefficient; see caption of Fig. 2). At present there is no theoretical basis for the apparent linear relation between \( E \) and \( r \). More accurate determination of activation energies for a greater range of molecular radii is necessary to resolve the exact relation between activation energy and molecular radius. Determination of the activation energy for Xe diffusion in these glasses will be very helpful in resolving this issue. For example, extrapolation using the \( E \) vs. \( r \) linear trend predicts the diffusional activation energy of Xe (\( r = 1.96 \) Å) to be 210 kJ/mol in rhyolitic glass and 140 kJ/mol in silica glass, whereas extrapolation using the \( E^0 \) vs. \( r \) linear trend predicts 248 and 156 kJ/mol, respectively. These differences in activation energies are resolvable from experimental data.

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