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van der Waals Volumes and Radii

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Intermolecular van der Waals radii of the nonmetallic elements have been assembled into a list of "recommended" values for volume calculations. These values have been arrived at by selecting from the most reliable X-ray diffraction data those which could be reconciled with crystal density at 0°K. (to give reasonable packing density), gas kinetic collision cross section, critical density, and liquid state properties. A qualitative understanding of the nature of van der Waals radii is provided by correlation with the de Broglie wave length of the outermost valence electron. Tentative values for the van der Waals radii of metallic elements—in metal organic compounds—are proposed. The paper concludes with a list of increments for the volume of molecules impenetrable to thermal collision, the so-called van der Waals volume, and of the corresponding increments in area per molecule.

Table of Nomenclature

$A_w$	Surface area of molecules (based on model of Fig. 3) per mole (cm. <sup>2</sup> /mole)
$b$	Covalent bond radius, Å.
$C$	Normalization constant in eq. 1
$d_w$	Nonbonded internuclear distance between atoms of neighboring molecules
$E^\circ$	Standard energy of vaporization, defined in ref. 2
$h$	Planck constant
$I_0$	First ionization potential
$l$	Bond distance, Å.
$m_e$	Rest mass of an electron
$N_A$	Avogadro number
$r$	Distance from atom nucleus
$r_0$	Distance between like nonbonded atoms (molecules) at potential energy minimum
$r_b$	van der Waals radius estimated by Pauling's approximation, $r_b = b + \text{const.}$
$r_w'$	van der Waals radius derived from nonbonded contact distance in crystals
$r_w$	van der Waals radius
$\bar{r}_w$	Mean van der Waals radius for volume calculations involving anisometric atoms
$V_0$	Molal volume

$V_0$	Molal volume at 0°K.
$V_c$	Molal volume at critical temperature and pressure
$V_w$	van der Waals volume (calculated on the basis of Fig. 3)
$Z$	Number of nearest neighbors of a molecule
$\delta(\ )$	Decrement of $V_w$ or $A_w$ (as indicated) due to intramolecular crowding or hydrogen bonding
$\lambda_B \equiv h\sqrt{m_e I_0}$	de Broglie wave length of outermost valence electron
$\rho_0^* \equiv V_w/V_0$	packing density at 0°K.
$\rho_c^* \equiv V_w/V_c$	packing density at critical point
$\sigma$	Distance between like atoms (molecules) at steepest ascent of repulsion branch of potential energy well (as obtained from gas properties by means of Lennard-Jones and Devonshire theory)
$\psi^2$	Charge density (probability of finding an electron) at distance $r$ from the atom nucleus

Purpose and Scope

The primary purpose of the present investigation is the calculation of the volume occupied by a molecule, *i.e.*, impenetrable for other molecules with thermal energies at ordinary temperatures. This volume, called here the van der Waals volume ( $V_w$ ), is to serve

as reducing parameter in the study of the physical properties of condensed phases.<sup>1,2</sup> The calculation of  $V_w$  assumes a knowledge of bond distances, bond angles, and the contact distances [*i.e.*, intermolecular van der Waals radii ( $r_w$ )] and shapes characteristic of atoms in various molecular configurations. While most of the important bond distances and angles in organic molecules are reasonably well known and recorded,<sup>3,4</sup> only a few and semiquantitative contact (= nonbonded intermolecular) distances in crystals have been collected.

The first task at hand was therefore the collection of contact distances from reliable X-ray diffraction data in the literature. Selection of the "best" values from the resulting mass of data is of necessity an arbitrary procedure, which will be discussed in the body of this paper. Two important approximations have been made which can only be excused by the desire to obtain results now; namely, the contact distances have not been corrected<sup>5</sup> to 0°K. and all atoms have been treated as spheres and sphere segments, although it is well known that many are more nearly pear-shaped. The reason for the first approximation is the absence of data regarding the often anisotropic thermal expansion coefficients of the crystals from which the contact distance data were obtained. Spherical shapes have been assumed because of the absence of generally agreed pear shapes for the various atoms and the mathematical complexity of testing various alternative shapes for internal consistency. The values of  $V_w$  presented in this paper are therefore subject to further improvement. However, they have proven sufficiently useful, even in their present state, so that their publication appears justified.

### General Principles

Assumption of the existence of a defined spatial extent of atoms is common to kinetic gas theory and X-ray crystallography. The extent of agreement between the dimensions of the rare gas atoms produced by both approaches can be considered as a measure of the status of kinetic gas theory.<sup>6</sup> However, this test is restricted to the rare gases because the crystals of metals, the only other monatomic species, are held together by covalent bonds and therefore do not permit comparison of the interatomic distances during thermal collisions of gas atoms with those prevailing in the solid.

This observation brings us to one of the tacit assumptions of this inquiry, the invariance of the van der Waals radius of an atom under the most drastic environmental changes, *i.e.*, irrespective of its chemical combination and of its nearest nonbonded neighbors

as well as of the phase state in which it is found. Closer examination shows that this assumption is surprisingly valid for heavy atoms, but is not very good with atoms containing only a few electrons, such as hydrogen, fluorine, etc. It may, therefore, be worthwhile to look at the nature of the van der Waals radius from the point of view of the electron density distribution around an atom.

The electron density  $\psi^2$  at distance  $r$  from the core of a hydrogenic one-electron atom is given by the well known relation

$$\psi^2 = C^2 \exp\left(-\frac{2\sqrt{2m_e I_0}}{\hbar} r\right) \quad (1)$$

where  $m_e$  is the rest mass of the electron,  $I_0$  the first ionization potential of the atom, and  $C$  is a normalization constant chosen such that  $\int \psi^2 d\omega = 1$  when the integration is carried out over the whole of space ( $\omega$ ). The shape of the electron density distribution for typical one and multielectron atoms is shown on Fig. 1. As two atoms approach each other from  $r = \infty$  their electron clouds interpenetrate more and more. The Pauli exclusion principle then causes a repulsion<sup>7</sup> of the two atoms in direct proportion<sup>8</sup> to the electron density in the region of interpenetration. One might define the van der Waals radius in terms of that distance  $r$  at which this repulsion just balances the attraction forces between the two atoms. Comparison of the abscissa of Fig. 1 with the empirically known van der Waals radii shows that this distance corresponds to a very low electron density—so low, in fact, that one cannot formulate a "critical" electron density and hope to calculate  $r_w$  from it because of the low degree of accuracy of the known calculating schemes in that part of the energy spectrum.

However, eq. 1 contains a parameter  $\hbar/\sqrt{m_e I_0}$ , the de Broglie wave length  $\lambda_B$  of the outermost valence

(1) A. Bondi, *A.I.Ch.E. J.*, **8**, 610 (1962).

(2) A. Bondi and D. J. Simkin, *ibid.*, **6**, 191 (1960).

(3) Landolt-Bornstein, "Zahlenwerte und Funktionen I/4: Kristalle," Springer, Berlin, 1955.

(4) L. Pauling, "The Nature of the Chemical Bond," Cornell University Press, Ithaca, N. Y., 1942, p. 192.

(5) Since the volume increase of most solids between 0°K. and the melting point is about 10%, the van der Waals radii at 0°K. differ from those given here, probably by less than 3%.

(6) J. O. Hirschfelder and R. B. Bird, "Molecular Theory of Gases and Liquids," New York, N. Y., 1959.

(7) See J. A. A. Ketelaar, "Chemical Constitution," Elsevier Publishing Co., Amsterdam, 1953, p. 146 ff. for a more detailed discussion; a somewhat different treatment is presented by K. S. Pitzer, "Quantum Chemistry," Prentice-Hall, Inc., New York, N. Y., 1953, p. 201.

(8) The repulsion potential  $E_{r2} \sim \exp(r/\alpha)$ ; hence, at the distances of the order  $r_w$ , considering eq. 1,  $E_r \sim \psi^2$ , the electron density in the region of interpenetration.

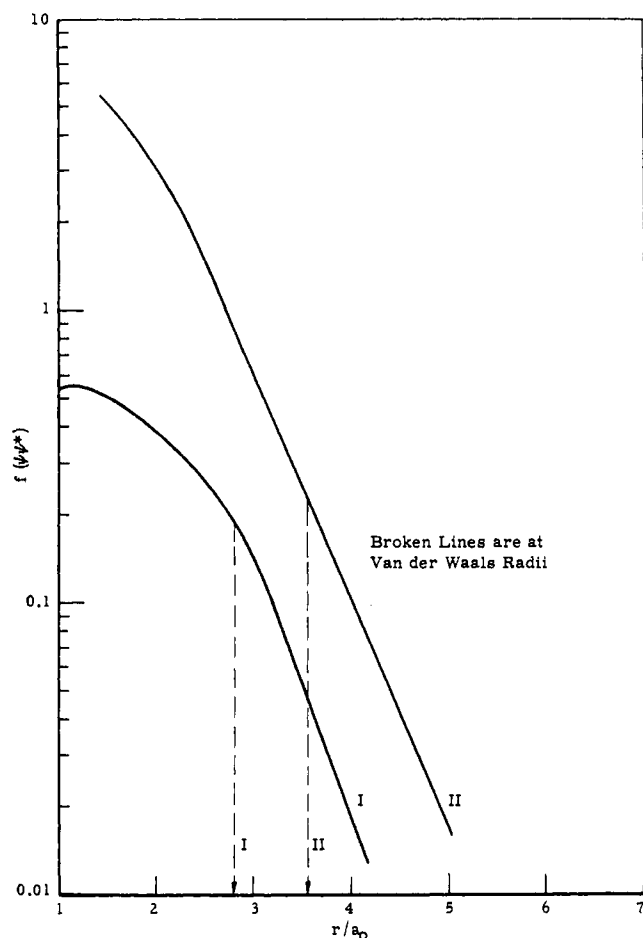


Figure 1. Electron density distribution near the atom "surface," for hydrogen (I) and argon (II) [D. R. and W. Hartree, *Proc. Roy. Soc. (London)*, A166, 450 (1938)].

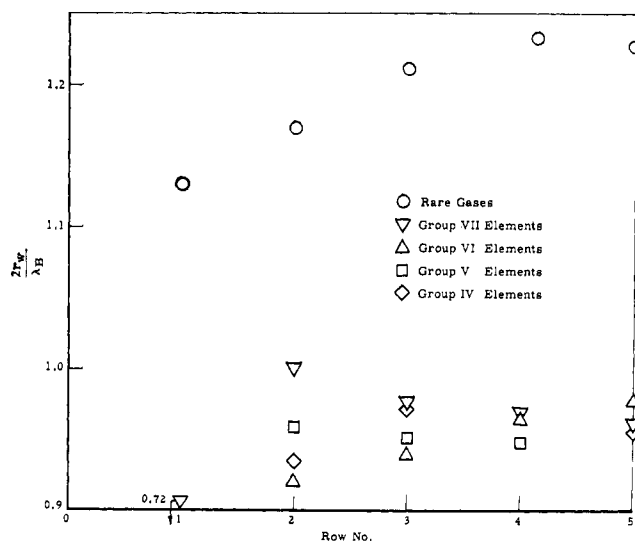


Figure 2. Relation of  $r_w$  (single bond value) to de Broglie wave length  $\lambda_B$  of outer valence electron for nonmetallic elements.

electron of an atom, which might be related to the van der Waals radius ( $r_w$ ). A few years ago Morrison<sup>9</sup> suggested that perhaps  $r_w = (\text{const.}) \lambda_B$ . Our examination of this suggestion brought to light that this simple correlation holds surprisingly well. One finds that the constant is 0.61 for the rare gas atoms, and is—as one might expect—appreciably smaller for bound atoms, *ca.* 0.53 for the halogens and about 0.48 for the remainder of the nonmetallic elements. Deviations from this correlation (shown in Fig. 2) are in the direction of predicting too large a diameter for the lightest elements. As the valence electrons of a covalently bound atom are concentrated between it and its bound neighbor to a degree that depends on the nature of the bond, it is surprising that  $r_w$  retains enough individuality to be correlatable in terms of the ionization potential of the free atom.

Table I: Comparison of Correlation with "Observed" Mean van der Waals Radii (in Å.)

					H	He
$r_b^a$					1.06	...
$\lambda_B/2^b$					1.67	1.24
$\bar{r}_w^c$					1.20	1.40
	B	C	N	O	F	Ne
$r_b$	1.65	1.53	1.46	1.42	1.40	...
$\lambda_B/2$	2.13	1.82	1.61	1.66	1.47	1.32
$\bar{r}_w$	..	1.70	1.55	1.52	1.47	1.54
	Al	Si	P	S	Cl	Ar
$r_b$	..	1.93	1.86	1.80	(1.75) <sup>d</sup>	..
$\lambda_B/2$	2.51	2.15	1.87	1.91	1.70	1.55
$\bar{r}_w$	..	2.10	1.80	1.80	1.75	1.88
	Ga	Ge	As	Se	Br	Kr
$r_b$	..	1.98	1.94	1.90	1.87	..
$\lambda_B/2$	2.51	2.19	1.96	1.97	1.79	1.64
$\bar{r}_w$	..	..	1.85	1.90	1.85	2.02
	In	Sn	Sb	Te	I	Xe
$r_b$	..	2.16	2.12	2.08	2.04	2.05
$\lambda_B/2$	2.55	2.27	2.09	2.05	1.90	1.76
$\bar{r}_w$	..	..	..	2.06	1.98	2.16

<sup>a</sup>  $r_b \equiv b + 0.76$ . <sup>b</sup>  $\lambda_B \equiv 6.13 \times 10^{-8} / \sqrt{I}$ . <sup>c</sup> Only the most frequently used values for single bonded forms of the elements are quoted here. <sup>d</sup> Reference point chosen for the system  $r_b = b + \text{constant}$ .

### Compatibility with Physical Properties

The range of numerical values of  $r_w$  for a given atom obtained from X-ray crystallographic contact distances is usually too wide for the direct application to a meaningful calculation of  $r_w$ . The "best" value of  $r_w$  must

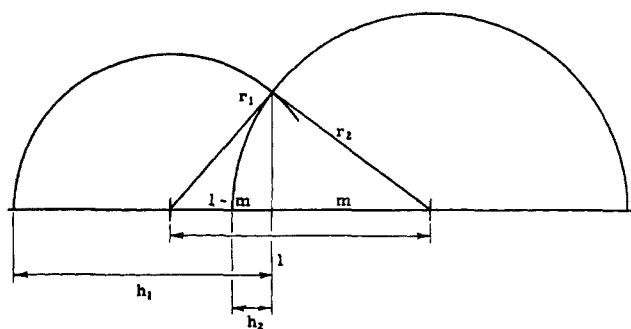
(9) J. D. Morrison, *Rev. Pure Appl. Chem.*, 5, 46 (1955).

then be obtained by appeal to extraneous information. In the ideal case enough crystal structure and zero point density data are available to fix through calculation that value of  $r_w$  which yields the correct packing density  $\rho_0^*$  at 0°K. This has been done for the simple tetrahalides by Sackmann.<sup>10</sup>

In general, when a detailed analysis is either too laborious or not possible, the "plausibility" of the packing density calculated from the experimental zero point volume<sup>11</sup> and  $V_w$  will fix at least the upper limit of  $r_w$ . The empirical observation that for molecular crystals  $\rho_0^*$  always  $>0.6$  may be used to fix a lower limit on  $V_w$ . The method used to estimate  $V_w$  from bond distance  $l$  and from  $r_w$  is shown on Fig. 3.

A lower limit is set for  $r_w$  by the requirement that the packing density at the critical temperature must be sufficiently high to permit the existence of a continuous three-dimensional network, *i.e.*, the number of nearest neighbors  $Z \geq 3$ .<sup>12</sup> A related criterium is the compatibility with the size of the equivalent sphere obtained by application of kinetic gas theory.<sup>13</sup> The somewhat surprising regularity that  $V_w \approx N_A(\pi/6)\sigma^3$  which we observed whenever  $r_w$  met the other two criteria, is useful when neither  $\rho_0$  nor  $\rho_c$  but only gas properties have been measured.

A particularly arbitrary criterium developed in the course of the present work is that the number of external degrees of freedom of rigid (nonlinear) poly-



$r_1, r_2$  = van der Waals radii  
 $l$  = covalent bond distance  
 $m$  = auxiliary parameter  
 $h_1, h_2$  = height of sphere segments  
 $m = \frac{r_2^2 - r_1^2 + l^2}{2l}$ ;  $h_1 = r_1 + l - m$ ;  $h_2 = r_2 - m$   
 $V_1^s = \pi h_1^2 (r_1 - \frac{h_1}{3})$ ;  $\Delta V_{2-1} = \pi h_2^2 (r_2 - \frac{h_2}{3})$ ;  $V_2 = \frac{4\pi}{3} r_2^3$

Example:  
 van der Waals volume of diatomic molecule:

$V_w = N_A [V_1^s + V_2 - \Delta V_{2-1}] \text{ cm}^3/\text{mole}$   
 where  $N_A = 6.02 \times 10^{23}$  molecules/mole  
 and  $r$ 's are given in Angstrom units

Volume of center atom = Total volume of atom  
 Surface area =  $2\pi rh$

Figure 3. Method of calculation.

atomic molecules calculated from liquid density and energy of vaporization ( $E^\circ$ ) data, as  $0.391E^\circ/RT \cdot (1.8) = 6 \pm 0.2$ .<sup>2</sup> The application of this criterium requires the availability of good liquid density and vapor pressure or calorimetric heat of vaporization data and great faith in the form of the corresponding states principle adopted for this purpose.

### Experimental Data for Nonmetallic Elements

In this account the long form of the periodic table is followed from right to left.

*The Halides. Fluorine.* The data of Table II show that Pauling's suggestion to set the ionic radius (1.35 Å.) =  $r_w$  yields rather improbable values for  $\rho_c^*$  of well studied fluorine compounds. Crystallographic data for  $\text{NH}_3 \cdot \text{BF}_3$ ,<sup>14</sup>  $\text{SiF}_4$ ,<sup>3,15</sup> phosphonitrilic fluoride,<sup>16</sup> and Teflon<sup>17,18</sup> yield  $r_w(\text{F}) = 1.50$  Å., a value which, according to Table II, is compatible with  $\rho_0$ ,  $\rho_c$ , and  $\sigma$ . The liquid state criterium led to the very similar value  $r_w(\text{F}) = 1.47$  Å. for perfluoroalkanes, aryl fluorides, and for secondary and tertiary alkane fluorides. However, for primary alkane fluorides  $r_w(\text{F}) = 1.40$  Å. was found more compatible with the data. In view of the exactly opposite trend of the C-F bond length, this is an unexpected result, for which neither conflicting nor confirmatory evidence could be found in X-ray diffraction data.

*Chlorine, Bromine, and Iodine.* The previously mentioned analysis of the crystallographic and density data of the tetrahalides of the group IV elements by Sackmann<sup>10</sup> yielded  $r_w(\text{Cl}) = 1.76$  Å.,  $r_w(\text{Br}) = 1.85$  Å.,  $r_w(\text{I}) = 1.96$  Å., in good agreement with many other X-ray diffraction data, shown in Table III. The data of Table IV indicate that the resulting values of  $V_w$  for various halogen compounds are more compatible with experimental density data than are the frequently quoted van der Waals radii based on Pauling's approximation  $r_w = r_i$ . The radii and volume increments derived from the liquid state criteria have been assembled in Table IV. They all show the same value for the primary alkanes previously noted for fluorine.

(10) H. Sackmann, *Z. physik. Chem.*, **208**, 235 (1958).

(11) W. Biltz, "Raumchemie der festen Stoffe," Leipzig, 1934.

(12) This point will be discussed in detail in a subsequent article.

(13) A. Bondi, *J. Phys. Chem.*, **58**, 929 (1954).

(14) J. L. Hoard, *et al.*, *Acta Cryst.*, **4**, 396 (1951).

(15) M. Atoji and W. N. Lipscomb, *ibid.*, **7**, 173 (1954).

(16) H. McGeachin and F. Tromans, *J. Chem. Soc.*, 4777 (1961).

(17) C. W. Bunn and E. R. Howells, *Nature*, **174**, 548 (1954).

(18) H. G. Killiam and E. Jenckel, *Z. Elektrochem.*, **63**, 308 (1959).

**Table II:** Compatibility of Proposed van der Waals Radii of Fluorine with Physical Properties

Substance	$r_w(\text{F})$ , Å.	$V_w$ , cm. <sup>3</sup> /mole	$\frac{V_w}{N_A(\pi/6)\sigma^3}$ <sup>a</sup>	$\rho_0^{*b,c}$	$\rho_0^{*d}$
F <sub>2</sub>	1.35	10.5	0.88	0.42	...
	1.50	14.2	1.19	0.57	...
CF <sub>4</sub>	1.35	16.5	0.51	0.56	0.161
	1.50	29.5	0.92	0.69 <sub>5</sub>	0.200
<i>n</i> -C <sub>7</sub> F <sub>16</sub>	1.35	105.8	..	...	0.160
	1.50	127.7	..	...	0.193

<sup>a</sup> See ref. 6. <sup>b</sup> See ref. 11. <sup>c</sup> G. A. Miller and R. B. Bernstein, *J. Phys. Chem.*, **63**, 710 (1959). <sup>d</sup> K. A. Kobe and R. E. Lynn, *Chem. Rev.*, **52**, 121 (1953), and Miller and Bernstein, preceding reference.

**Table III:** Recent Intermolecular Contact Radii  $r'$  for Chlorine, Bromine, and Iodine in Molecular Crystals

Compound	$r_w'(X)$ , Å.
$\lambda$ -C <sub>6</sub> H <sub>6</sub> Cl <sub>6</sub> (Gammexane)	(Cl) 1.70 <sup>a</sup>
Perchlorofulvalene	(Cl) 1.74 <sup>b</sup>
Tetrachloro- <i>p</i> -benzoquinone	(Cl) 1.78 <sup>c</sup>
1,3,5-Trichlorobenzene (90°K.), 293°K.	(Cl) (1.77), 1.82 <sup>d</sup>
1,4-Dibromocyclo[3.2.2]azine	(Br) 1.80 <sup>e</sup>
<i>p</i> -Dibromobenzene	(Br) 1.88 <sup>f</sup>
1,3,5-Tribromobenzene	(Br) 1.88 <sup>d</sup>
Iodoform, 0°K.	1.99 <sup>g</sup>
Phosphorus thioiodide	1.95 <sup>h</sup>
<i>p</i> -Diiodobenzene	2.07 <sup>i</sup>

<sup>a</sup> G. W. Van Vlater, *et al.*, *Acta Cryst.*, **3**, 139 (1950). <sup>b</sup> See ref. 21. <sup>c</sup> S. Chu, *et al.*, *Acta Cryst.*, **15**, 661 (1962). <sup>d</sup> H. J. Milledge and L. M. Pant, *ibid.*, **13**, 285 (1960). <sup>e</sup> A. Hanson, *ibid.*, **14**, 124 (1961). <sup>f</sup> U. Croatto, *et al.*, *ibid.*, **5**, 825 (1952). <sup>g</sup> See ref. 30. <sup>h</sup> D. A. Wright and B. R. Penfold, *Acta Cryst.*, **12**, 455 (1959). <sup>i</sup> S. B. Hendricks, *et al.*, *J. Chem. Phys.*, **1**, 549 (1933).

*Oxygen, Sulfur, Selenium, and Tellurium.* As the diameter of singly bonded oxygen and sulfur is smaller than that of methylene or other functional groups to which they might be attached, one finds only few good contact distance data for them. Moreover, in the present context all contacts involving hydrogen bonds must be excluded. As a consequence of being too deeply "buried" within the molecule for frequent (or any) collision with neighboring molecules, the "effective volume" occupied by an ether oxygen atom differs from molecule to molecule. The upper limit on  $r_w(>O)$  set by the few available X-ray data (Table VI), 1.52 Å., corresponds to  $V_w(>O) = 5.5$  cm.<sup>3</sup>/mole. Only in ethylene oxide is the ether oxygen sufficiently

**Table IV:** Compatibility of Proposed van der Waals Radii of Cl, Br, and I with Physical Properties

Substance	$r_w(X)$ , Å.	$V_w$ , cm. <sup>3</sup> /mole	$\frac{V_w}{N_A(\pi/6)\sigma^3}$ <sup>a</sup>	$\rho_0^{*b}$	$\rho_0^{*c}$
Cl <sub>2</sub>	1.75	24.1	1.09	0.74	0.195
	1.85	28.9	1.16	0.75	0.200
Br <sub>2</sub>	2.00	35.6	1.42	0.93	0.240
	1.96	37.6	0.96	0.76	...
I <sub>2</sub>	2.16	46.0	1.17	0.93	...
	1.75	51.4 <sup>d</sup>	0.8	0.69	0.186
CCl <sub>4</sub>	1.75	51.4 <sup>d</sup>	0.84	0.69	...
CBr <sub>4</sub>	1.85	60.8 <sup>d</sup>	..	0.68	...
CI <sub>4</sub>	1.97	74.9 <sup>d</sup>	..	0.68	...

<sup>a</sup> See ref. 6 and R. A. Svela, NASA Technical Report R-132 (1962). <sup>b</sup> See ref. 11. <sup>c</sup> See ref. *d* in Table II. <sup>d</sup> No allowance has been made for possible volume reduction due to overlap of the halogen atoms or for their probable pear shape. Such correction would reduce  $V_w$  and the other reduced densities, but would leave the constancy in the series unchanged.

**Table V:** Effective van der Waals Radii of Bound Halogen Atoms from Liquid State *p-v-t* Property Calculations

X	Attachment	$r_w(X)$ , Å.
F	Primary alkyl	1.40
	<i>sec</i> , <i>tert</i> , alkyl, perfluoro alkane, phenyl	1.47
Cl	Primary alkyl	1.73
	Vinyl	1.75
	<i>sec</i> , <i>tert</i> , alkyl, polychloroalkyl, phenyl	1.77
Br	Primary alkyl	1.84
	<i>tert</i> , polybromoalkyl	1.85
	Phenyl	1.92
I	Primary alkyl	2.01
	<i>tert</i> , polyiodoalkyl, phenyl	2.06

exposed to yield this value of  $V_w(>O)$ . In alkyl ethers, especially polyethers,  $V_w(>O) = 3.7$  cm.<sup>3</sup>/mole and in polyphenyl ethers it is 3.2 cm.<sup>3</sup>/mole. These values were obtained by fitting polyether data to the generalized density correlation for liquids.

For single-bonded sulfur, X-ray diffraction data point to  $r_w(>S) = 1.83$  Å., while 1.80 Å. is compatible with various physical properties (Table VII) and with liquid phase density. For single-bonded selenium, a single set of data<sup>19</sup> yields  $r_w(>Se) = 1.87$  Å. and for tellurium  $V_w(>Te) = 2.06$  Å.

The geometry of double-bonded oxygen and sulfur is probably quite anisometric. In the direction parallel

(19) R. E. Mush, *Acta Cryst.*, **5**, 458 (1952).

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