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ESTIMATED VISCOSITY, SURFACE TENSION, AND DENSITY OF LIQUID DT FROM THE TRIPLE POINT TO 25 K

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Abstract

The literature data for the saturated viscosity, liquid-vapor surface tension, and density of liquid hydrogen is reviewed for the temperature range 14–25 K. Extrapolations are made to estimate the corresponding values for DT and T_2. Solution mixing data is considered to estimate values for the solution eD_2-DT-eT_2. Estimated values at the 1:1 D-T triple point at 19.75 K are: viscosity, 550×10^{-7} Pa·s; surface tension, 4.23×10^{-3} N/m; and density, 225 kg/m³. These derived values are used to illustrate, via simplified equations, some basic properties of liquid DT droplets, which may eventually be used as targets in laser-induced hydrogen fusion experiments. Slow creation of individual drops and the liquid jet method are considered. The combination of low liquid density with low surface tension will allow formation of hundreds of μm-diameter droplets with little problem. The viscosity is sufficient to damp out oscillations on a millisecond time scale.

Introduction

Currently, laser fusion is of great interest as a possible means of producing power. The current targets are hollow glass microspheres filled with DT gas.[1-3] Future targets for higher energy lasers will be solid shells of DT. When very high-energy lasers (perhaps 10^5 J) are available, the targets may be liquid or solid drops of DT.[4] This use of DT will require information about its cryogenic properties. Thus, we are embarking on a program to experimentally measure those physical properties of DT (near the triple point) which would be most affected by the presence of tritium. We are also reviewing a number of physical properties of hydrogen in order to extrapolate to the values for DT. Two such studies have already been published[5,6] as part of this program.

Cryogenic laser targets will probably be created from liquid droplets. Hence, the liquid viscosity, surface tension, and density on the phase boundary near the triple point are of considerable interest. We review here the literature data, which is almost all on H_2,

HD, and D_2, and then estimate DT and T_2 values.

The actual laser fuel will be the three-component solution of eD_2-DT-eT_2, with a 1:1 atomic D:T ratio. We have estimated the relative amounts of these three components by extrapolation of 0.1-Pa thermodynamic gas calculations.[5,7] These predict the approximate proportions in the range 19–25 K of 30 mole % D_2, 40 mole % DT, and 30 mole % T_2. The triple point should occur at about 19.75 K, and we have assigned an extrapolated value of 40.20 % DT at this temperature. We assume that the percent of DT increases linearly with temperature to 41.04 % at 25 K. It should be emphasized, however, that no measurements or calculations have actually been made for this chemical equilibrium in the liquid state.

We feel that the properties of viscosity, surface tension, and density will probably not be affected by the presence of the tritium beta particles and the resultant radiation damage. Hence, estimating these properties for the liquid state seems feasible.

Liquid Viscosity

Recent experimental measurements of the liquid viscosity of hydrogen* are: sym-H_2, 15 K to the critical point[8,9]; nH_2, 14–32 K[8,10,12]; HD, 16.6 to 34 K[11–13]; nD_2, 18.8–36 K[10,12]; and H-D solutions, 18.8 and 20.3 K.[13] For Refs. 10, 12, and 13, one must assume the normal form of H_2 and D_2 as the form is not specifically indicated. Most of the work cited in this section represents a sustained Soviet program.[9–13] All known viscosity data has been recently collected in a National Bureau of Standards (NBS) survey.[14] We shall not consider the calculated HT and T_2 values, since these do not fit with experimental data taken on the other hydrogens.[15]

For the above data, two experimental methods were used. What we considered the best data was taken by Diller[8] using the torsional crystal method, in which the fluid produces a damping effect about an

*The types of hydrogen are defined in Appendix A. Symbols used here are: sym- for symmetric and asym- for antisymmetric.

oscillating piezoelectric crystal in the shape of a right circular cylinder.[8,12] Diller's data is higher than the corresponding torsional crystal data of the Soviets[12] by 6–7 % at 14 K and 3–4 % at 25 K. Diller's data is 6 % higher than other Soviet work[10,16] done by the capillary viscometry method between 14–20 K.

The nH_2 viscosity ratios of Diller to the Soviets are 1.068, 1.085, 1.075, 1.063, and 1.041 for 14, 16, 18, 20, and 22 K, respectively. Within error, the first four points may be averaged to a constant value of 1.073; it is only above 20 K that the ratio definitely begins to diminish. We expect that the correction is a function not of the temperature but of the relative position to the triple point, i.e., to the ratio of measured temperature divided by triple point temperature. By this algorithm, we find that the 1.073 ratio may be applied to all isotopes to 25 K, with only HD beginning to slip at 24–25 K to lower values. We have, therefore, multiplied the Soviet HD and D_2 data to 25 K[12] by 1.073 to bring it into line with Diller.[8] All values are given in Table 1.

Table 1. Viscosity of the hydrogen isotopes from 14 to 25 K.

Temperature (K)	Triple point of	Viscosity (10^{-7} Pa·s)[a]					
		nH_2 (Refs. 8,14)	HD[b] (Refs. 11–14)	D_2[b,c] (Refs. 10,12,14)	DT	T_2	D_2-DT-T_2 (1:1 DT)
13.957	nH_2	257.0					
14.00		254.3					
15.00		230.2					
16.00		203.9					
16.60	HD	191.0	363				
17.00		182.9	344				
18.00		165.6	305				
18.71	D_2	156.0	282	466			
19.00		151.5	274	449			
19.71	DT	143.0	255	407	(590)	(736)	(550)
20.00		139.2	248	395	(560)	(715)	(529)
20.62	T_2	133.0	237	376	(520)	(662)	(494)
21.00		128.4	230	365	(498)	(632)	(473)
22.00		118.7	215	341	(469)	(596)	(445)
23.00		110.5	202	319	(439)	(562)	(418)
24.00		102.6	193	296	(406)	(515)	(385)
25.00		95.7	179	279	(382)	(486)	(363)

[a]Same unit as micropoise. Parentheses are estimates.
[b]Adjusted.
[c]Assume normal.

We plotted the viscosity as a function of molecular weight at each temperature and found it to be linear for H_2, HD, and D_2. We therefore extrapolated linearly to DT and T_2. These values are given in Table 1. The T_2 values are extrapolated to 19.71 K (even though T_2 is solid below 20.62 K) in order to calculate the three-component solution value of viscosity.

As previously mentioned, the liquid D_2-DT-T_2 contains about 40 mole % DT and 30 mole % of each of the other components for a 1:1 D-T solution from 19.71 to 25 K. Assuming additivity of the values so far obtained, we can easily use them to calculate the expected solution viscosity. The effects of non-additivity may be estimated from the Soviet work, which is shown in Fig. 1.[13] Their non-additivity factor, f_η, is[13]

$$f_\eta = \frac{\Delta \eta}{\eta} = \frac{\eta - \eta_{exp}}{\eta}. \tag{1}$$

where η_{exp} is the measured value and η the additive value. We obtain

$$\eta_{exp} = (1 - f_\eta)\eta. \tag{2}$$

We note that η_{exp} is less than η, since f_η is positive. Figure 1 shows non-additivity maxima at 18.8 K of 11.5, 7.0, and 5.1 % for H_2-D_2, H_2-HD, and HD-D_2, respectively. We shall follow the Soviet lead by plotting these values versus the relative molecular weight difference, M_{12}:[13]

$$M_{12} = \frac{M_2 - M_1}{\left(\dfrac{M_1 + M_2}{2}\right)} \tag{3}$$

where M_1 is the molecular weight of the lighter component and M_2 that of the heavier.

Equation (3) assumes that the maximum non-additivity occurs for 50-50 solutions, whereas Fig. 1 shows that the peak in two cases occurs for 35-65 mixtures. We shall ignore this and use a f_η-M_{12} plot to obtain maximum non-additives for the heavier hydrogen species. We obtain the values 7.0, 3.8, and 3.1 % as maximum non-additives for D_2-T_2, D_2-DT, and DT-T_2, respectively. Figure 2 is a triangular, phase diagram plot, with D_2, DT, and T_2 at each corner. The non-additives are plotted vertically and form a surface, bowed upward in the center by an unknown amount. The maximum heights of the projections on the binary mixture sides of the triangle are the maximum non-additives just calculated. The expected D_2-DT-T_2 composition lies almost exactly in the center of the diagram, where the degree of curvature is hardest to estimate. The average of the three maxima is 4.6 %; in Fig. 2, we increase it to 5 % to account for some upward curvature. It is possible that the curvature is much greater and the non-additivity much more extreme. It is also uncertain whether the variable M_{12} is the correct variable to use in calculating non-additivity. These questions must await an actual measurement of viscosity. Table 1 gives the estimated D_2-DT-T_2 viscosity as the additive value less a value of 0.05 for f_η in Eq. (2).
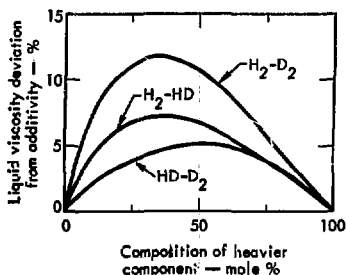

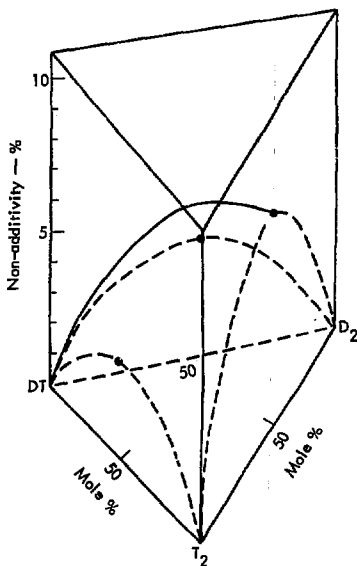Fig. 1. Non-additivity of liquid viscosity of H-D solutions at 18.8 K.[13]

Fig. 2. Schematic non-additivity surface for the viscosity, f_η, of D_2, DY, and T_2.

Liquid-Vapor Surface Tension

Compiling liquid-vapor surface tension data is somewhat complicated. Modern experimental measurements are all reported in a series of Soviet papers from 1964–1966. These papers cover the following topics: sym-H_2, 17–20.5 K[17]; nH_2, 16–20.5 K[18]; and 21–33 K[19]; HD, 16.5–20.5[18,20]; and nD_2, 19–20.5 K[18]. Solutions of H_2-D_2, 16.5–20.5 K[18]; and H_2-HD, HD-D_2, 16–20.5 K[21], were also studied. A summary of the actual data appears in the recent NBS cryogenic hydrogen review.[22]

All measurements were made by the method of liquid rise in capillaries,[23] with two different diameters being used in each set of experiments. The wetting angle hydrogen was found to be zero within $1\frac{1}{2}^\circ$.[19] The corrections to the simple formulae for capillary rise become quite complex.[24]

To obtain values below 16 K, however, one must go to older data.[25] This data was readjusted with that of the first two Soviet papers by Corruccini[24] who produced corrected points for nH_2 from the triple point to the critical point. His nH_2 data from 20.5–33 K is by extrapolation. Corruccini also estimated sym-H_2 from the triple point to the critical point based on the Soviet observation that sym-H_2 is 1–2 % less than nH_2 from 17–20.5 K.[17] Most of Corruccini's values were, therefore, estimated. Unfortunately, he also carried out his analysis just before the Soviets published the 21–33 K data for nH_2.[19] Therefore, his surface tensions are about 0.04×10^{-3} N/m higher than the probably more accurate Soviet data of the 21–33 K range.[19] This suggests that the HD and D_2 data, taken in earlier Soviet papers, may also be slightly high.

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Therefore, Table 2 contains the nH_2 values from 21–25 K as the fiducial data. All other experimental data has been lowered 0.04×10^{-3} N/m to be in agreement with the fiducial range. DT and T_2 data have been obtained by plotting the nH_2, HD, and nD_2 data at each temperature as a function of molecular weight and extrapolating to the higher weight species. The DT and T_2 curves have also been smoothed out and inherently contain the -0.04×10^{-3} N/m correction. The surface tension of sym-H_2 is 1–2 % below that of nH_2.[17] In many cases, differences in physical properties between the symmetric and antisymmetric forms of hydrogen diminish with increasing molecular weight. Therefore, the values given for D_2 and T_2 can probably be interchangeably used for both normal and equilibrium forms, within the errors of measurement and adjustment.

We next calculate the surface tension for the 19.75 K equilibrium D_2-DT-T_2 solution. Again, the deviations from additivity have been investigated in Soviet work, which is summarized in Fig. 3.[18,21] The HD-D_2 and H_2-HD data is averaged over 16 to 20.5

K[21]; the H_2-D_2 data is averaged for 18.8 and 20.4 K.[18] The experimental surface tension is found, as with viscosity, to be less than the additive surface tension. As with Eq. (1), we write

$$f_\gamma = \frac{\gamma - \gamma_{exp}}{\gamma}.\tag{4}$$

where γ_{exp} is the experimentally measured surface tension and γ the additively calculated one.* Then,

$$\gamma_{exp} = (1 - f_\gamma)\gamma.\tag{5}$$

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*The Soviet papers define the deviation from additivity for surface tension as being negative[18,21]:

$$f'_\gamma = \frac{\gamma_{exp} - \gamma}{\gamma},$$

where the denominator is somewhat unclear from the text. We have used the positive formulation as the Soviets used for viscosity.[13]

Table 2. Liquid surface tension of the hydrogen isotopes from 14 to 25 K.

Temperature (K)	Triple point of	nH_2 (Refs. 18,19,22)	HD (Refs. 18,20,22)	nD_2 (Refs. 18,22)	DT	T_2	D_2-DT-T_2 (1:1 DT)
13.957	nH_2	2.95^b					
14.00		2.94^b					
15.00		2.78^b					
16.00		2.61^b					
16.60	HD	2.51^b	(3.36)				
17.00		2.45^b	3.29^b				
18.00		2.28^b	3.09^b				
18.71	nD_2	2.17^b	2.96^b	(3.77)			
19.00		2.13^b	2.92^b	3.72^b			
19.71	DT	2.01^b	2.78^b	3.57^b	(4.36)	(5.03)	(4.23)
20.00		1.96^b	2.72^b	3.51^b	(4.28)	(4.95)	(4.17)
20.62	T_2	1.85^b	(2.61)	(3.37)	(4.13)	(4.78)	(4.02)
21.00		1.79	(2.54)	(3.28)	(4.00)	(4.68)	(3.91)
22.00		1.63	(2.35)	(3.06)	(3.74)	(4.40)	(3.66)
23.00		1.46	(2.16)	(2.84)	(3.50)	(4.14)	(3.43)
24.00		1.30	(1.96)	(2.62)	(3.26)	(3.84)	(3.18)
25.00		1.14	(1.79)	(2.40)	(3.01)	(3.57)	(2.94)

[a]Same as dyne/cm. Parentheses indicate estimates.
[b]Adjusted experimental values.

From Fig. 3, we observe maximum deviations from additivity of 3.8, 2.7, and 1.5 % for H_2-D_2, H_2-HD, and HD-D_2, respectively. Plotting these maximum deviations from additivity versus the relative mass difference [Eq. (3)], we obtain estimated values of 2.3, 1.3, and 1.0 % for D_2-T_2, D_2-DT, and DT-T_2, respectively. As with viscosity, this will form a mountain in composition space. The curvature of the mountain top is unknown. We shall average the above values to 1.5% and increase this to 2.0% to account for probable curvature. Thus, we insert a value of 0.02 for f_γ into Eq. (5) as a modifier to the additive surface tension of D_2-DT-T_2. This calculated value is given in the last column of Table 2.
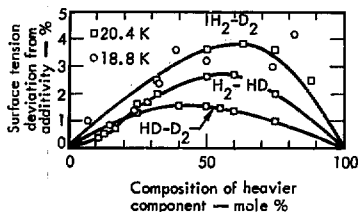

Fig 3. Experimental additivity of surface tension in binary hydrogen mixtures.[18,21]

Liquid Density

There is considerable literature on liquid hydrogen densities, but we shall use the data recommended by the NBS cryogenic hydrogen survey.[26] These data are: nH_2, 14-26[8,27]; HD 16.6-27.0 K[28,29]; nD_2, triple point to critical point[30,31]; and T_2 (probably e), 20.6-29.1 K.[32,33] The nH_2 and nD_2 data are correlations and smoothings of earlier sources. The density of sym-H_2 is only 0.3-0.4 % less than that of nH_2 in the temperature range, 13.97 to 25 K.[8,34] The sym-asym differences for the heavier hydrogen species may certainly be neglected. Several studies have been done on the volumetric properties of H_2-D_2 mixtures, and we cite the work of three publications.[35-37] Further, Kerr reports volume changes in a 1:1 mixture of D_2 and T_2 from 19.5 to 24.5 K.[38]

Liquid densities are measured with a calibrated flask. This may be combined with a direct mass measurement with a balance[28,37] or by reduction of a known gas pressure upon liquefaction.[32] Similar methods are used for mixtures with volume changes, but extra care is needed to accurately fill the calibrated volume with the two liquified components.[35-37]

The data for each isotope is interpolated and extrapolated to yield values in the temperature range of interest to us. At each temperature, the densities are plotted as a function of molecular weight. The resulting curves are linear and allow estimation of DT densities. The compiled data is shown in Table 3.

We now consider the non-additivity of densities for solution. As with Eqs. (1) and (4), we define this non-additivity as

$$f_\rho = \frac{\rho - \rho_{exp}}{\rho}. \tag{6}$$

Oddly enough, there is less data on densities of solutions than for viscosity and surface tension. Only the system H_2-D_2 has been measured and the data is shown in Fig. 4.[35-37] The function, f_ρ, is negative, indicating an increase in density above that calculated additively. This corresponds to a shrinkage of the mixed volumes. As can be seen from Fig. 4, there is considerable scatter in the data, and we cannot estimate a best value at this time.

Kerr has mixed liquid D_2 and T_2 in 1:1 ratios, and he finds a decrease in density non-additivity.[38] He obtains an f_ρ of +0.7 % at 21 K and +0.2 % at 26K. By substituting our own densities for additive calculation, we find a further increase in Kerr's f_ρ to +1.0% at 21 K to 1.5% at 26 K. This behavior is quite surprising as one would expect all mixtures of hydrogen isotopes to show the density increase found in the three H_2-D_2 sources. Because of the uncertainty in the deviation from additivity, we have used only additively calculated data for the D_2-DT-T_2 mixture listed in Table 3. These figures are the same as those for DT because of the symmetry of the chemical equilibrium with 1:1 D-T present.

Table 3. Liquid density of the hydrogen isotopes from 14 to 25 K.

| Temperature (K) | Triple point of | Liquid density (10^3 kg/m^3)[a] | | | | | |
		nH$_2$ (Refs. 8,27)	HD (Refs. 28,29)	nD$_2$ (Refs. 30,31)	DT	T$_2$[b] (Refs. 32,33)	D$_2$-DT-T$_2$ (1:1 DT)
13.957	nH$_2$	0.0773					
14.00		0.0772					
15.00		0.0763					
16.00		0.0754					
16.50	HD	0.0749	0.1228				
17.00		0.0745	0.1223				
18.00		0.0735	0.1209				
18.71	nD$_2$	0.0728	0.1199	0.1739			
19.00		0.0725	0.1195	0.1733			
19.71	DT	0.0718	0.1184	0.1719	(0.2250)	(0.2772)	(0.225)
20.00		0.0714	0.1180	0.1713	(0.2240)	(0.2761)	(0.224)
20.62	T$_2$	0.0707	0.1170	0.1700	(0.2220)	0.2739	(0.222)
21.00		0.0702	0.1164	0.1691	(0.2200)	0.2724	(0.220)
22.00		0.0690	0.1147	0.1669	(0.2175)	0.2686	(0.218)
23.00		0.0676	0.1130	0.1645	(0.2145)	0.2649	(0.215)
24.00		0.0662	0.1111	0.1621	(0.2110)	0.2611	(0.211)
25.00		0.0647	0.1091	0.1595	(0.2075)	0.2572	(0.208)

[a]Same units as g/cm^3. Parentheses indicate estimates.
[b]Probably cT$_2$.
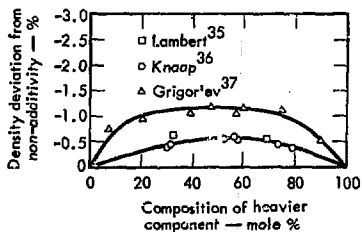

Fig. 4. Density non-additivity of H$_2$-D$_2$ liquid solutions at 20–20 4 K.

It is difficult to estimate accuracies as one proceeds down the chain of estimates. The authors of our fiducial measurements were consulted for their estimates. Their best accuracies are: viscosity: $\pm 0.5\%$; surface tension: $\pm 0.02 \times 10^{-3}$ N/m (± 0.7 to $\pm 2\%$ for H_2 in the range 14–25 K)[19]; and liquid density: $\pm 0.1\%$.[8] The isothermal plots of all three properties are quite linear as a function of molecular weight. The density is especially well known since T_2 has been measured. We may estimate the errors of adjustment and extrapolation to obtain DT and T_2 data to be: viscosity, $\pm 3\%$; surface tension, $\pm 2.5\%$; and density, $\pm 0.6\%$. The last extension to D_2-DT-T_2 mixtures is especially uncertain, because of total lack of knowledge of the non-additivities of ternary solutions. We have been conservative in our estimates of non-additivity, and the possible errors might be of the order: viscosity, $+2\%$, -6%; surface tension, $+1\%$, -3%; and liquid density, $+2\%$, -1%.

Slow Droplet Formation

We have noted that the laser target of the future may well be a DT droplet. We shall use the estimated values of surface tension, viscosity, and density to consider some properties of liquid DT droplets. We shall make our calculations for liquid eD_2-DT-eT_2, with 1:1 atomic D-T, at the estimated triple point of 19.75 K. We use the values estimated above: viscosity, 550×10^{-7} Pa·s; surface tension, 4.23×10^{-3} N/m; and density, 225 kg/m^3.

There are two basic ways to create drops: to slowly drop one at a time or to fire them as a liquid jet. We shall first consider the slow creation of droplets from a capillary tube, for which the control of timing and dimensions is not good. If the capillary is stable, the largest drop that can adhere to the capillary against the force of gravity is[39]

$$a_{max} = \left[\frac{3a_{co}\gamma F}{2\rho g} \right]^{1/3}, \quad (7)$$

where a_{max} is the maximum radius of the suspended drop, a_{co} is the outer radius of the capillary, γ the liquid-vapor surface tension, F the shape factor, ρ the liquid density, and g the acceleration due to gravity. The shape factor, F, is in fact, an empirical correction to the experimental inability to achieve truly maximum size drops. For a truly stable system, F may be larger than 0.6.

Equation (7) applies to a droplet hanging only from the bottom of the capillary. In fact, droplets tend to climb the capillary to increase their contact area. Some continual flow would probably be needed to offset this phenomenon, thus again possibly reducing the shape factor.

Let us assume a small capillary tip of 10-μm diameter and a shape factor of 0.6. For our liquid DT mixture, we obtain a 410-μm-diameter droplet–quite large. This occurs because the low surface tension is offset by the low density and the ratio is diluted by the one-third power argument.

Liquid Jets of Droplets

Probably a better method of creating DT droplets is by the liquid jet method, which produces a stream of very uniformly sized droplets. The generator may be turned on for short periods of time... that it need not produce large numbers of unwanted drops. A method has been developed for charging a single water drop and then electrostatically deflecting it to a specified point.[40–43] The early work in the liquid jet field was done largely with water and organic liquids,

but recent work has extended the method to metals, inorganic salts, and plastics.[44–47] The detailed theory of the method has also received considerable attention.[44,46,47] Most recently, the jet technique has been extended to the creation of liquid hydrogen laser targets by C.D. Hendricks[48] of this Laboratory.

The effect has been known quite a long time–Lord Rayleigh provided the early theory.[49] If a capillary squirting a jet of liquid has an oscillation of the

proper frequency applied, the liquid stream distorts into a wave pattern and then breaks into drops. Surface tension pulls the drops into spherical form, and they continue on the trajectory of the jet. The quantitative treatment of the liquid jet effect can be quite complex, but an impression of the orders of magnitude may be gained from the following simplified set of equations for a low-viscosity liquid.[42] The equations below are given from the point of view of the final droplet radius, a:

$$a_{ci} = 0.66a \tag{8}$$

$$a_j = 0.529a \tag{9}$$

$$u > 2.75 \left(\frac{\gamma}{\rho a}\right)^{1/2} \tag{10}$$

$$f_m = \frac{u}{9.016a_j} = 0.21 \frac{u}{a} \tag{11}$$

$$\Delta P = \frac{18.4\eta \ell u}{a^2} , \tag{12}$$

where a is the radius of the final droplet, a_{ci} the inner radius of the vibrating capillary, a_j the radius of the narrowed liquid jet upon leaving the capillary, u the average flow velocity inside the capillary, γ the surface tension, ρ the liquid density, f_m the optimum applied frequency for droplet break-up and the resulting rate of droplet formation, ΔP the driving pressure down the length of the capillary to create the liquid jet, η the viscosity, and ℓ the capillary length. Equation (10) gives the minimum flow velocity for the liquid jet effect. The jet may be operated at higher flow rates with corresponding increases in the frequency and pressure.

Suppose we want liquid DT droplets of 100-μm diameter. This requires an inner capillary diameter of 66-μm, which will narrow to a 53-μm-diameter jet before droplet break up. The liquid jet phenomenon will begin with a minimum flow velocity of 1.7 m/s, which will produce 7100 droplets/s. It will require only 6800 Pa to maintain this flow in a 1-cm-long capillary.

Oscillation of Drops

When a droplet is created, it will possess at least one tail. This tail will be pulled in by the surface tension until viscosity damps out the oscillation. We want to know how far a droplet need move before the oscillations damp out.

The frequency of oscillation of a non-viscous sphere is[50]

$$f_n = \frac{1}{2\pi} \left\{ \frac{\gamma}{a^3} \left(1 - \frac{\Delta a_{max}}{a}\right) \right.$$
$$\left. \left[\frac{n(n+1)(n-1)(n+2)}{(n+1)\rho + n\rho_g}\right] \right\}^{1/2} , \tag{13}$$

where f_n is the nth order of frequency of oscillation, γ the surface tension, a the equilibrium droplet radius, Δa_{max} the maximum initial deformation from sphericity, n the order of oscillation, ρ the liquid density, and ρ_g the gas density. The first versions of this equation were derived by Lord Rayleigh[51] and Sir Horace Lamb.[52] For liquid DT, $\rho \gg \rho_g$. We may further simplify by considering the modes of oscillation. The zero-order mode represents a translation and is trivial. The first-order mode is a pulsation and is largely forbidden for almost incompressible liquids. Hence, it is

the second-order dumbbell oscillation that is of prime consideration. We reduce to the form:

$$f_2 = \frac{1}{2\pi} \left\{ \left(1 - \frac{\Delta a_{max}}{a}\right) \frac{8\gamma}{\rho a^3} \right\}^{1/2} . \tag{14}$$

The ($\Delta a_{max}/a$) term is actually an empirical correction for large amplitude oscillations.

The next step is to consider the viscous damping force as a perturbation of f_2. The degree to which this perturbation shifts the undamped frequency may be inverted to give the time constant for the exponential decay of the oscillations due to viscosity. This was first computed by Sir Horace Lamb for the oceans of the earth (the oscillations are small enough to be considered a surface phenomenon).[52-54] His result, for the second-order oscillation is (without the a_{max} term):

$$\tau_2 = \frac{\rho a^2}{5\eta} , \tag{15}$$

where τ_2 is the first-order damping time constant for the second-order oscillation, ρ the liquid density, a the radius, and η the viscosity. Suppose we wish to

estimate the length of time it takes for the oscillation amplitude to decay to less than some fraction of the radius. We use the exponential decay:

$$\frac{\Delta a}{a} = \frac{\Delta a_{max}}{a} \exp (- t/\tau_2).$$ (16)

If the initial deformation was 20 % and we want $< \frac{1}{2}$ %, then we need wait about four time constants. For a drop falling under the influence of gravity, the distance the drop falls, y, is[55]

$$y = \frac{g}{2} t^2,$$ (17)

where g is the acceleration due to gravity and t the time. For liquid DT drops of 100-μm diameter and a 20 % initial deformation, we find an oscillation frequency of about 5000 Hz and a damping time constant of only 2 ms. If we wait four time constants, a drop will fall only 330-μm under the effect of gravity and will move only 1.4 cm in the minimum velocity liquid jet calculated above. It appears that liquid oscillations will rapidly damp out.

Acknowledgments

References

1. P. C. Souers, R. T. Tsugawa, and R. R. Stone, *Fabrication of a Glass Microballoon Laser Target*, Lawrence Livermore Laboratory, Rept. UCRL-51609 (1974).

2. P. C. Souers, R. T. Tsugawa, and R. R. Stone, *Manufacture of DT-Filled, Hollow Glass Microsphere Laser Targets*, Lawrence Livermore Laboratory, Rept. UCRL-76151 (1974). To be published. *Revs. Sci Instr.*

3. R. R. Stone, D. W. Gregg, and P. C. Souers, *Nondestructive Inspection of Transparent Microtargets for Laser Fusion*, Lawrence Livermore Laboratory, Rept.-76113 (1974). To be published. *J. Appl Phys.*

4. J. Emmett, J. Nuckolls, and L. Wood, *Scientific American 230* (6), 24 (1974).

5. P. C. Souers, R. G. Hickman, W. Z. Wade, and R. T. Tsugawa, *A Simple Model of D_2-DT-T_2 Equilibrium at Cryogenic Temperatures*, Lawrence Livermore Laboratory, Rept. UCRL-51681 (1974).

6. P. C. Souers, R. G. Hickman, W. Z. Wade, and R. T. Tsugawa, *Estimated Infra-red Spectra of Cryogenic D_2-DT-T_2*, Lawrence Livermore Laboratory, Rept. UCRL-51674 (1975).

7. W. M. Jones, *J. Chem. Phys. 17*, 1062 (1949).

8. D. E. Diller, *J. Chem. Phys. 42*, 2089 (1965).

9. N. S. Rudenko and V. G. Konareva, *Soviet Physics – JETP 21*, 509 (1965).

10. N. S. Rudenko and V. G. Konareva, *Russ J Phys Chem 37*, 1493 (1963).

11. N. S. Rudenko and V. G. Konareva, *Russ J Phys Chem 40*, 1058 (1966).

12. V. G. Konareva and N. S. Rudenko, *Russ J Phys Chem 41*, 1289 (1967).

13. N. S. Rudenko and V. G. Konareva, *Soviet Physics JETP 22*, 313 (1965).

14. H. M. Roder, G. E. Childs, R. D. McCarty, and P. E. Angerhofer, *Survey of the Properties of the Hydrogen Isotopes Below Their Critical Temperatures*, National Bureau of Standards, Washington, D.C.. Technical Note 641 (1973), Section 5.5. This source is hereafter referred to as NBS-641.

15. J. D. Rogers and F. G. Brickwedde, *Physica 32*, 1001 (1966).

16. K. N. Linov'eva, *Soviet Physics JETP 7*, 421 (1958).

17. V. N. Grigor'ev, *Soviet Physics JETP 20*, 321 (1965).

18. V. N. Grigor'ev and N. S. Rudenko, *Soviet Physics JETP 20*, 63 (1965).

19. Y. P. Blagoi and V. V. Pashkov, *Soviet Physics JETP 22*, 999 (1966).

20. V. N. Grigor'ev, *Soviet Physics JETP 18*, 72 (1964).

21. V. N. Grigor'ev, *Soviet Physics – Technical Physics 10*, 266 (1965).

22. NBS-641, Sec. 5.1.

23. Walter J. Moore, *Physical Chemistry* (Prentice-Hall, Englewood Cliffs, N.J.. 1963), 3rd ed., pp. 730–731.

24. R. J. Corrucini, *Surface Tensions of Normal and Para Hydrogen*, National Bureau of Standards, Washington, D.C., Technical Note No. 322 (1965).

25. H. Kummerlingh-Onnes and H. A. Kuypers, *Commun. Phys. Lab. Leiden 13* (142d) (1914).

26. NBS-641, Sec. 2.2.

27. NBS-641, Table 5-5.

28. N. S. Rudenko and V. P. Slyusar, *Russ. J. Phys. Chem. 43*, 434 (1969).

29. NBS-641, Table 2-11.

30. R. Prydz, *The Thermodynamic Properties of Deuterium*, National Bureau of Standards, Washington, D.C., Rept. 9276 (1967).

31. NBS-641, Table 2-7.

32. E. R. Grilley, *J. Amer. Chem. Soc. 73*, 5307 (1951).

33. NBS-641, Table 2-9.

34. NBS-641, Table 5-4.

35. M. Lambert, *Phys Rev Lett 4*, 555 (1960).

36. H. F. P. Knaap, M. Knoester, and J. J. M. Beenakker, *Physf 27*, 309 (1961).

37. V. N. Grigor'ev and N. S. Rudenko, *Soviet Physics JETP 13*, 530 (1961).

38. E. C. Kerr, *J. Amer. Chem. Soc. 74*, 824 (1952).

39. G. Manfre, *J. Appl Phys. 37*, 1955 (1966).

40. J. M. Schneider and C. D. Hendricks, *Rev. Sci. Instr. 35*, 1349 (1964).

41. J. M. Schneider, N. R. Lindblad, and C. D. Hendricks, *J. Colloid Sci. 20*, 610 (1965).

42. N. R. Lindblad and J. M. Schneider, *J. Sci. Instr. 42*, 635 (1965).

43. N. R. Lindblad and J. M. Schneider, *Rev. Sci. Instr. 38*, 325 (1967).

44. R. J. Pfeifer and C. D. Hendricks, *Phys Fluids 10*, 2149 (1967).

45. T. Erin and C. D. Hendricks, *Revs Sci Instr 39*, 1269 (1968).

46. C. D. Hendricks and S. Babil, *J. Phys E 5*, 905 (1972).

47. J. M. Schneider, N. R. Lindblad, C. D. Hendricks, and J. M. Crowley, *J. Appl Phys. 38*, 2599 (1967).

48. C. D. Hendricks, Lawrence Livermore Laboratory, private communication (1975).

49. Lord Rayleigh, *Proc. Lond. Math. Soc. 10*, 4 (1879).

50. R. R. Schroeder and R. C. Kintner, *American Inst. Chem. Eng. J. 11*, 5(1965).

51. Lord Rayleigh, *The Theory of Sound*, (Dover Co., New York, 1945), vol. I, pp. 371–375.

52. Sir Horace Lamb, *Hydrodynamics* (Dover Co., New York, 1932), 6th ed., pp. 473–475.

53. S. Chandrasekhar, *Proc. London Math Soc. 9*, 141 (1959).

54. W. H. Reid, *Quart J. Appl. Phys. 18*, 86 (1960).

55. F. W. Sears, *Mechanics, Wave Motion, and Heat* (Addison-Wesley, Reading, Mass. 1958), pp. 82–86.

APPENDIX A: Definition of Symmetric and Antisymmetric

The terminology symmetric (sym) and antisym-metric (asym) refers to the even or odd rotational states of the hydrogen molecule respectively, as listed in Table A-1. At low temperatures, molecular hydrogen is either in the ground (symmetric) state or the first excited (antisymmetric) state. Moreover, the transition from the first excited to the ground rotational state is largely forbidden by the symmetry of the H_2, D_2, and T_2 molecules. Upon cooling, their first, excited levels are trapped in metastable states, unless a catalyst is present. The species HD, HT, and DT also have symmetric and antisymmetric rotational energy levels, but the lack of molecular symmetry allows rapid equilibration.

When the metastable behavior of asym-H_2 and asym-D_2 was first discovered, researchers thought there were two "kinds" of hydrogen. The kind predominant at room temperature was called "ortho" (o); the other kind "para". As can be seen in Table A-1, the definitions of oH_2 and oT_2 are the same, but the definition of oD_2 is the opposite. The species HD, HT, and DT are not defined at all. The ortho-para terminology is widely used but is so confusing that we employ the spectroscopic names, which are identical for all the isotopic species.

The high-temperature sym-asym equilibrium is regulated by another physical phenomenon. The rotational levels of H_2, D_2, and T_2 are locked to the molecular nuclear magnetic moments. The high-temperature equilibrium is, therefore, set by equal distribution across the number of nuclear magnetic states indicated in Table A-1. Because the molecular nuclear spin, I, of D_2 is different from that of H_2 and T_2, the room (essentially "high") temperature behavior and subsequent difference of ortho-para definitions occurs.

Most actual samples of hydrogen are mixtures of sym and asym. The equilibrium mixture at room temperature is called "normal (n)." The equilibrium mixture at any other temperature is called "equilibrium (e)." H_2 and D_2, if cooled, will remain essentially normal for days, unless deliberately catalyzed to the equilibrium form. T_2, however, will reequilibrate in hours because of the catalyzing effects of the beta particle. [A-3,A-4] The species HD, HT, and DT quickly lose rotational energy upon cooling and will assume some equilibrium mix of sym and asym rotational states at any given temperature.

Table A-1. Definition of sym and asym hydrogen.

Rotational quantum numbers, J	% at 0 K		Molecular nuclear spin, I	No. of magnetic states, $2I + 1$	% at high temperature	Nuclear magnetic resonance active?
H_2 and T_2			$(1/2 + 1/2)$			
0, 2, 4, . . . Symmetric (sym)/even	100	Para (p)	0	1	25	No
1, 3, 5, . . . Anti-symmetric (asym)/odd	0	Ortho (o)	1	3	75	Yes
D_2			$(1 + 1)$			
0, 2, 4, . . . Symmetric (sym)/even	100	Ortho (o)	0,2	1,5	66 2/3	No,Yes
1, 3, 5, . . . Anti-symmetric (asym)/odd	0	Para (p)	1	3	33 1/3	Yes
H_2, D_2, and T_2						
		Normal (n)	The equilibrium mixture of sym-asym forms at room temperature (close to "high temperature" %).			
		Equilibrium (e)	The equilibrium mixture of sym-asym forms at any temperature except room temperature.			

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References

A-1. G. N. Lewis and M. Randall, *Thermodynamics*, revised by K. S. Pitzer and L. Brewer (McGraw-Hill, New York, 1961), 2nd ed., pp. 595–600.

A-2. NBS-641, Sec. 9.1. See Ref. 14.

A-3. R. Frauenfelder, F. Heinrich, and J. B. Olin. *Helv Phys Acta 38*, 279 (1965).

A-4. E. W. Albers, P. Hartech, and R. R. Reeves, *J. Amer Chem Soc 86*, 204 (1964).