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**DENSITY OF TRIETHANOLAMMONIUM NITRATE
AND LIQUID PROPELLANT**

RONALD A. SASSÉ

DECEMBER 1988

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The density of hydroxylammonium nitrate based liquid propellants has been evaluated with particular attention given to the system 1845 containing triethanolammonium nitrate as fuel. It was found that density relationships of such systems could be correlated to the molten density of ingredients as opposed to crystalline density values. Equations are presented relating densities to concentrations and satisfactory comparisons are made to experimental measurements. <i>Keywords: Liquid Gun Propellants.</i>					
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I. INTRODUCTION

For several reasons, this author was convinced that it would be both proper and expedient to present an evaluation of the densities of liquid propellant solutions in two parts using the natural divisions of oxidizer and fuel. This approach offers the advantage of following the modes of preparation where the constituents are first prepared as separate solutions which are then mixed to make propellant. This division also afforded the opportunity to bring to print, at an early time, some practical data on the solution characteristics of the oxidizer hydroxylammonium nitrate (HAN) and allowed an extended time period to administratively release data pertaining to the fuel triethanolammonium nitrate (TEAN). In addition, the delay allowed new experiments to be performed and new ideas evaluated.

II. BACKGROUND

Eli Freedman (BRL-Emeritus) as early as 1981 had considered HAN-based propellants as a mixture of ingredients having linear additive properties and he offered generalized equations relating individual component densities and their respective concentrations to propellant densities.¹ Such predictions were useful, at that time, for estimating some thermodynamic quantities and also gave some anticipated solution densities. In fact, at the start of writing this paper his value of 1.20 g/cm^3 for the density of TEAN was the only value available. He recently sparked further interest in this subject by asking if recent work by Sasse', et al.,² could render better estimates of propellant density. This paper attempts to answer that question affirmatively.

It was recently reported² that the functional relationship between HAN concentration and density is represented by a second order equation that, when extrapolated to pure HAN, intersects the density of melted anhydrous HAN as opposed to the crystalline density value. The following regression equation was derived from several HAN solutions at 20°C , where concentration has units of moles/liter and density has units of g/cm^3 :

$$\rho_{\text{HAN}} = 0.9935 + 0.046630 M - 0.00040 M^2 \quad (1)$$

The kernel of the present approach is to assume the above equation embraces both the non-linear ion-solute and ion-ion interactions and linearly add to this second order function the density/volume/concentration characteristics of various fuels such as TEAN which is used in the liquid propellants 1845 and 1846. Results of these approximations will be compared to experimentally derived density measurements for the propellant 1845.

III. DISCUSSION

To undertake this calculation the densities of several TEAN solutions are required to form a data base much like the one previously used for determining HAN-solution relationships. Fortunately, Gail Stein,³ of the Naval Ordnance Station, made such measurements using four temperatures ($21, 34, 53$ and 69°C) and presented density and viscosity data for three fuels (TEAN, diethanolammonium nitrate and monoethanolammonium nitrate). Table 1 gives her values for the density of TEAN solutions at 21°C .

Table 1. Density of TEAN Solutions

Percent by Weight	Concentration moles/liter	Density g/cm ³
20	0.9952	1.056
40	2.1149	1.122
60	3.3618	1.189
70	4.0375	1.224
80	4.7613	1.263
100*	6.2580*	1.328*
100	6.0789	1.29**
100	6.7857	1.44***

-
- * Extrapolated from solution values
 ** Anhydrous melt
 *** Crystalline value

These data were fitted to two linear regression lines, one in units of weight percent and the other in units of moles, M.

$$\rho_{\text{TEAN}} = 0.985896 + 0.003424 (\text{wt.}\%) \quad (2)$$

or,

$$\rho_{\text{TEAN}} = 1.003784 + 0.0546873 M \quad (3)$$

Excessive digits are given for the coefficients to prevent successive round off errors but obviously the constants are not known to better than one part in a thousand. The extrapolated density at 100% TEAN is 1.328 g/cm³. In contrast, six values for the density of crystalline TEAN gave an average of 1.44 ± 0.01 g/cm³ as measured by Warren Hillstrom of the BRL using a Micromertics automatic helium pycnometer. This sharp difference between the crystalline and extrapolated solution density values for TEAN parallels a similar relationship found for the HAN system.²

To complete the relationships, the density of a liquid melt of TEAN was determined to be 1.29 g/cm³ which of necessity had to be obtained above the melting point of TEAN at a temperature of about 90°C. At this temperature the melt has expanded and does not truly represent just the breaking of crystalline bonds at 20°C. Of the two values of 1.29 and 1.328, the later estimate was selected to better represent solution properties of TEAN and will be used in the calculations that follow. Lastly, the chemical composition of the propellant is required and the "ideal" formulation for propellant 1845 is given in Table 2.

Table 2. The Ideal Chemical Composition of 1845

Species	Composition	
	weight %	moles/liter
HAN	63.23	9.62
TEAN	19.96	1.38
H ₂ O	16.81	13.64

IV. CALCULATIONS

In order to separate the constitutive effects of TEAN and HAN solutions, a liter of propellant is considered, consisting of two parts: one of pure TEAN and the other part a solution of HAN. These volume elements are noted as V_{TEAN} and $V_{\text{HAN-H}_2\text{O}}$.

To estimate the concentration of HAN in this idealized division, the volume of TEAN is subtracted from the propellant volume. Equations (3), (4), and (5) are offered to achieve this result. From the relationship:

$$M_1 V_1 = M_2 V_2 \quad (4)$$

where $M_1 V_1$ represents HAN in the propellant and $M_2 V_2$ represents HAN in its sub-divided volume element.

Expansion first yields,

$$M_1 V_1 = M_2 [V_1 - V_{\text{TEAN}}] \quad (5)$$

and further expansion yields,

$$M_1 V_1 = M_2 \left[V_1 - \frac{M_{\text{TEAN}} (\text{MW}_{\text{TEAN}})}{\rho_{\text{TEAN}}} \right] \quad (6)$$

where M_{TEAN} is the number of moles of TEAN in a liter of propellant or 1.38 moles/liter and MW is the molecular weight of TEAN (212.21).

Substitution gives:

$$9.62 (1000) = M_2 \left[1000 - \frac{1.38 (212.21)}{1.328} \right] \quad (7)$$

Thus the concentration of HAN in the divided system is M_2 or 12.34 moles/liter, and it has a volume of $V_1 - V_{\text{TEAN}}$ or 779.48 cm^3 .

The density of HAN at this concentration is given by applying the second order equation 1 and is 1.5080. The density of propellant $\rho_{1,2}$ is then assumed to be the weighted average of its constituents or:

$$\rho_{1,2} V_{1,2} = \rho_1 V_1 + \rho_2 V_2 \quad (8)$$

expansion yields

$$\rho_{1845} = \rho_{\text{HAN-H}_2\text{O}} \left[\frac{V_{\text{HAN-H}_2\text{O}}}{V_{1,2}} \right] + \rho_{\text{TEAN}} \left[\frac{V_{\text{TEAN}}}{V_{1,2}} \right] \quad (9)$$

and substitution yields

$$\rho_{1845} = 1.5080 \left[\frac{779.48}{1000} \right] + 1.328 \left[\frac{1000-779.48}{1000} \right] \quad (10)$$

and thus the density of 1845 is calculated to be 1.4676 g/cm³. Propellant 1846 is slightly more dilute than 1845; thus, incorporating the chemical composition of 1846 into the equations offered would characterize that systems density.

Messina, et al.,⁴ offered an equation relating the density of 1845 (lot 244) to temperature, T in degrees centigrade, based on data obtained at 5, 25, 45, and 65 degrees. The equation offered was:

$$\rho_{1845} = 1.4695 - 7.1188 \times 10^{-4} (T) \quad (11)$$

which at 20 degrees yields a value of 1.4688 g/cm³. The agreement between this value and the value obtained using Eqs. (4) through (10) gives a result that is better than the number of significant figures used to express concentration. Another equation was offered⁵ that differed from Eq. (11) by about one percent; however, that equation was the result of experiments conducted at but two temperatures and the experimenters used a laboratory prepared sample that was not as documented as lot 224. Thus, that equation is not considered the equation of merit.⁶

A simpler calculation predicting the density of propellant can be accomplished by mixing two properly constituted solutions of HAN and TEAN. Once the concentration of one constituent is chosen, within the boundary conditions of the respective crystalline densities, then all required sub-quantities can be calculated. The various inter-relationships are shown by Figure 1 where a line normal to the volume axis cuts the two curves giving the volume and normality of HAN and TEAN required to make a liter of the propellant 1845. For example, an arbitrary line was drawn through the 700 ml HAN and 300 ml TEAN values that also intersected concentrations of 13.7429 m/l and 4.6000 m/l; the required concentrations for HAN and TEAN. Respected densities were given by Eqs. (1) and (3) and a final propellant density of 1.46775 g/cm³ was obtained by employing the weight averaging Eq. (8). This density value agrees with the experimentally measured value and also with the value obtained earlier using Eqs. (4) through (10).

V. CONCLUSION

From the equations and corresponding arguments presented, the density of the system HAN, TEAN and water can be calculated from their respective concentrations. Further, the density of either HAN or TEAN solutions, considered individually, can be also be estimated from their respective concentrations. This may be of benefit to those that concentrate such materials. Such relationships may be of help in the general art of formulation but clearly final concentrations will be established by chemical analysis and not by density.

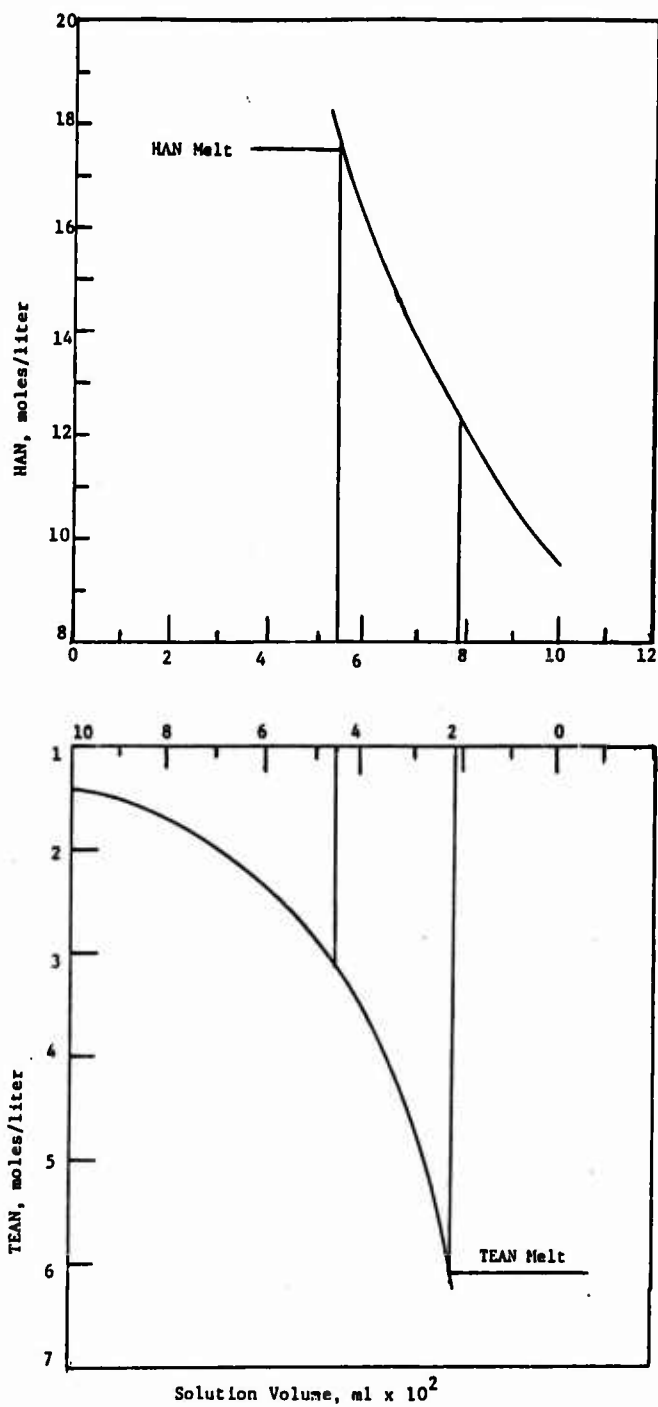


Figure 1. Mixing Solutions of Various Concentrations of HAN and TEAN to Make Propellant 1845

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