PHYSICAL & THERMODYNAMIC PROPERTIES OF HYPERGOLIC PROPELLANTS: A REVIEW AND UPDATE

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ABSTRACT

Significant errors and omissions were found in some of the reported literature values for nitrogen tetroxide, monomethylhydrazine, and Aerozine-50. The methods used to try and resolve some of these errors included (1) a comparison of various literature values, including an assessment of data quality, to determine whether reported values were measured or estimated, (2) a derivation of temperature dependent correlation coefficients and validation with independent measurements (where sufficient measured data were available), and (3) an estimation of the missing parameters using modern techniques such as the method of corresponding states or group contribution methods. Utilizing these methods resulted in a validated set of properties (many as functions temperature) for hypergolic propellants, which are suitable for environmental modeling applications and more general engineering calculations.

The complete parameter set is provided, along with references and examples illustrating the above methods. Also, mixing rules, pseudo-critical properties, and mixture properties are provided for a nominal composition Aerozine-50 mixture.

INTRODUCTION

The objective of this work was to find and validate existing hypergol parameters for use in environmental modeling applications. However, standard references and chemical engineering journals either do not have all of the required data, such as vapor viscosity as a function of temperature, or they contain crudely estimated values and outright errors. Although they contain some of the more hard to find parameters, the current propellant manuals no longer contain the ancillary information regarding data quality, sources, etc. Also, some of the reported data is mis-represented, i.e., if the original experiment was not specifically designed to measure critical properties, it's most likely not an appropriate source for those parameters (especially when the original author has so stated).

This work required a very extensive review of the available reference and technical literature, as well as the use of numerous estimation techniques, both to fill in data gaps and to evaluate multiple reference values for the same properties. Also, many sources neglect to report key information such as whether certain parameters are measured or estimated, or what the uncertainty estimate is for various measurements. The end result is that misinformation gets propagated from one reference to the next, while valuable information gets lost along the way.

RESULTS AND DISCUSSION

PROBLEMS IN THE LITERATURE

The equations and units for each property are shown in Table 1. There are many different functional shapes discussed and used in the literature, based on either theoretical or empirical factors (or both). For many parameters, there are multiple forms presented by different investigators. For this work, the forms for the individual equations are those defined and used by the propellant evaporation model. However, alternative forms and independent data sets were used as a check whenever possible.

Where there were multiple existing values (e.g., certain critical properties), they were evaluated in the light of various factors such as: 1) Were the values reported in the literature measured or estimated? 2) If measured, were the measurements repeatable, or were there experimental difficulties? 3) If estimated, what estimation techniques were used? Also, the results of other calculations involving the values in question were compared to measurements, and those results were used to gauge the data quality. Many other ancillary parameters were used in the

calculations, estimation techniques, etc. These values are shown in Table 2, along with the critical properties and ancillary data required for the various estimation technique used. They are the recommended values for use in engineering calculations and modeling (at or near ambient temperatures and pressures) involving hypergolic propellants. Some typical data integrity problems are cited in the next few paragraphs.

It was somewhat surprising to find so many conflicting values and typographical errors in the main references. For example, the table of critical properties in Lange's Handbook (Dean, 1992) gives the critical pressure of hydrazine as 14.5 atmospheres, which is an order of magnitude too low. Also, the low temperature vapor pressure coefficients for Aerozine-50 given in Schmidt (1984) are missing a digit in the third term, which results in calculated vapor pressures that are also an order of magnitude too low.

Table 3 is an example of the available critical property data for monomethylhydrazine (MMH). The critical molar volume (V_c) and critical density (d_c) are equivalent quantities (i.e., one can be derived from the other); derived values are shown in italics. Some quantities show little scatter, while there is a large difference between the V_c given in Reid, et al (1987) and the d_c given by Barger (1958). Many later references (including CPIA/M4) give the values reported by Aston, et al (1951), and imply they are measured values, even though Aston specifically states that they are estimated and "should not be compared to measurements." In fact, Aston, et al, were not trying to measure critical properties, yet their work is repeatedly cited by later references as a source for MMH critical temperature and pressure (albeit completely inappropriately). Other references cite the work of Aerojet (Barger, 1958) as the "best" values, however, the USAF Propellant Handbook (March & Knox, 1970) specifically states "the values obtained by Aerojet are accepted, but only as reasonably good estimates due to experimental difficulties encountered from decomposition."

Differences in the physical properties of N_2O_4 versus Mixed Oxides of Nitrogen (MON-3, MON-10, etc.) were also investigated. The MONs are designated based on the nitric oxide (NO) content, i.e., MON-1 has 1% NO, MON-3 has 3% NO, etc. Although the physical and thermodynamic properties of MON-10 and MON-25 are quite different than N_2O_4, the physical properties, vapor pressure correlations, etc., for N_2O_4 are considered adequate to describe the characteristics of MON-1 and MON-3 (Wright, 1977), therefore the MONs were not included in this work. Unfortunately, the physical properties given in Chapter 14 (Nitrogen Oxides) of CPIA Pub. 394 are incorrect; the MON-10 properties and N_2O_4 properties were mixed up and included in the table describing MON-1 and MON-3. Also, some of the information presented in the table of N_2O_4 properties in CPIA Pub. 394 is incorrect.

Estimated critical properties for the hypergols were calculated, via Ambrose's group contribution method (see Reid, et al, 1987) and are shown in Table 4, along with the best available literature values. Both sets of values were then used to calculate low pressure gas viscosities (also shown in Table 4) in an attempt to validate the original model viscosity values. The only literature source of hydrazine vapor viscosities turned out to contain rather simple estimates (i.e., the estimation technique used was much less rigorous than those described in Reid, et al). The most reliable values available appear to be those calculated from the measured critical properties.

HYPERGOL CORRELATION COEFFICIENTS

All model parameters are shown in Table 5. These include molecular weight, boiling and freezing temperatures, and various vapor and liquid properties (as well as the applicable temperature ranges) required by the propellant evaporation model. Table 6 gives the data sources and estimation techniques used. Validated or estimated properties for pure quantities are discussed below, while the mixture properties for Aerozine-50 are discussed in the following section.

Most of the standard references (e.g., Lange's Handbook, Perry's Handbook, etc.) contain data and/or correlation coefficients for typical organic (alcohols, esters, etc.) compounds, inorganic compounds, and common substances. However, hypergols are apparently uncommon enough that it becomes increasingly difficult to find appropriate data/correlations, especially for vapor properties. In this case, there were no available measurement data or correlation coefficients found for vapor viscosities. Although modern estimation techniques are generally accurate, most of them assume they will be used to estimate properties of non-polar compounds, so the accuracy tends to degrade as polarity increases. Also, without measured data there is no way to quantify the error in the estimated values. The viscosity values shown in Table 5 are calculated values at 298°K (25°C); the sensitivity of the

model to vapor viscosity did not justify the additional uncertainty in estimating viscosity as a function of temperature.

The correlation coefficients for vapor heat capacity per unit mass (C_p) , or specific heat, are also shown in Table 5. Where reference heat capacity measurements were found, the calculated results have been validated against these data. Existing correlation coefficients were found for nitrogen dioxide (NO_2), but not for nitrogen tetroxide (NTO), so Joback's group contribution method (see Reid, et al, 1987) was used to estimate the NTO vapor heat capacity parameters. Reid, et al (1987) also contained hydrazine (HYD) parameters (based on measured data), so these were verified and used. For the case of monomethylhydrazine (MMH), there were no C_p measurements, nor were there any existing correlation coefficients, so appropriate parameters for the Miller Polynomial were generated from measured data found in Aston (1951). Joback's group contribution method was used for the 1,1-dimethylhydrazine (UDMH) parameters.

The vapor thermal conductivity coefficients for both NTO and HYD were found in Table 10-3 of Reid, et al (1987). The MMH coefficient is a single value, referenced at 298°K, which is the average of the Eucken and modified Eucken methods. This was the recommended approach (Perry, 1984), since the former method tends to underestimate, while the latter tends to overestimate thermal conductivity. The UDMH parameters are a linear fit to measured data (recommended by Schmidt, 1984), while the A-50 parameter is a single value referenced at 298°K.

The equation for heat of vaporization (ΔH_v) as a function of temperature (T) is shown in Table 1, where ΔH_v is in J kg^{-1} and T is in Kelvins. This equation (also known as the Watson relation) is used to scale ΔH_v at the normal boiling point (ΔH_{vb}) to other temperatures, where $C = \Delta H_{vb}$, $A = T_c$ (critical temperature), $B = T_b$ (boiling temperature), and $D = n$, a constant (Reid, et al 1987). The coefficients for NTO, HYD, MMH, and UDMH were all updated with measured data (ΔH_{vb}, T_b, T_c) from Dean (1992), Reid et al (1987), Schmidt (1984), and Giauque & Kemp (1938), with new exponents (D) calculated using Equation 7-12.2 of Reid, et al, (1987). The resulting errors are all 2% or less.

The single case where there is actually an abundance of data is vapor pressure. Numerous data sets were checked against independent data, and, except for a few specific instances, the quality was generally excellent. The data sets shown in Table 5 were chosen based on the available temperature range and the error when compared to the measurements, as well as other factors such as any reported experimental difficulties.

The coefficients for liquid thermal conductivity were taken from both Reid, et al (1987) and Marsh & Knox (1970), while the coefficients for UDMH were derived from Rocketdyne data reported in the latter. The agreement with measured data (where available) was generally very good.

MIXING RULES AND MIXTURE PROPERTIES OF AEROZINE-50

The propellant evaporation model uses an energy balance approach, and iteratively calculates an average pool temperature and evaporation rate. It also assumes the evaporating liquid is a pure substance. This required the derivation of pseudo-critical properties for Aerozine-50, as well as equivalent physical and thermodynamic properties. Although there are some measured data available (e.g., liquid density and vapor pressure), most of the desired properties had to be estimated.

The issue of the appropriate value for the apparent molecular weight of Aerozine-50 may seem at first glance to be a trivial one, however, there are various combining rules for chemical mixtures, and each has a particular application. As seen in Table 7, there are at least four valid methods for calculating the apparent molecular weight of a mixture. The molecular weight calculated from the total moles of the mixture is mass-conserving, and is probably the most appropriate characterization of the properties of the bulk liquid mixture. Because it is mass-conserving, it is also appropriate for converting concentration units of mg/m^3 to parts per million. However, because it is descriptive of the bulk liquid, it may not be appropriate for the saturated vapor calculations in the model. Also, the proper treatment of a liquid mixture is beyond the current basis of the model.

The proposed value for the apparent molecular weight of Aerozine-50 is based on the liquid mole fractions shown in Table 7. In this case, the interaction terms Q_{ij} were calculated via an arithmetic mean (originally proposed by van der Waals) and the binary interaction parameters k_{ij} were set equal to unity. According to Reid, et al (1987), the arithmetic mean is typically used for size parameters, while the geometric mean is used for energy parameters. The above approach yields a value very close to the results of a simple "total moles" calculation.

The actual evaporation of the Aerozine-50 mixture is a much more complex process than the modeled representation. The true boiling curves are shown in Pannetier & Mignotte (1963) for a complete liquid solution series between two end-member components (specifically, hydrazine and UDMH). Assuming a nominal composition Aerozine-50 (50:50 by weight), at 350°K the mixture begins to boil, however, the composition of the vapor is initially approximately 80% UDMH. As temperature (and time) increases, the composition of the vapor follows the vaporus curve, while the composition of the remaining liquid follows the liquidus curve. When the liquid is entirely evaporated, the bulk vapor composition is the same as the initial liquid (i.e., 50:50) but that is only an "average" composition. Since the evaporation model is steady-state (i.e., no time dependence), this process is not reflected in the model calculations. The pseudo-critical properties for a nominal mixture composition Aerozine-50 are given in Tables 2 and 5.

The vapor viscosity value for Aerozine-50 was estimated via the method of corresponding states, with a polar correction factor (as recommended in Reid, et al, 1987). The vapor heat capacity coefficients were estimated via the group contribution method from functional groups given in Perry's Handbook (1984), while the vapor thermal conductivity is a weighted mole fraction average of the component thermal conductivities at 298°K. The heat of vaporization was estimated using Reidel's method (Reid, et al, 1987). The Reidel method was chosen because the calculated values for other methylated hydrazines had the least error when compared to measurements.

The low and high temperature vapor pressure correlations for Aerozine-50 given in Schmidt (1984) appear to be based on measured data, however, the low temperature correlation is missing a digit in the third term. The low temperature vapor pressure correlation given in CPIA/M4 correctly reproduces the reference data at 25°C.

The liquid thermal conductivity coefficients were taken from Schmidt (1984). This correlation was compared to the results of a mole fraction average calculation (using measured values of mixture components), and also compared to the results of two different mixture estimation techniques. The latter three techniques all appeared to over-estimate the thermal conductivity of liquid Ae-50 by about 25%. This is most likely due to the large dipole moment of UDMH, since mixture estimation techniques generally perform much better with non-polar mixtures.

SUMMARY AND CONCLUSIONS

The quantity and quality of available reference data on hypergolic propellants does not approach that of typical organic/inorganic compounds. This seems due to the high toxicity and resulting low industrial use of these compounds, thus most of the existing data is only found in propulsion related technical reports. The lack of peer review for some of the experimental work, coupled with the problems related to high temperature decomposition, have also contributed to the large scatter evident in many of the measurements that do exist. However, if one takes the time to examine some of the older reports, it is possible to extract the high quality measurements, as well as estimate the quality and uncertainty in the bulk of the remaining data. Also, modern estimation techniques, when applied appropriately, can both help to fill the existing gaps, as well as help to gauge the quality of the existing data.

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Table 1. Equation Forms for Temperature Dependent Parameters

Saturated Liquid Density [gms/ml]	$\rho_L = A + B \bullet T + C \bullet T^2$
Liquid Thermal Conductivity [W/m-°K]	$\lambda_L = A + B \bullet T + C \bullet T^2$
Vapor Thermal Conductivity [W/m-°K]	$\lambda_g = A + B \bullet T + C \bullet T^2 + D \bullet T^3$
Vapor Viscosity [Pa•sec]	$\eta_g = A + B \bullet T + C \bullet T^2$
Vapor Heat Capacity [J/kg-°K]	$C_p = A + B \bullet T + C \bullet T^2 + D \bullet T^3$
Heat of Vaporization [J/kg]	$\Delta H_v = C \left(\dfrac{A - T}{A - B} \right)^p$
Saturation Vapor Pressure [mm Hg]	$Log(P) = A - B/(T + C) - D/T^2 + E \bullet T + F \bullet T^2$

Table 2. Hypergol Physical Properties and Ancillary Parameters

Property (symbol)	Units	Compound				
		A-50	N_2O_4	N_2H_4	MMH	UDMH
Critical Temperature (T_c)	°K	593	431.4	653	567	523
Critical Pressure (P_c)	atm	92.5	101	147	82.4	53.6
Critical Volume (V_c)	cm^3/mole	135	167.8	96.1	271.2	230
Critical Density (d_c)	gms/cm^3	0.310	0.548	0.333	0.170	0.261
Critical Compressibility Factor (z_c)	none	0.266	0.473	0.375	0.474	0.272
Pitzer's Accentric Factor (ω)	none	0.396	0.834	0.316	0.425	0.343
Dipole Moment (μ)	debyes	N/A	0.5	3.0	1.68	2.93

Table 3. Critical Property Values for Monomethylhydrazine (MMH)

T_c(°K)	P_c(atm)	V_c(cm^3/mole)	d_c(gm/cm^3)	Source	Comments
567	82.4	271.2	*0.17*	Reid, et al (1987)	measured
567	79.3	271		Dean, ed. (1992)	source is probably Reid (T_c and V_c), source of P_c unknown
585	81.3	*160*	0.29	Barger (1958)	measured (with experimental difficulties), d_c estimated from phase diagram
530	75	n/a	n/a	Aston, et al (1951)	estimated
561	72	155		Arnold (this work)	calculated via Ambrose's group contribution method.

Table 4. Comparison of Measured vs. Estimated Hypergol Critical
Properties and Their Effect on Calculated Viscosity

	Measured/Estimated Critical Properties			Calculated Viscosity	
	T_c(°K)	P_c(atm)	V_c(cm^3/mole)	(pa-sec)	(AFGL)
N_2O_4	431.4	101	167.8	1.05 E-5	1.25 E-5
	443	65.3	196	1.03 E-5	
N_2H_4	653	147	96.1	8.59 E-6	8.57 E-6
	620	115	100	8.32 E-6	
MMH	567	82.4	271.2	5.37 E-6	7.83 E-6
	561	72.9	155	7.86 E-6	
UDMH	523	53.6	230	7.41 E-6	7.35 E-6
	523	53.2	210	7.88 E-6	

Table 5. Temperature Dependent Correlation Coefficients for Hypergols

Description	Units	A-50	N_2O_4	N_2H_4	MMH	UDMH
Molecular Weight	gms/mole	41.8018	92.0110	32.0454	46.0724	60.0994
Boiling Temperature (T_b)	Kelvins	*350	294.25	386.65	360.65	337.05
Freezing Temperature (T_f)	Kelvins	*266	261.95	275.15	220.75	215.15
Liquid Density A	gms/ml	1.17423	2.066	1.02492	1.15034	1.09450
Liquid Density B		-9.2417 E-4	-1.979 E-3	-8.65 E-4	-9.3949 E-4	-1.0343 E-3
Liquid Density C		0	-4.826 E-7	0	0	0
High Temp	°K	340	325	450	360	330
Low Temp	°K	255	265	273	220	205
Vapor Viscosity A	Pa-sec	8.32 E-6	1.05 E-5	8.59 E-6	5.37 E-6	7.41 E-6
Vapor Viscosity B		0	0	0	0	0
Vapor Viscosity C		0	0	0	0	0
High Temp	°K	298	298	298	298	298
Low Temp	°K	298	298	298	298	298
Vapor Heat Capacity A	J/Kg-°K	183.3	150.74	308.78	129.14	-52.08
Vapor Heat Capacity B		6.041	2.7192	5.9904	5.6084	6.137
Vapor Heat Capacity C		-4.811 E-3	-1.4455 E-3	-5.2380 E-3	-2.9038 E-3	-4.010 E-3
Vapor Heat Capacity D		1.595 E-6	3.0866 E-7	1.9046 E-6	0	1.015 E-6
High Temp		400	400	400	500	400
Low Temp		275	270	275	270	270
Vapor Thermal Conductivity A	W/m-°K	.01294	-1.404 E-2	-2.257 E-2	.01522	-5.929 E-4
Vapor Thermal Conductivity B		0	1.108 E-4	1.193 E-4	0	4.283 E-5
Vapor Thermal Conductivity C		0	-3.162 E-8	8.375 E-9	0	0
Vapor Thermal Conductivity D		0	4.485 E-12	-7.956 E-13	0	0
High Temp	°K	298	1670	1670	298	310
Low Temp	°K	298	300	273	298	273
Heat of Vaporization A	°K	593	431.4	653	567	523
Heat of Vaporization B	°K	350	294.25	386.65	360.65	337.05
Heat of Vaporization C	J/kg	7.899 E+5	4.146 E+5	1.269 E+6	8.119 E+5	5.269 E+5
Heat of Vaporization D	None	.387	.441	.405	.402	.387
High Temp	°K	593	431	653	567	523
Low Temp	°K	266	262	275	221	215
Vapor Pressure A	Mm Hg	9.55828	9.00436	-6.50603	7.056791	6.73578
Vapor Pressure B		2820.587	1753.000	653.880	1065.025	875.89
Vapor Pressure C		0	0	0	0	0
Vapor Pressure D		-181627.9	0	0	158905.5	140001.0
Vapor Pressure E		0	-1.18078 E-3	4.7914 E-2	0	0
Vapor Pressure F		0	2.0954 E-6	-4.9886 E-5	0	0
High Temp	°K	345	295	386	360	337
Low Temp	°K	262	262	275	208	238
Liquid Thermal Conductivity A	W/m-°K	3.0139 E-1	2.176 E-1	1.198	1.4246 E-1	7.428 E-2
Liquid Thermal Conductivity B		1.5687 E-4	2.604 E-5	-7.337 E-4	9.2114 E-4	9.639 E-4
Liquid Thermal Conductivity C		-7.0074 E-7	-1.077 E-6	-1.017 E-6	-1.9030 E-6	-2.245 E-6
High Temp	°K	425	415	591	325	340
Low Temp	°K	283	262	275	256	255
Molecular Diffusion Volume		55.63	33.52	18.32	38.84	59.36

*Boiling range: 350-379°K, melting range: 265-267°K

Table 6. Data Sources and Estimation Techniques

	N_2O_4	N_2H_4	MMH	UDMH	A-50
Vapor Viscosity	Estimated using the method of Chung, et al, from measured critical properties (Reid, et al, 1987).	Chung (same as previous)	Chung (same as previous)	Chung (same as previous)	Estimated via the method of corresponding states, with polar correction factor (Reid, et al, 1987).
Vapor Heat Capacity	Estimated using Joback's group contribution method w/$N0_2$ groups (adjusted units).	Coefficients from Reid with 1.3% adjustment for propellant grade N_2H_4 (Marsh & Knox, 1970); also adjusted units.	Generated coefficients from Aston (1951) data (less error than Joback results).	Estimated using Joback's group contribution method (no measured data) .	Estimated via the group contribution method.
Vapor Thermal Conductivity	Coefficients for Miller polynomial from Reid ($N0_2$).	Coefficients from Miller polynomial from Reid.	Single value; average of Eucken methods.	Linear fit to measured data reported in Schmidt (1984).	Weighted mole fraction average of component thermal conductivities at 298°K.
Heat of Vaporization	Uses measured value for Watson coefficient.	Vetere's method.	Vetere's method.	Reidel's method.	Reidel's method.
Vapor Pressure	Giaque & Kemp (problems with multi-set correlation reported in Marsh & Knox (original G&K work in cm Hg).	Multi-set data from Schmidt.	NASA data extends low end (most recent work). Adjusted units.	Marsh & Knox data covers larger temperature range than Lange's.	Use low temp temp coeff's from Marsh & Knox or CPIA/M4 (Schmidt has typo).
Liquid Thermal Conductivity	Coefficients from Reid.	Reid coefficients (Rocketdyne data considered provisional).	Adjusted units from original Rocketdyne units.	Generated coefficients from Rocketdyne data.	Coefficients from Marsh & Knox (adjusted units).

Table 7. Mixture Combining Rules for Apparent Molecular Weight
of Aerozine-50 (50/50 Mixture by weight of N_2H_4 & UDMH)

	N_2H_4 fraction	UDMH fraction	MW_m [gms/mole]
Mass fraction	.5	.5	46.0721
Volume fraction*	.447	.568	42.6779
Liquid mole fraction	.652	.348	41.8018
Total moles	1.56 moles	.832 moles	41.8015

*Note: The final volume of the mixture may not equal the sum of the volumes of the mixture
components.