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An Assessment of Metals as Rocket Fuels

REVIEW ON Mar. 80 N.W. Luft and L.A. WisemanPICATINNY ARSENAL
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An Assessment of Metals as Rocket Fuels.

L4

by

N.W. Luft and L.A. Wiseman.

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CONTENTS.

	<u>Page</u>
1. Summary.	1
2. Introduction.	1
3. Method of Calculation.	2
3.1. Selection of Data.	2
3.2. Computing procedure.	2
3.3. Errors involved.	4
4. Results.	5
5. Discussion.	5
6. Conclusions.	7
7. Bibliography.	8
8. Acknowledgment.	9
Appendix I. Performance Data of Metals with HNO_3 .	10
Appendix 2. Performance Data of Suspensions of metal in n-Decane, with HNO_3 .	11
Appendix 3. Performance Data of Hydrides of metals with HNO_3 .	11
Appendix 4. Composition of Combustion Gases.	12
Fig.1. Performance (S.I.) of metals with Nitric Acid.	
Fig.2. Performance (S.I.) of (1) Suspensions of metal in n-decane and (2) BeH_2 and B_2H_6 with Nitric Acid.	
Fig.3. Performance (density impulse) of suspensions of metal in n-decane with Nitric Acid.	

/1. SUMMARY.

U.S. Naval Air Rocket Test Station
Lake Denmark, Dover, N. J.

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Reference: X.R. 460/40.

1. SUMMARY.

An assessment has been made of the use of the metals Li, Be, B, Mg, Al alone, in suspension in a hydrocarbon, and as hydrides, with the oxidant Nitric Acid. The best performance is obtained with systems based on Be and B, but the increase in performance, compared with a hydrocarbon fuel, is not sufficient to warrant a great deal of research on these fuels, particularly for medium size motors (~5000 lbs. thrust). Some objections to the use of metals, even for lower performance systems, are discussed.

2. INTRODUCTION.

The large heats of combustion of many metals have attracted attention to their possible use as rocket fuels. Only those metals which have a low atomic weight can be considered in this application and even in these cases it must be realised that all the energy cannot be utilised. This is because, above 3000°K., many molecules are appreciably dissociated and, in the case of metal oxides, heats of fusion and evaporation are large. Thus a general review of heats of formation, such as that given by Dartnall (1), is not more than the first step in an assessment of the use of metals. A slightly more elaborate treatment by Saenger (2) neglects dissociation and phase changes and gives, therefore, rather optimistic values for the exhaust velocities with such fuels. The system H₂O/Al. has been discussed by Dartnall (3) and Pike (4) and H₂O/Mg. by Dartnall (3). Borrow (5) has summarized a certain amount of American information, but the combustion temperatures quoted are extremely high, so high that no serious weight can be given to some of the results.

Recently Stosick (6) has given some performance figures for the system liq.O₂/B₂H₆ which are reasonable and agree with those of Huff, Calvert and Erdmann. The optimum specific impulse of this system is 300 secs. but the optimum density impulse is only ~245. This should be compared with the figure of ~280 for the optimum density impulse of HNO₃/n-decane (8). The density impulse is relevant to an assessment of the performance of propellants for Guided Weapons. For such a purpose liquid oxygen has been excluded from consideration (9) and finally Nitric Acid was selected as the oxidant for the naval weapon "Seaslug", partly as the result of the recommendation of the Ordnance Board (10).

Since there was a lack of comparative data on the performance of oxidants with metal compounds and since it was clear that the research effort on Nitric Acid systems would be increased, a comparative series of performance calculations on Nitric Acid with metal fuels, suspensions of metals in hydrocarbons, and two metal hydrides has been carried out.

/3.

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3. METHOD OF CALCULATION.

3.1. Selection of Data.

The available data on the thermochemistry of metals and metal oxides at high temperatures has been critically reviewed by Luft (11). These data are used in the calculations in this report. It has been necessary to assume further that :-

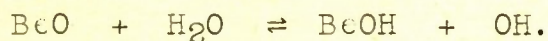
- a) Al_2O_3 exists as such in the vapour. This is unlikely, but as the equilibrium constants for the reactions between probable radicals such as AlO_2 , AlO etc., are unknown there is no alternative. Al_2O_3 , in the vapour phase, was assumed to behave as a chemical molecule, i.e. with a specific heat of ~ 24 cal./mol. deg.C. B_2O_3 vapour was treated similarly. Stosick (6) has made calculations for liq. $\text{O}_2/\text{B}_2\text{H}_6$, in which he considered two cases, viz: B_2O_3 vapour as being monomeric and as being dimeric. The existence of a dimer, $(\text{B}_2\text{O}_3)_2$, at high temperatures is unlikely but, in any case, this makes little difference to his results.
- b) in the combustion of Li with HNO_3 the oxide formed is Li_2O . As there are no data on the specific heat of Li_2O at high temperatures, that of water has been used.

3.2. Computing Procedure.

The following assumptions are made in the calculation of the Specific Impulse:-

- a) The calculations are applied only to systems with excess of oxidant. This has been done for two reasons. Firstly because, under these conditions, equilibria involving metal atoms and molecules, nitrides, carbides and hydrides can be neglected. Secondly, because combustion of metals occurs most readily with excess of oxidant. Thus Fennel (12) has reported up to 10% Al. metal in the exhaust from ram-jets using Al. powder as the fuel.
- b) Full account is taken of phase changes, i.e. melting and volatilisation of the oxide, but dissociation of the oxide in the vapour phase is neglected. To compensate somewhat for this neglect of dissociation phenomena the upper limit for the vapour pressure of oxide (in ref.11) is used. The error involved in these assumptions is small for BeO but may be appreciable at the highest temperatures for the other oxides.

The reason for neglecting dissociation phenomena, apart from the simplification of computing and low accuracy of the available data, is that the number of molecular species present in the vapour phase is unknown. Thus it is stated that BeO is appreciably more volatile, at high temperatures, in an atmosphere of H_2O than in one of N_2 or H_2 (13). This must be due to some chemical reaction, e.g.



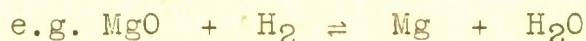
Al_2O_3 and B_2O_3 in the vapour phase must be partially dissociated to AlO and BO . Apart from spectroscopic data on the existence of AlO and BO in the vapour phase, there is other evidence. Thus Zintl (14) has found a method for refining Al_2O_3 and Al by distilling a mixture of crude Al_2O_3 and Al in vacuo. He ascribes this

/to

to the formation of AlO in the vapour phase. Evidence for the existence of aluminium monohydrides in the vapour phase has been given by Gross et alia (15) and thus the existence of AlO in appreciable concentration in the vapour phase is not surprising. Similarly there is evidence for the existence of SiO in the vapour phase (16).

In all these examples the required thermodynamic data for estimating equilibrium constants are uncertain and therefore all dissociation of metal oxide vapours has been neglected. Dissociation of the other gases is taken into account.

c) Reactions of the gases with metal oxides have also been neglected



because oxidising conditions predominate.

d) The expansion of the combustion products has been treated as a frozen equilibrium process. This seems a reasonable assumption in view of the short time available for the formation and growth of nuclei of liquid and solid oxides. It should be pointed out that the formation and growth of nuclei are difficult under such conditions owing to the large heats of fusion and evaporation.

Maxwell, Dickinson and Caldin (17) and Willcock (18) have derived expressions for the exhaust velocity of hot combustion gases, containing solid or liquid particles, expanding through a venturi. In their formulae it is assumed :-

- i) that chemical equilibrium is frozen
- ii) that particles undergo no phase changes during the expansion
- iii) that thermal equilibrium is maintained during the expansion process
- iv) that the particles have everywhere the local gas velocity.

Maxwell et alia (17) have shown that assumptions iii) and iv) are reasonable.

The expression for the jet velocity used in this report is

$$(m/sec) = 91.53 \left\{ \frac{C_p T_c}{M} \left\{ 1 - \left(\frac{P_e}{P_c} \right)^m \right\} \right\}^{\frac{1}{2}} \quad 1.$$

where C_p = mean molar specific heat of all combustion products at constant pressure.

M = mean molecular wt. of combustion products

T_c = combustion temperature in $^{\circ}\text{K}$

P_e = external pressure

P_c = combustion chamber pressure.

$$m = \frac{R}{C_{pg}} \left\{ 1 + \frac{x_s}{x_g} \frac{C_{ps}}{C_{pg}} \right\}^{-1} = \frac{R}{C_p} \cdot \frac{\sum n_g}{\sum n_g + n_s} \quad 2.$$

/s.g.

s.g. as subscripts, refer to particles and gas respectively

$x =$ molar fraction i.e. $x_s + x_g = 1$

x_s applies to the only solid product, i.e. the metal oxide

$x_g = \sum x_i$ where summation is over all gaseous products, i , including the gaseous oxide

$C_{pg} = \frac{\sum n_i C_{pi}}{\sum n_i}$ where summation is over all gaseous products
 n_i being the number of mols. of the i^{th} gas.

This equation is identical with equation (4) given by Willcock (18). The Specific Impulse is obtained by dividing the jet velocity by $g = 981 \text{ cm/sec}^2$. In all these calculations $P_e = 1 \text{ atm.}$ and $P_c = 20 \text{ ats.}$

3.3. Errors involved.

The main objection to the method of calculation used is that no account is taken of the dissociation of the metal oxide in the gas phase. Luft's data (11) indicate that the inaccuracy in the case of BeO is small because the vapour pressure and dissociation constant of BeO are small even at 4000°K. It is impossible to allow for the dissociation of Al_2O_3 , B_2O_3 and Li_2O as the necessary information is not available, but uncertainty due to this cause will arise only at the higher combustion temperatures. In any case the error due to not allowing for dissociation of the metal oxide is partly compensated by the high values for the vapour pressures which have been used.

Neglecting therefore the effect of dissociation of the metal oxide, the main uncertainty will be the vapour pressure assigned to the metal oxide. The equation for the jet velocity can be rewritten in the form

$$\omega = 91.53 \sqrt{F} \quad \text{where} \quad 3.$$

$$F = \frac{C_p T_c}{M} \quad \eta \quad \text{and}$$

$$\eta = \left\{ 1 - \left(\frac{P_e}{P_c} \right)^m \right\} \quad 4.$$

It may be shown that

$$-\frac{\delta \omega}{\omega} = 0.5 (\delta \ln T_c + \delta \ln \eta) \quad 5.$$

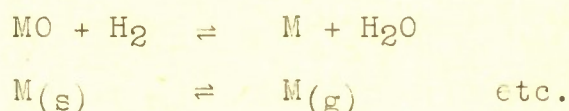
The error in T_c is due either to an inaccuracy in the heats of formation of the reactant and product molecules or an inaccuracy in the vapour pressure of the metal oxide, i.e. in the latent heat of evaporation. Any error in the heats of formation is unlikely to be large. An error in the vapour pressure leading, for example, to a lower value of T_c is partly compensated by the change in η which is in the opposite direction. Thus a lower heat of vaporisation leads to a lower value of T_c but to a higher concentration of metal oxide vapour, which gives an increase in η as can be seen from equations (4) and (2).

/It

It is impossible to state whether equilibrium is maintained or not during the expansion process, although the authors consider, for reasons mentioned previously, that equilibrium is not maintained between gaseous and condensed phases. Huff et alia (7) calculated both cases for the propellants $B_2H_6 + \text{liq. } O_2$, and $B_2H_6 + H_2O_2$, and the maximum difference in S.I. was 4% or less. Since, in this report, the S.I.'s have been calculated with the most unfavourable values (from the point of view of performance) of the vapour pressures of the metal oxides, it is considered that they may be a little low at the maximum S.I.'s but not by more than 5%. At the lower combustion temperatures the error is probably smaller.

4. RESULTS.

Performance figures for the systems HNO_3/metal , with Li, Be, B, Al and Mg. as the metals, are given in Appendix I and Fig.1. All the systems considered have an excess of oxidant. This was stipulated in order to eliminate the necessity of considering equilibria such as



Since, however, only relative performance figures were required this is no serious disadvantage, particularly as the optimum performance figures can be estimated from the curves.

Suspensions of the metals Be, Mg and Al in a hydrocarbon, n-decane, have also been considered as fuels (Appendix 2, Figs. 2 and 3). The concentration of metal is 55 to 60% by weight as this is believed to be about the maximum concentration which will still give a fluid system. The density of the fuel was calculated on the assumption of no volume change on mixing hydrocarbon and metal.

Finally, in Appendix 3 and Fig.2, the performance figures for BeH_2 (hypothetical) and B_2H_6 have been given, zero heats of formation for the metal hydrides being assumed. There is no information on the heat of formation of BeH_2 , and various figures are given for that of B_2H_6 , the latest being $\Delta H_f (B_2H_6) = 6.73 \pm 0.52 \text{ k.cals./ml. (19)}$. The value $\Delta H_f (B_2H_6) = 0$, which is used here, will not lead to any serious error. The density of liq. B_2H_6 is taken to be 0.4818 gr./cc. at $-129^\circ C$. as given in Ref.7.

5. DISCUSSION.

The performance of the systems $HNO_3/\text{n-decane}$ and $HNO_3/\text{liq. } H_2$ are used for comparison purposes. To justify serious consideration any propellant HNO_3/metal must give a significantly better performance than $HNO_3/\text{n-decane}$. Only Be and B satisfy this requirement, B being by far the best of the metals considered.

Suspensions of metal in nitric acid would be possible monopropellants but it is highly unlikely that they would be chemically stable. Thus the rate of penetration of 98-100% HNO_3 into massive Al is about 0.01 ins./year at $50^\circ C$. (20). Such a rate of attack on finely divided Al would render the suspension of Al in HNO_3 valueless. It would also be difficult to stabilise such suspensions physically by the addition of surface active agents, in view of the reactivity and oxidising power of Nitric Acid.

/Suspensions

Suspensions of metals in a hydrocarbon have also been considered (25). In this case the density impulse ($S.I. \times \delta$) is, however, only slightly better than that of the pure hydrocarbon with Nitric Acid (Fig.3) and the gain, except in the case of Beryllium, is not worth further consideration.

Two hydrides have been considered; hypothetical BeH_2 , and B_2H_6 . B_2H_6 shows a slight improvement on n-decane but the difference in optimum S.I.'s is only 10% to 15%. The density impulse is, however, little, if at all, better. Pentaborane B_5H_9 would make a better fuel than Diborane because of its more convenient physical properties. Thus its melting point is $-46.6^\circ C.$, boiling point $+48^\circ C.$, and density, at $0^\circ C$, 0.61 (21). There are, however, serious objections to the use of metal or metallic compounds as fuels. These are :

i) It is a difficult problem to make suspensions of metal in liquids such as water or Nitric Acid chemically stable on account of the enormous specific surface of the metal. Thus rates of reaction, which are negligible with the metals in bulk, are serious with finely divided metals.

ii) Physical stability of a suspension of metal in a hydrocarbon is difficult to produce, and constitutes a formidable research problem.

iii) The combustion of some metals is not easy. Combustion of Mg or Li should not be difficult but Fennel (12) has shown that, in Ramjet combustion of Al powder, there is an appreciable amount of unburnt Al in the combustion products and a number of American firms (Ref.22, p.20) have found Diborane disappointing as a rocket fuel since it requires a combustion chamber having a large L^* and tends to give free boron due to incomplete combustion or thermal cracking.

iv) The combustion temperatures are high and thus the cooling of the motor becomes a more difficult problem. The cooling problem may still further be aggravated by the formation of oxide layers (with release of the latent heat of fusion or evaporation) but Miss Schilling at R.A.E., Farnborough has shown that it is possible to overcome this by sweat-cooling. Oxides may also tend to choke the venturi.

v) The exhaust products will contain large quantities of metallic oxides. Apart from the obvious disadvantages, the oxide (with probably some unburnt metal) may form an anti-radar screen in the sky with consequent difficulties in guiding the missile.

Finally a word should be said about Beryllium. This is the most promising of the metals (together with Boron) but it is reported that some of its compounds are highly toxic although the available information is not definite (23). However, it is very probable that other demands (such as Cu/Be alloys) will preclude its use as a propellant. The quantity available is certainly not more than a few hundred tons per annum (23). This is based on an estimate for the U.S.A.

/6. CONCLUSIONS.

6. CONCLUSIONS.

The main purpose of this note is to determine whether the performance of the propellant, HNO_3 /Hydrocarbon, can be appreciably improved by the use of metals in suspension. The application in view was to medium thrust weapons (~ 5000 lbs. thrust) for which Density Impulse is more important than Specific Impulse. The only metals which would give an appreciable increase in density impulse are Be and B and, apart from other difficulties discussed in the previous section, the supply of these metals is inadequate at the moment.

B_2H_6 shows some promise (max. S.I. ~ 250 secs.) but the density impulse is not particularly good (~ 280). Should manufacture of this fuel, or pentaborane, be satisfactorily developed it seems more reasonable to use it in a high energy propellant with an oxidant such as liq.O_2 or with the fluorine group of oxidants.

It is possible that BeH_2 , if it can be made, will have a high positive heat of formation (24). This will give a better performance (10-15%) in bipropellant systems but such a compound would be far more useful as a monopropellant and will not, therefore, be further discussed here.

A considerable amount of experimental work is being done at R.A.E. on the monopropellant $\text{Al/H}_2\text{O}$ (i.e. suspension of Al in H_2O). This is being investigated primarily, it is believed, for use in ramjets. The serious problems of physical and chemical instability of the suspension may be overcome but early consideration should be given whether the smoke can be tolerated in the application of metal fuels in rockets (e.g. visibility, radar screening).

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The authors are indebted to Miss M.A. Hooper for carrying out the numerous and tedious calculations in this memorandum.

/Appendix I.

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APPENDIX I.

Performance Data of metals with HNO₃.

a) Beryllium.

<u>M.R.</u>	<u>T°K.</u>	<u>S.I. (secs)</u>	<u>S.I. x δ</u>
19.32	2000	159	243
15.54	2400	173	266
10.23	2800	183	283
8.99	3000	188	291
7.70	3200	192	299
4.90	3600	203	319
3.27	3800	211	335

b) Boron.

<u>M.R.</u>	<u>T°K.</u>	<u>S.I. (secs)</u>	<u>S.I. x δ</u>
32.64	2000	160	246
26.46	2400	175	269
21.26	2800	188	290
16.83	3200	200	310
13.22	3600	212	330

c) Magnesium.

<u>M.R.</u>	<u>T°K.</u>	<u>S.I. (secs)</u>	<u>S.I. x δ</u>
7.44	2000	153	236
5.94	2400	166	256
4.70	2800	175	190
3.29	3000	175	274
2.33	3200	176	279
1.33	3400	174	279

d) Aluminium.

<u>M.R.</u>	<u>T°K.</u>	<u>S.I. (secs)</u>	<u>S.I. x δ</u>
9.05	2000	154	244
6.59	2400	165	265
5.18	2800	175	287
3.92	3000	178	297
2.58	3200	180	311

e) Lithium.

<u>M.R.</u>	<u>T°K.</u>	<u>S.I. (secs)</u>	<u>S.I. x δ</u>
8.55	2000	158	201
5.81	2400	173	207
3.86	2800	188	207
2.70	3200	198	201

M.R. = Mixture Ratio = $\frac{\text{wt. of oxidant}}{\text{wt. of fuel}}$

/Appendix 2.

APPENDIX 2.

Performance Data of Suspensions of Metal in n-Decane with HNO₃.

a) Be in n-Decane (55.9 wt. % of metal = 20 Be + C₁₀H₂₂)

<u>M.R.</u>	<u>T^ok.</u>	<u>S.I. (secs)</u>	<u>S.I. x δ</u>
16.32	2000	162	241
12.93	2400	178	263
8.95	2800	191	279
7.65	3000	198	288
6.27	3200	206	298

b) Mg in n-Decane (57.8 wt. % of metal = 8 Mg + C₁₀H₂₂)

<u>M.R.</u>	<u>T^ok.</u>	<u>S.I. (secs)</u>	<u>S.I. x δ</u>
9.64	2000	162	237
7.58	2400	177	258
5.76	2800	191	275
4.12	3000	203	287

c) Al in n-Decane (57.1 wt. % of metal = 7 Al + C₁₀H₂₂)

<u>M.R.</u>	<u>T^ok.</u>	<u>S.I. (secs)</u>	<u>S.I. x δ</u>
10.60	2000	162	241
8.00	2400	176	262
5.79	2800	191	281
4.23	3000	200	291

APPENDIX 3.

Performance Data of Hydrides of Metals with HNO₃.

a) BeH₂. (hypothetical: ΔH_f assumed zero)

<u>M.R.</u>	<u>T^ok.</u>	<u>S.I. (secs)</u>	<u>S.I. x δ</u>
21.52	2000	165	-
17.01	2400	182	-
13.13	2800	198	-
8.22	3200	214	-
6.05	3400	228	-

b) B₂H₆ (ΔH_f assumed zero)

<u>M.R.</u>	<u>T^ok.</u>	<u>S.I. (secs)</u>	<u>S.I. x δ</u>
19.79	2000	168	232
15.70	2400	185	249
12.29	2800	203	266
9.15	3200	221	277
7.68	3400	231	281

Note: The dotted lines in the curves denote that a change of phase of the oxide occurs in that temperature range.

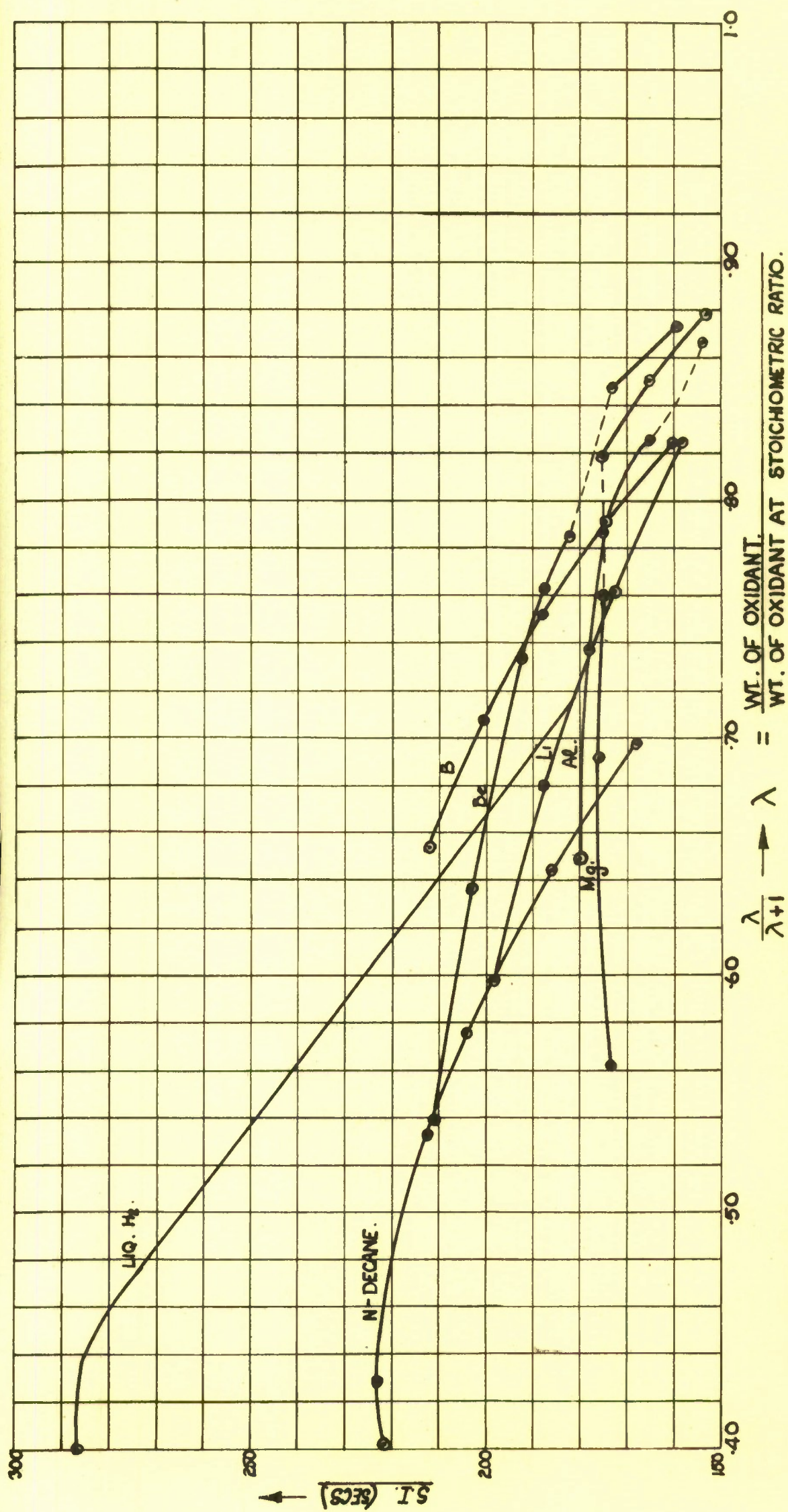
/Appendix 4.

Composition of Combustion Gases.

Propellant		Tc (°K)	Composition of Combustion Products at 20 ats.											
Fuel	Mols.of HNO ₃		O ₂	H ₂	H ₂ O	OH	O	H	N ₂	N	NO	Oxide (c)	Oxide (g)	
1.0 Be	.437	3800	.059	.045	.125	.072	.057	.055	.217	.004	.030	.637	.563	
1.0 Be	2.760	2000	2.95	-	1.375	.010	.001	-	1.38	-	-	1.000	-	
2.0 B	2.268	3600	1.000	.058	.802	.455	.325	.092	1.012	.049	.223	-	1.000	
2.0 B	5.600	2000	5.456	-	2.790	.020	.001	-	2.762	-	.076	-	1.000	
1.0 Mg	.513	3400	.111	.022	.196	.060	.032	.017	.241	.001	.030	.556	.444	
1.0 Mg	2.868	2000	3.061	-	1.429	.010	.001	-	1.413	-	.041	1.000	-	
1.0 Al	1.107	3200	.548	.019	.467	.120	.056	.014	.513	.001	.080	-	.500	
1.0 Al	3.878	2000	4.066	-	1.932	.014	.001	-	1.911	-	.056	.500	-	
1.0 Li	.297	3200	.102	.006	.125	.031	.013	.004	.139	-	.018	-	.500	
1.0 Li	.944	2000	.923	-	.471	.003	-	-	.466	-	.013	.442	.058	

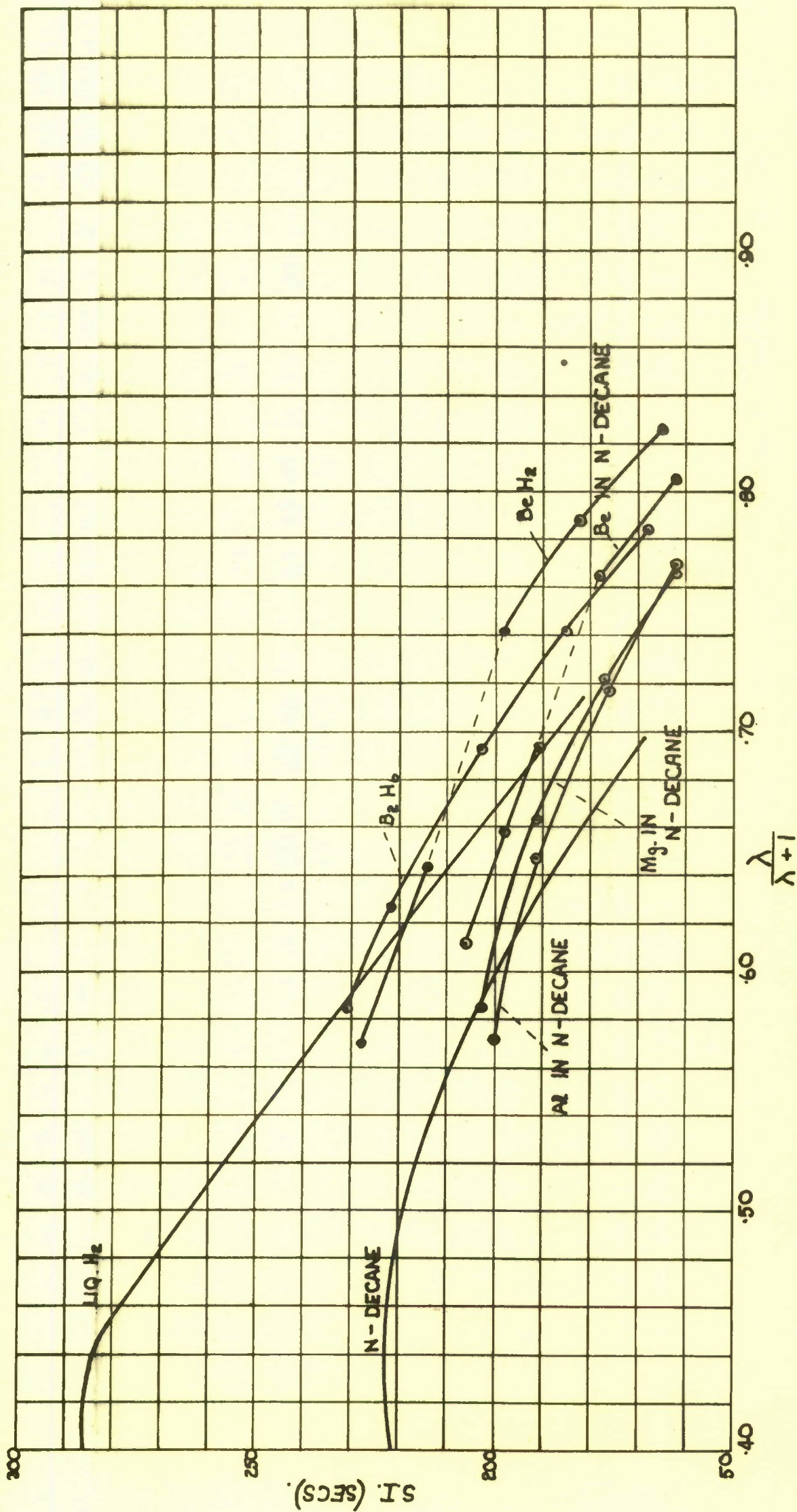
Note: All figures refer to the No. of gr. mols.

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M.No.86/50.

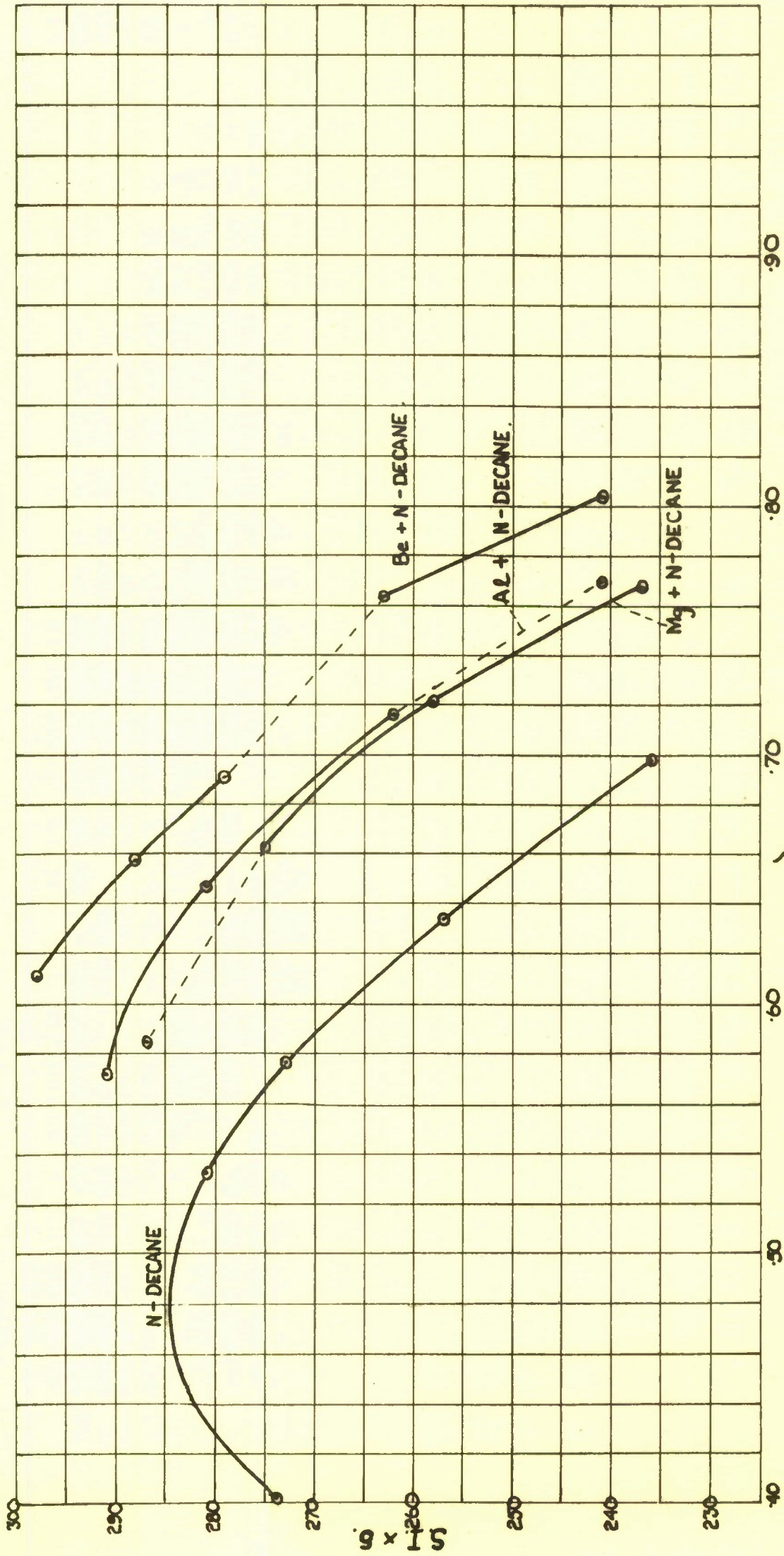


PERFORMANCE (S.I.) OF METALS WITH NITRIC ACID.

FIG. 1.



PERFORMANCE (S.I.) OF (1) SUSPENSION OF METAL IN N-DECANE
AND (2) WITH B_2H_6 AND B_2H_6 WITH NITRIC ACID



PERFORMANCE (DENSITY IMPULSE) OF SUSPENSIONS
OF METAL IN N-DECANE WITH NITRIC ACID.

δ = MEAN DENSITY OF PROPELLANT.

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