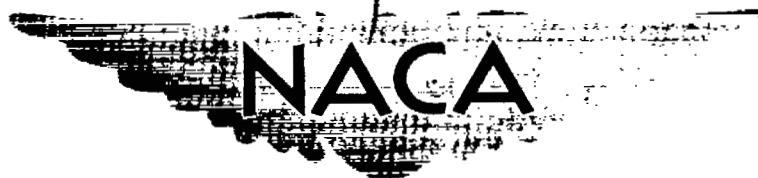NACA RM E58DO3

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RESEARCH MEMORANDUM

CHEMICAL AND PHYSICAL FACTORS AFFECTING COMBUSTION

IN FUEL - NITRIC ACID SYSTEMS

NOT TO BE TAKEN FROM THIS ROOM

By Louis Baker, Jr.

Lewis Flight Propulsion Laboratory
Cleveland, Ohio

FOR REFERENCE

CLASSIFIED DOCUMENT

NATIONAL ADVISORY COMMITTEE
FOR AERONAUTICS
WASHINGTON
July 28, 1958

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NATIONAL ADVISORY COMMITTEE FOR AERONAUTICS

UNCLASSIFIED

RESEARCH MEMORANDUM

CHEMICAL AND PHYSICAL FACTORS AFFECTING COMBUSTION

IN FUEL - NITRIC ACID SYSTEMS*

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SUMMARY

Characteristic exhaust-velocity measurements were made of the JP-4 fuel - red fuming nitric acid propellant combination in 40-pound-thrust rocket engines with various combustion-chamber lengths from 1.5 to 12.3 inches and diameters of 1 and 1.5 inches (characteristic length, 30 to 240 in.). The effect of adding unsymmetrical dimethylhydrazine was determined. Also measured was the effect of added water on the characteristic exhaust velocity of red fuming nitric acid with JP-4, toluene, or hydrazine fuels.

The results, along with those from previous studies, are discussed in terms of a vaporization model of combustion. The experimental trends are consistent with the hypothesis that for spontaneously reacting fuels heat generation within mixed fuel-acid droplets can accelerate combustion; added water has very specific effects in such cases. The combustion rate of nonreactive fuels such as saturated hydrocarbons and petroleum fractions, on the other hand, is controlled by the relative volatility of the fuel and the acid; adding water causes a relatively slight decrease of the characteristic exhaust velocity in this case.

INTRODUCTION

Fuming nitric acid is an important oxidizer, which can be used with a number of readily available fuels. An understanding of the rate-controlling step in the combustion process and the factors affecting it are of great practical importance in rocketry.

Recently, detailed studies have been reported on the combustion of fuming nitric acid with spontaneously igniting fuels (ref. 1), the aniline - furfuryl alcohol system (ref. 2), and hydrocarbon fuels (ref. 3). It will be shown herein that the results of these studies can be interpreted by assuming that droplet vaporization is the rate-limiting step in the combustion process.

UNCLASSIFIED

*Title, Unclassified

This report describes several experiments designed to further test
the vaporization model of combustion discussed in reference 2 and further
discussed in reference 3. The characteristic exhaust velocity c^* of a
JP-4 - nitric acid propellant system was measured under conditions iden-
tical to those used in reference 1 in order to determine the relative
combustion rates of spontaneously igniting fuels with red fuming nitric
acid (RFNA). The effect on c^* produced by adding unsymmetrical dimeth-
ylhydrazine (UDMH) to JP-4 fuel was determined in order to further relate
the hydrocarbon data of reference 3 to the data for self-igniting fuels
of references 1 and 2.

The effect on c^* of water added to either the RFNA or the aniline -
furfuryl alcohol fuel mixture of reference 2 appeared to verify the va-
porization model. Tests were therefore made on the c^* performance with
water added to JP-4 fuel, toluene, or anhydrous hydrazine fuels, all with
RFNA, to learn whether the effect on exhaust velocity was a general one.

The experiments were carried out in a 40-pound-thrust rocket engine
using a high-speed mixing injector (ref. 1).

APPARATUS AND PROCEDURE

The apparatus, instrumentation, and operation are described in de-
tail in references 1 and 3. A schematic drawing of the rocket engine
and injector is shown in figure 1. Combustion chambers having diameters
of 1 and $1\frac{1}{2}$ inches were used. The chamber lengths and their correspond-
ing characteristic lengths L^* are given in the following table:

Combustion-chamber diameter, in.	Length, in.	Characteristic length, L^*, in.
1	3.7	30
	4.9	40
	6.1	50
	9.2	75
	12.3	100
$1\frac{1}{2}$	1.5	30
	12.2	240

Fuels and Acids

The acid was red fuming nitric acid containing from 18 to 19 percent NO_2 and from 0.2 to 2.5 percent water. Several investigations were made of the effect of adding water. In all cases, control runs were made within a few days of the test runs using RFNA containing less than 1 percent water. The same batch of acid was then used with 4 percent water added.

The physical characteristics of the JP-4 fuel are given in table I. Mixtures with UDMH were from commercial grade UDMH and proportioned by weight. The hydrazine was commercial anhydrous hydrazine. The stoichiometric mixture ratios $(o/f)_{st}$ for the fuels are given in the following table:

Fuel	Stoichiometric mixture ratio, $(o/f)_{st}$
Toluene	4.96
JP-4	5.44
JP-4 plus 10% UDMH	5.23
JP-4 plus 40% UDMH	4.61
Hydrazine	1.48

Treatment of Data

The characteristic length L^* was calculated from

$$\frac{\text{Chamber volume}}{\text{Nozzle-throat area}}$$

The characteristic exhaust velocity c^* was calculated from

$$\frac{(\text{Chamber pressure})(\text{nozzle throat area})}{\text{Total flow rate}}$$

The mixture ratio o/f was calculated as

$$\frac{\text{Acid weight flow}}{\text{Fuel weight flow}}$$

In order to compare fuels, the parameter $\beta = (o/f)/(o/f)_{st}$ was used.

RESULTS AND DISCUSSION

The c^* data for JP-4 fuel in 1-inch-diameter combustion chambers are plotted in figure 2 as a function of mixture ratio and L^*. The data are cross-plotted, c^* as a function of L^*, at ratio β of 0.8 in figure 3 from which the characteristic combustion-chamber length L^*_{97} corresponding to a c^* of 97 percent of maximum experimental c^* is found to be about 70 inches. In the following table, the combustor length L^*_{97} required for JP-4 fuel is compared with that for the self-igniting fuels reported in reference 1:

Fuel	Combustor length, L^*_{97}, in.
Hydrazine	19
Trimethyl trithiophosphite	27.5
Furfuryl alcohol	31
UDMH	37
JP-4	70
Allylamine	75
o-Toluidine	83

The fuel JP-4 has a considerably slower combustion rate than the first four fuels and approximately the same rate as allylamine and o-toluidine.

The self-igniting fuels of reference 1 burned more rapidly in fuel-rich mixtures than in acid-rich mixtures. This characteristic as well as the higher combustion rates of some of the fuels as compared with JP-4 fuel can be explained by taking into account the liquid-phase heat generation. Calculated evaporation rates based only on convective heat transfer would predict much smaller differences in combustion rates than are shown in the previous table, and would reverse the order of the combustion rate in hydrazine and JP-4 fuel. Liquid-phase heat generation could increase evaporation rate directly as well as by droplet shattering. In reference 1, hydrazine is shown to be most reactive of the previously mentioned fuels in the liquid phase in agreement with the observed combustion rate. The abnormally large decrease in c^* with an increase in water content of fuel or acid reported in reference 2 is attributed also to a decrease in liquid-phase heat generation.

Liquid-phase heat generation should be most pronounced in fuel-rich mixtures. Ignition-lag studies with a triethylamine - white fuming nitric acid mixture (ref. 4) showed that the shortest ignition lags, and, hence, the maximum liquid-phase heat generation, occur in fuel-rich mixtures. This results from the fact that the stoichiometric mixture ratios for liquid-phase reactions such as neutralization, nitration, and partial

oxidation are more fuel-rich than those for complete combustion. Hydra-
zine, of course, presents an exception to this generalization. Most
fuels showing a high degree of liquid-phase reactivity, therefore, should
have relatively high combustion rates in fuel-rich mixtures, as was ob-
served in reference 1.

Effect of UDMH

Runs in short chambers $(L^* = 30$ in.) were made in $1\frac{1}{2}$-inch-diameter
chambers with 10 and 40 percent UDMH in JP-4 fuel. Runs in long chambers
$(L^* = 240$ in.) were made in the $1\frac{1}{2}$-inch-diameter chambers only with the
40 percent UDMH mixture. The results are shown in figure 4 along with
the data for neat JP-4 taken from reference 3. The UDMH mixtures have
widely differing stoichiometric mixture ratios with RFNA so that the c^*
data are compared in terms of β rather that o/f.

The long-chamber c^* with the 40-percent UDMH mixture exceeds that
of the pure JP-4 fuel by about 4 percent. In the short chambers, the
c^* values approach each other near the stoichiometric mixture ratio
$(\beta = 1)$, while in fuel-rich mixtures, the c^* of the 40-percent mixture
exceeds that of pure JP-4 fuel by as much as 20 percent. The high per-
formance of the 40-percent blend in fuel-rich mixtures can probably be
attributed to liquid-phase heat generation within mixed droplets of fuel
and acid, as discussed previously.

Effect of Water on Performance of Hydrocarbon Fuels

The effect of 4 percent water added to RFNA on the c^* of toluene
and JP-4 in a short chamber $(L^* = 30$ in.) is shown in figure 5. The
data for the neat acid is again taken from reference 3. The experimen-
tal c^* decreased approximately 7 percent in both cases. The data were
obtained with mixture ratios near 3 so that the water added to the pro-
pellant system was about 3 percent. Although the results are not con-
clusive, it can be argued that the decrease is smaller than that which
would be obtained with propellants having a high degree of liquid-phase
reactivity.

A reduction in c^* of approximately 7 percent is reported in ref-
erence 2 for furfuryl alcohol - aniline mixtures and RFNA with 3 percent
added water. The engines used in reference 2, however, were longer
$(L^* = 73.5$ in. as compared with 30 in. in the present investigation),
and the c^* values for the undiluted propellants were higher (4450
ft/sec as compared with 3800 and 3300 ft/sec). The effect of the water
addition noted in reference 2 was therefore greater since the water ef-
fect should become very small in long engines that give experimental

values of c^* close to calculated theoretical values. The decrease in theoretical c^* calculated for the addition of 3 percent water is less than 1 percent (ref. 2).

In reference 2, the effect of water was attributed to a decreased rate of heat generation within mixed droplets, since the same effect was noted whether the water was added initially to the fuel or the acid, and since known ignition catalysts such as ferric nitrate overcame the effect of water. It is concluded in reference 3, however, that there is a negligible rate of liquid-phase heat generation between ordinary hydrocarbon-type fuels and RFNA. The relatively small c^* decrease, which resulted from addition of 3 percent water to the propellants, could be due to a decrease in vaporization rate if it is assumed that the heat-transfer rate to the drops remains constant and that the total enthalpy of vaporization increases slightly.

### Effect of Water on Performance of Hydrazine Fuel

The effect of water on the c^* of the RFNA - anhydrous hydrazine propellant combination is shown in figure 6. The lower curve represents runs made without added water. On the upper curve the square data points represent runs made with 4 percent of water added to the RFNA and the triangular data points represent runs made with 8 percent of water added to the hydrazine. The total amount of water added to the system varied from 1.4 to 3.3 percent depending upon whether the water was added to the fuel or the acid. The c^* performance increased nearly 12 percent in all cases.

The large effect on exhaust velocity is undoubtedly due to changes in the rate of liquid-phase heat generation. The rapid nature of the liquid-phase reactions between hydrazine and nitric acid is demonstrated by ignition-delay measurements in reference 5 and by the extreme cup pressure drop developed in the injector of reference 1. Both references show that very great heat liberation results within 1 millisecond after the initial contact of the propellants.

The addition of small amounts of water to the hydrazine - nitric acid system apparently increases the rate of liquid-phase heat generation; conversely, the effect on the furfuryl alcohol - aniline and nitric acid system is to decrease the rate. This behavior suggests that there are fundamental differences in the reaction mechanism. It is reasoned in reference 2 that the NO_2^+ ion might be involved in the reaction of the acid with the furfuryl alcohol - aniline mixture, that is, that the reaction is a nitration. Addition of a small percentage of water greatly reduces the NO_2^+ ion concentration according to the following equation (ref. 6):

$$2HNO_3 \rightleftharpoons NO_2^+ + NO_3^- + H_2O \qquad (1)$$

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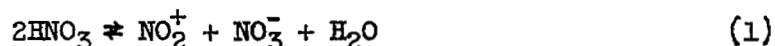
In the hydrazine reaction, a compensating effect of the water may be to increase the concentration of either hydrazinium hydroxide or the hydrazinium ion according to the following equation:

$$N_2H_4 + H_2O \rightleftharpoons N_2H_5OH \rightleftharpoons N_2H_5^+ + OH^- \tag{2}$$
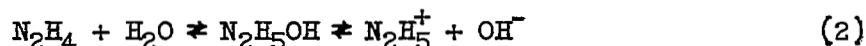
A neutralization reaction with HNO_3 might proceed more rapidly with N_2H_5OH or OH^- than with N_2H_4, in which case small additions of water would increase the rate of the over-all neutralization. Another possibility is that water addition reduces the vigor of the initial reactions sufficiently to permit much better liquid-phase mixing than is otherwise possible, and that the net effect for this particular case is increased over-all heat liberation in the liquid phase and better atomization.

CHEMICAL AND PHYSICAL FACTORS AFFECTING COMBUSTION

Liquid-Phase Reactivity

The concept that heat generation within mixed droplets of fuel and acid directly affects performance is used in reference 2 to explain the effect on c^* of water added to the propellant combination of furfuryl alcohol - aniline and RFNA. Heat generated within mixed droplets by liquid-phase chemical reactions supplements heat transferred from the hot gas atmosphere. If the rate of liquid-phase heat generation is significant compared with the rate of heat transfer from the gas phase to the droplets, the evaporation rate of the mixed propellant droplets is accelerated, resulting in increased c^* in short combustors, as compared with that of chemically inert fuels like saturated hydrocarbons.

Of the self-igniting fuels studied in reference 1, it appears that, relative to JP-4, only hydrazine, trimethyl trithiophosphite, furfuryl alcohol, and UDMH react with RFNA with sufficient rapidity and heat liberation to satisfy the previous concept. JP-4 fuel with 40 percent of the highly reactive UDMH also gives a large c^* increase over neat JP-4. A further indication of the rapidity of liquid-phase reactions among the fuels hydrazine, UDMH, and trimethyl trithiophosphite with RFNA is the high pressure drop developed in a rapid mixing injector (ref. 1).

The c^* performance of the self-igniting fuels, allylamine and o-toluidine, was similar to that for JP-4. This indicates that the rate of heat generation in the liquid phase is not significant compared with the rate of heat transfer from the gas phase. Therefore, the fact that the fuels are self-igniting does not automatically imply that liquid-phase reactions are fast enough to be effective in improving combustion rates within an engine.

Added Water

Water added initially to either the fuel or the acid has a very specific and possibly a very large effect if the fuel-acid combination has rapid exothermic reactions in the liquid phase. Thus, water added to the hydrazine - RFNA mixture increases c^*, whereas water added to furfuryl alcohol - aniline and RFNA produces the opposite effect. Studies of the ignition delay of various fuels with nitric acid oxidizers have shown that added water can either increase or decrease the ignition delay. Triethylamine (ref. 4) and triethylamine mixtures with o-toluidine, allylamine, and ethylenimine (ref. 7) show increased ignition delay with added water using WFNA at -40° and 70° F. When NO_2 is added to the acid, the effect is less pronounced and for RFNA containing 24 percent NO_2, at 70° F, the effect is reversed for the allylamine and ethylenimine mixtures with triethylamine.

Whether water added to fuel-acid mixtures will increase or decrease rates of liquid-phase reactions depends on the mechanism of the reactions. Detailed discussions of the chemistry of ionic reactions in reactive fuel-acid mixtures may be found in references 4, 6, and 7. The water effects may be expected to apply to steady-state combustion in engines only when the liquid-phase reactions are fast enough to liberate heat at a rate that is significant compared with the heat-transfer rate from the combustion gas. It was pointed out in a previous section that self-ignition may not be a sufficient criterion.

Water added to fuel-acid mixtures that have little or no liquid-phase reactivity may be expected to produce a relatively small c^* decrease. The decrease may be considerably greater than that in the theoretical c^* as shown by the data for toluene and JP-4 fuel. The reasons for the decreased c^* performance are not well understood. It might be caused by changes in physical properties such as total enthalpy of evaporation.

Fuel Volatility

For the case in which liquid-phase heat generation is negligible compared with the convective heat-transfer rate (e.g., paraffins, JP-4 fuel, toluidine), fuel volatility becomes an important factor in determining combustion rate. In reference 3 it is shown that the general shape of the c^* mixture ratio curves for this case can be predicted from the relative volatilities of the propellants. It was only necessary to assume that the injector produced drops of mixed propellant and that the fraction of propellant vaporized depended only on L^*.

Injector Configuration

The concept that vaporization is the principal rate-determing step in rocket combustion combined with the concept of heat generation within mixed droplets leads to important conclusions concerning injection methods. In order to utilize heat generation as an aid to vaporization, an injector must be used that produces efficient liquid-phase mixing. This effect was demonstrated in reference 2 in which a mixing-type injector gave improved performance over a nonmixing-type injector with furfuryl alcohol - aniline fuel. Both injectors presumably produced similar degrees of atomization.

The vaporization model of combustion used to interpret the hydrocarbon performance data of reference 3, however, implied that a nonmixing injector would give better performance for very volatile or very nonvolatile hydrocarbon fuels. The principle should apply for any fuel that has negligible liquid-phase reactivity with nitric acid oxidizers.

Slow Reactions in Gas Phase

There is considerable evidence that thermal decomposition of nitric oxide, NO, is a rate-limiting step in combustion processes involving gaseous fuels and vaporized nitric acid or nitrogen oxides (refs. 8 and 9). In reference 1, the decrease in combustion rate of self-igniting fuels with an increase in o/f was explained in terms of a slow decomposition of NO. It now appears that the decrease could also be explained by a corresponding decrease in the rate of heat liberation in mixed droplets.

Recent kinetic studies of NO decomposition are discussed in reference 10 where the half-life of NO was calculated as a function of temperature. The result is shown in figure 7. From the experimental data of references 1 and 3, it is possible to estimate combustion temperature and the half-life of NO. When it is assumed that the gas specific heat ratio and molecular weight are 1.24 and 25, respectively, and that the flow is one-dimensional, the expression for combustion temperature T_c becomes

$$T_c = 2.195 \times 10^{-4} (c^*)^2 \quad (^{\circ}R)$$

$$= 1.22 \times 10^{-4} (c^*)^2 \quad (^{\circ}K)$$

and the mean stay time t_s in the combustor is given by

$$t_s = 0.191 \frac{L^*}{c^*} \quad (sec)$$

The following table shows estimated combustion temperatures, NO half-lives, and half-life to stay-time ratios for several fuel-RFNA systems at a mixture parameter β of 0.8:

Fuel	Characteristic chamber length, L^*, in.	Characteristic exhaust velocity, c^*, ft/sec	Combustion temperature, T_c, °K	Nitric oxide half-life, $t_{1/2}$, sec	Stay time, t_s, sec	Ratio of half-life to stay time, $\dfrac{t_{1/2}}{t_s}$
Hydrazine	30	4970	3010	0.00076	0.00115	0.14
	100	4770	2780	.0007	.004	.18
UDMH	30	4760	2760	.0009	.0012	.75
	100	4960	3000	.0008	.0039	.05
Allylamine	30	4050	2000	.8	.0014	200
	100	4860	2880	.00035	.0039	.09
o-Toluidine	30	3860	1820	1.0	.0015	667
	100	4700	2700	.0015	.0041	.37
Toluene	30	3620	1600	6.0	.0016	>1000
	240	4730	2730	.001	.0097	.1
n-Heptane	30	3460	1460	>10.0	.0017	>1000
	240	4790	2800	.0006	.0096	.06
JP-4	30	3430	1435	>10.0	.0017	>1000
	240	4740	2740	.001	.0097	.1

It is apparent from these calculations that in the short combustors the NO formed is essentially undecomposed, except for the hydrazines, unless there is considerable reaction with fuel or partially oxidized species. In the long combustors NO should be completely consumed. Although these results have no particular bearing on the validity of the vaporization model of combustion, they do indicate that the extent of NO decomposition, rather than the percent of liquid evaporated, may determine the actual c^* obtained in very short combustors.

CONCLUDING REMARKS

The vaporization model of combustion has been used to interpret a variety of steady-state performance measurements. A rigorous proof of the model, however, will depend upon future experimentation. Experimental comparisons between fuels of similar volatility but different liquid-phase reactivity would add to the state of understanding of the phenomena. Thorough studies of the effect of injection configuration with carefully chosen fuel-acid systems are also required.

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SUMMARY OF RESULTS

The hypothesis that droplet vaporization is the principal rate-limiting step in combustion has been applied to fuel - nitric acid rocket systems. Experimental results from this study and from three others have been found to be consistent with the vaporization model. The following trends were indicated:

1. The fuels hydrazine, trimethyl trithiophosphite, furfuryl alcohol, unsymmetrical dimethylhydrazine, furfuryl alcohol - aniline mixtures, and 40 percent unsymmetrical dimethylhydrazine in JP-4 appear to be sufficiently reactive toward red fuming nitric acid in the liquid phase to promote vaporization in mixed propellant droplets markedly.

2. Ordinary hydrocarbon-type fuels and the self-igniting fuels, allylamine and o-toluidine, are not sufficiently reactive toward red fuming nitric acid to promote vaporization in mixed propellant droplets to an extent that affects performance.

3. Water added to fuel-acid mixtures that react vigorously in the liquid phase has a very specific effect on the characteristic exhaust velocity which may depend on the chemistry of the reactions. For example, water added to the anhydrous hydrazine - red fuming nitric acid propellant combination improved the characteristic exhaust velocity in short chambers by 12 percent; whereas, other investigators found that water decreased the characteristic exhaust velocity of the furfuryl alcohol - aniline and red fuming nitric acid systems.

4. Water added to fuel-acid mixtures that are unreactive in the liquid phase decreases the characteristic exhaust velocity c^* in short combustors by a relatively small amount, which is greater however, than the corresponding decrease in theoretical performance.

Lewis Flight Propulsion Laboratory
     National Advisory Committee for Aeronautics
          Cleveland, Ohio, April 18, 1958

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TABLE I. - PHYSICAL CHARACTERISTICS

OF JP-4 FUEL

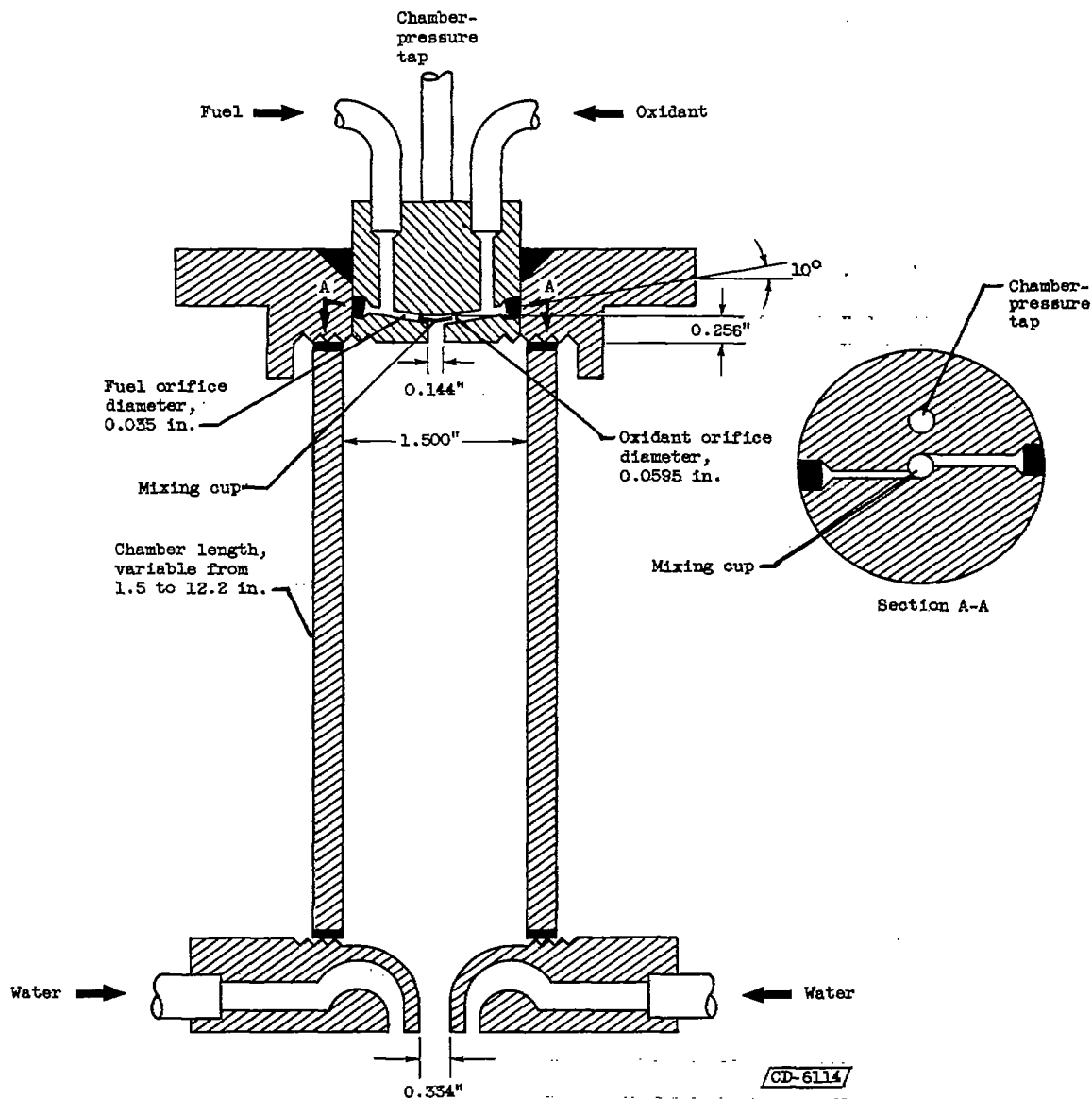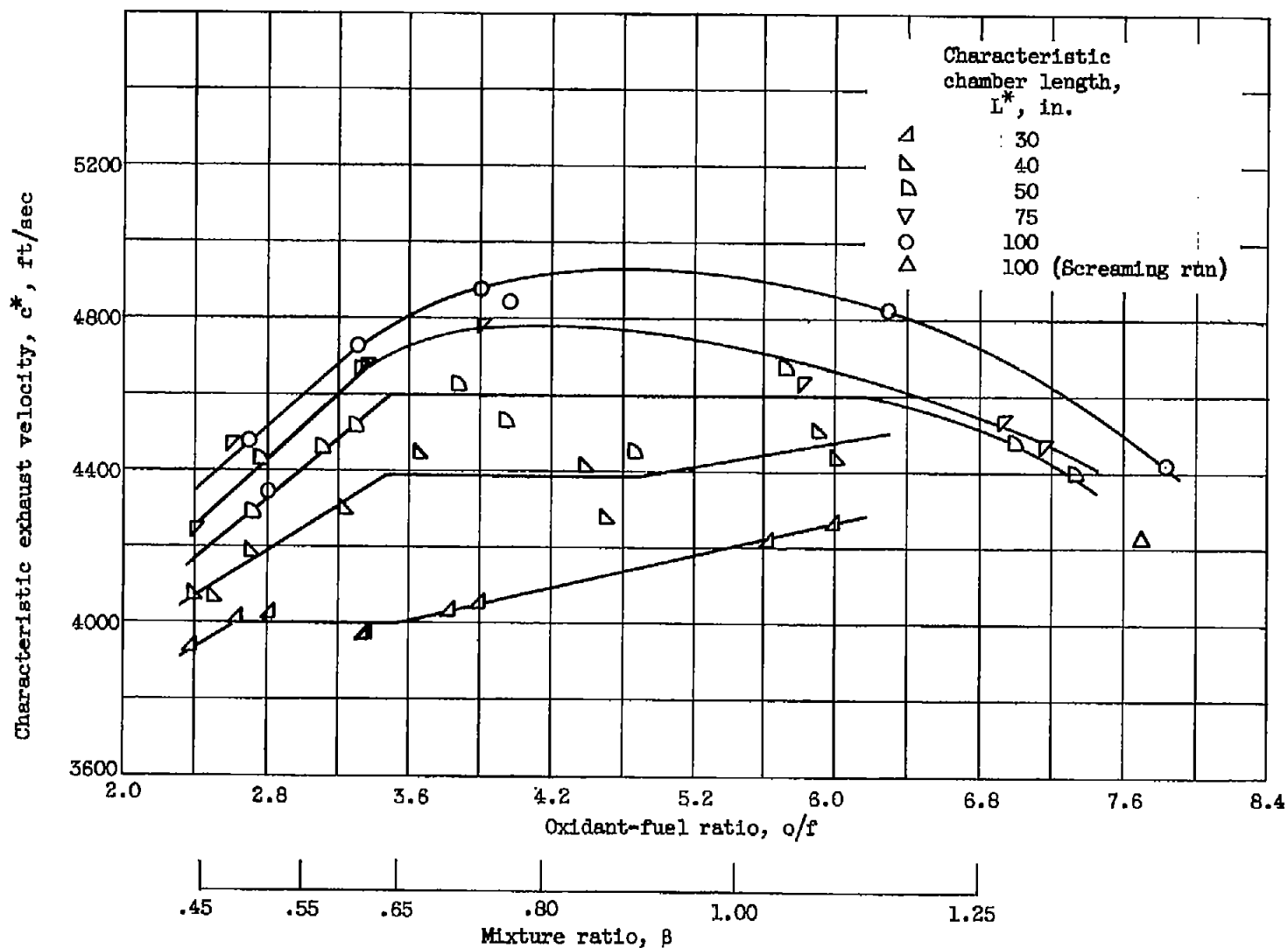| Boiling point, °F Percent evaporated	
0	157
5	224
10	246
20	263
30	273
40	282
50	291
60	301
70	312
80	324
90	345
95	364
100	418
Residue, percent	1.1
Loss, percent	.5
Reid vapor pressure, lb/sq in.	2.6
Specific gravity, 60/60° F	.763
Hydrogen-carbon ratio, by weight	.171
Net heat, Btu/lb	18,700
Aniline point, °F	134.9

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Chamber-
pressure
tap

Fuel ➡ ⬅ Oxidant

10°

0.256"

A   A

Chamber-
pressure
tap

Fuel orifice
diameter,
0.035 in.

0.144"

Oxidant orifice
diameter,
0.0595 in.

1.500"

Mixing cup

Mixing cup

Section A-A

Chamber length,
variable from
1.5 to 12.2 in.

Water ➡ ⬅ Water

0.334"

CD-6114

Figure 1. - Schematic drawing of 40-pound-thrust rocket engine.

Characteristic
chamber length,
L^*, in.

◁   30
◺   40
◹   50
▽   75
○   100
△   100 (Screaming run)

5200

4800

4400

4000

3600

Characteristic exhaust velocity, c^*, ft/sec

2.0   2.8   3.6   4.2   5.2   6.0   6.8   7.6   8.4

Oxidant-fuel ratio, o/f

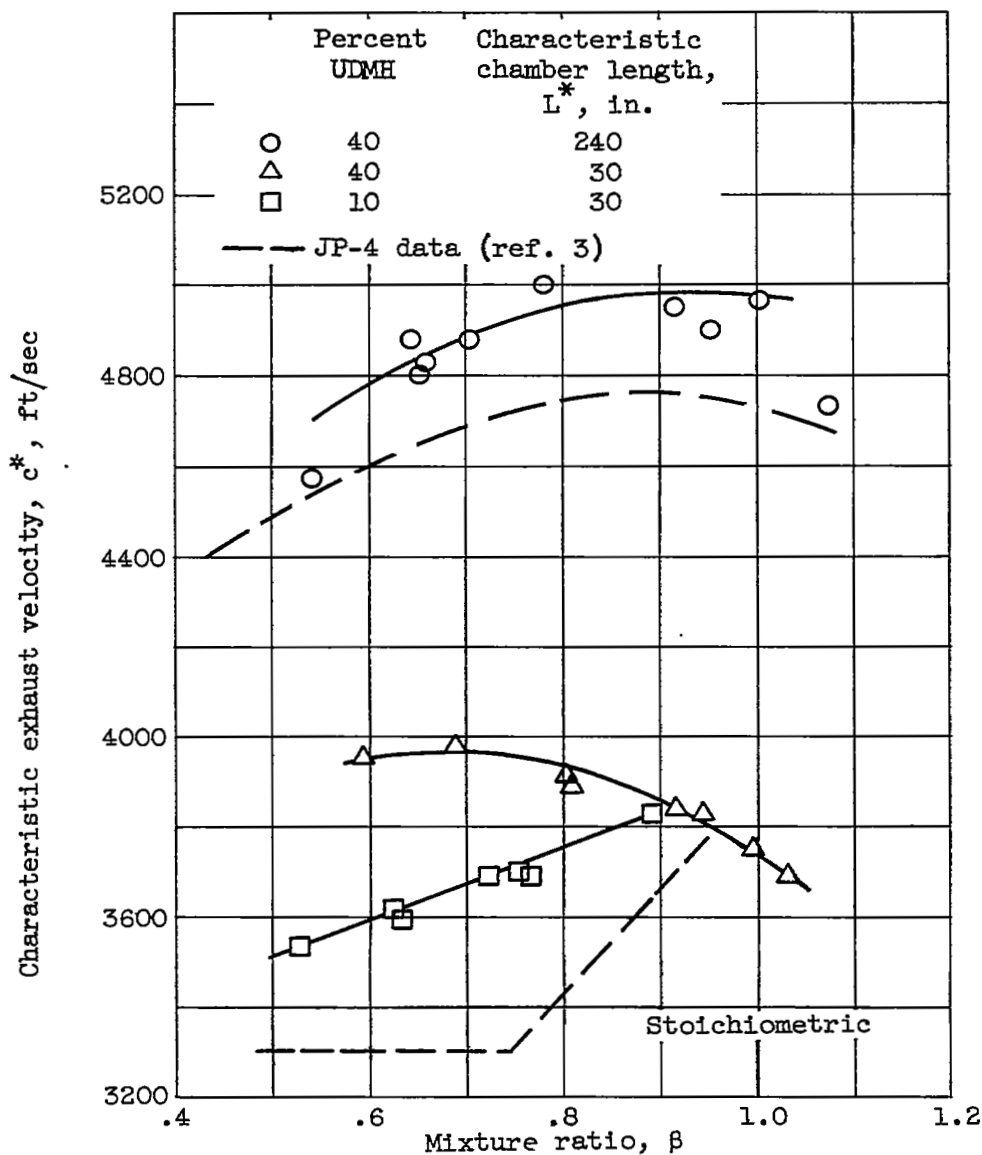.45   .55   .65   .80   1.00   1.25

Mixture ratio, β

Figure 2. - Characteristic exhaust velocity c^* as function of mixture ratio β and characteristic chamber length L^* for red fuming nitric acid and JP-4 fuel mixture in 1-inch-diameter combustor.

Figure 3. - Characteristic exhaust velocity as
function of characteristic chamber length for
red fuming nitric acid and JP-4 fuel at a mix-
ture ratio of 0.8 showing the chamber length
L_{97}^{*} required to reach an exhaust velocity of
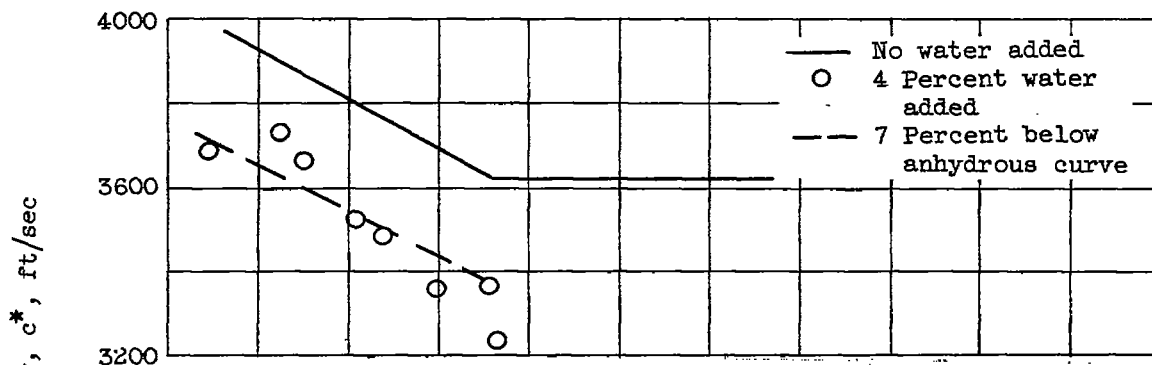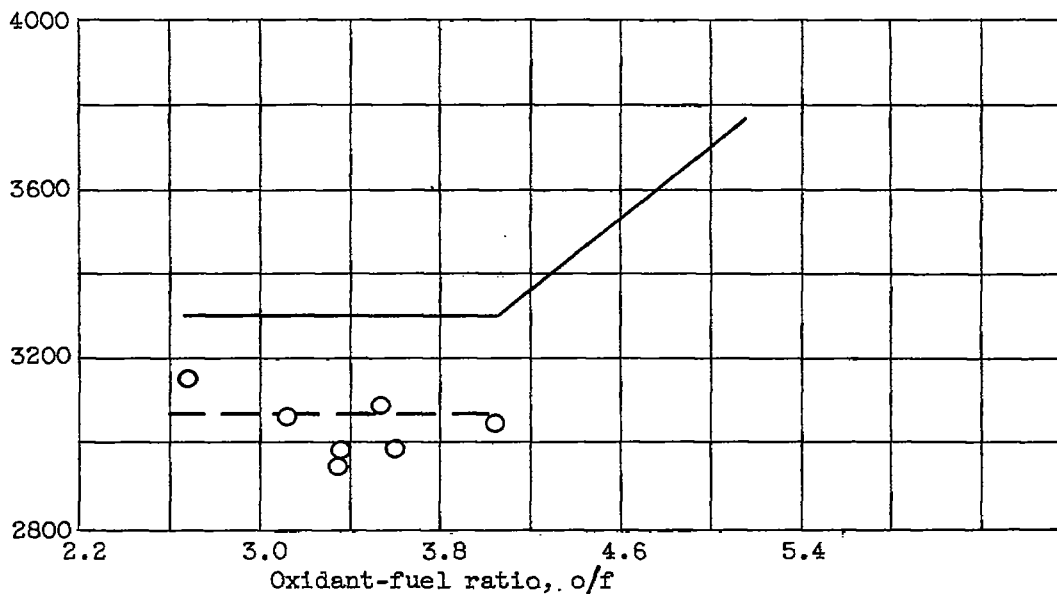97 percent of maximum experimental exhaust
velocity.

Figure 4. - Characteristic exhaust velocity as function of mixture ratio and characteristic chamber length for red fuming nitric acid with mixtures of JP-4 fuel and unsymmetrical dimethyl hydrazine.

(a) Toluene.

(b) JP-4 fuel.

Figure 5. - Effect of 4 percent water added to red fuming nitric acid on characteristic exhaust velocity for toluene and JP-4 fuel in $1\frac{1}{2}$-inch-diameter combustor with a chamber length of 30 inches.
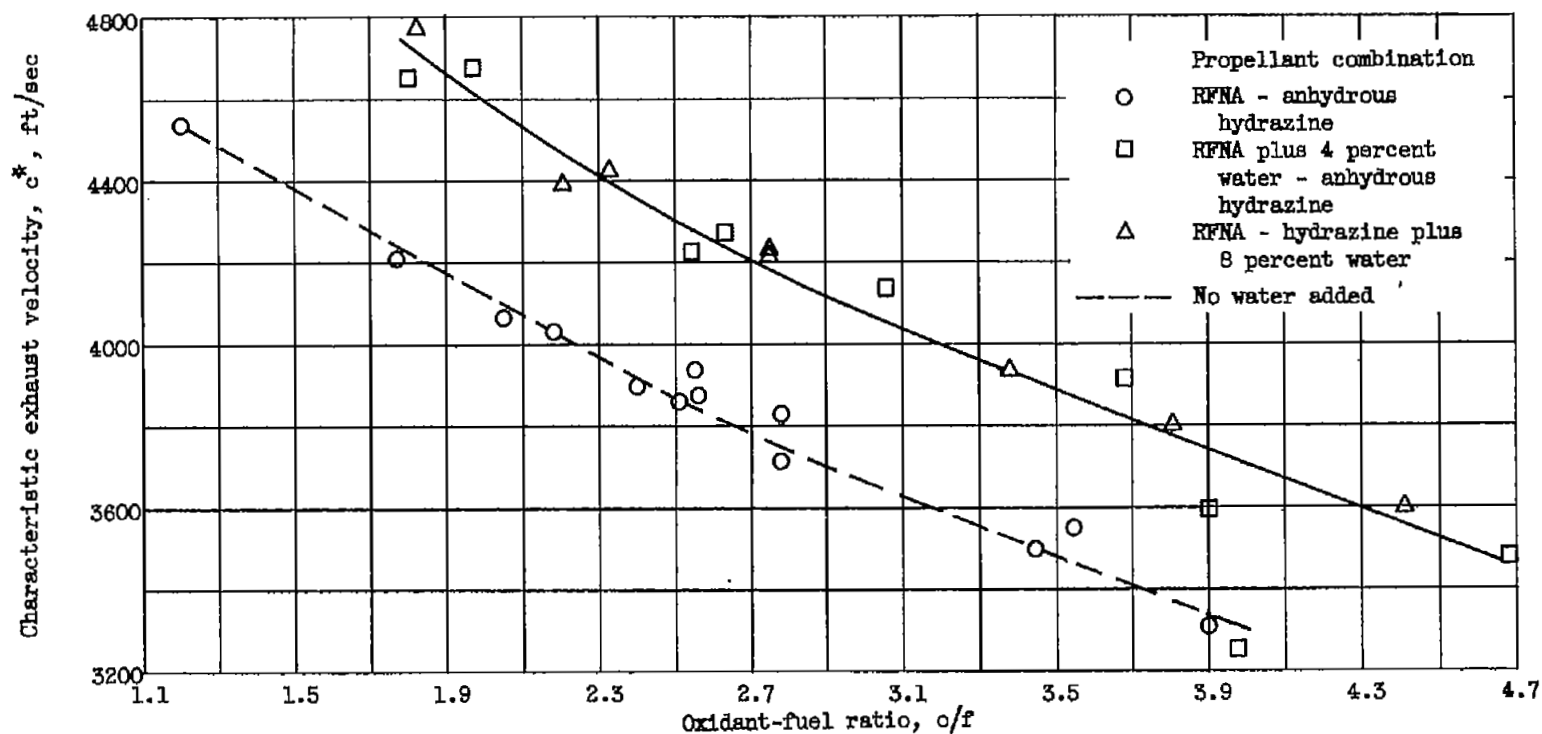
Figure 6. - Effect of added water on characteristic exhaust velocity of red fuming nitric acid and anhydrous hydrazine propellant combination in $1\frac{1}{2}$-inch-diameter combustor with a chamber length of 30 inches.
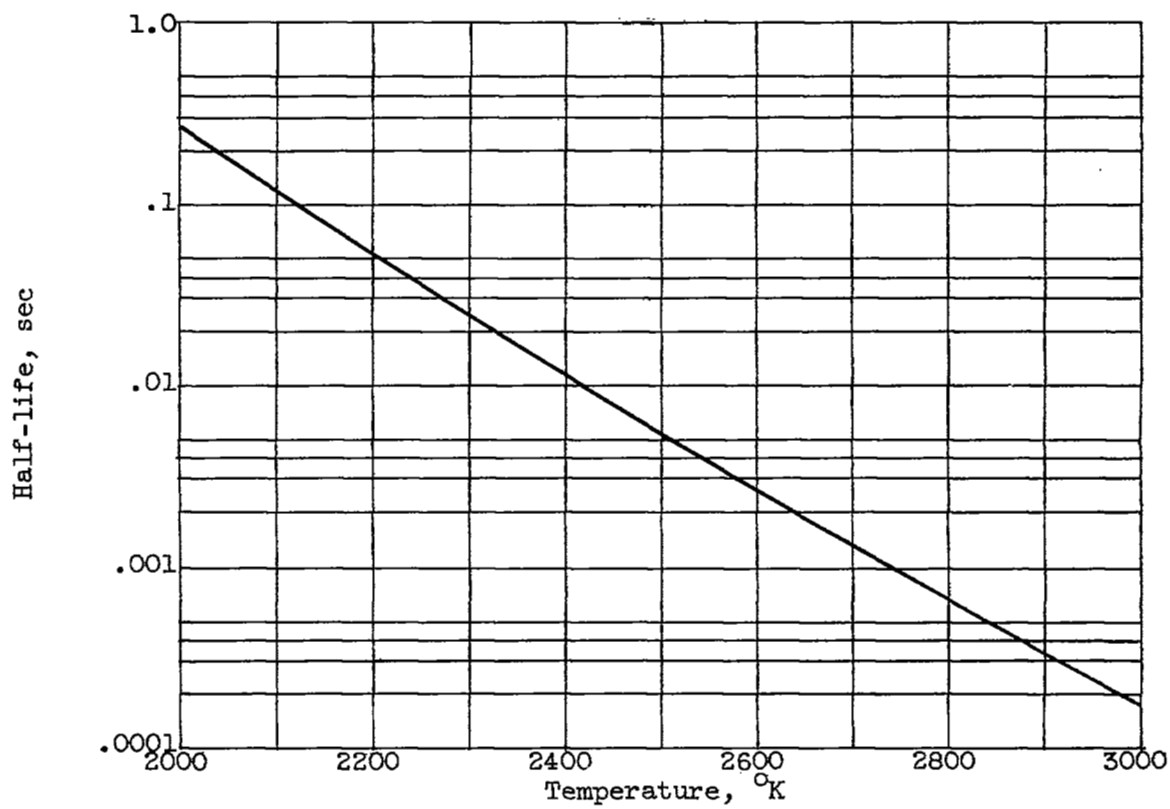
Figure 7. - Half-life as a function of temperature for iso-
thermal decomposition of nitric oxide (ref. 10). Pressure,
1 atmosphere.