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RESEARCH MEMORANDUM

SOLUBILITY OF WATER IN HYDROCARBONS

By R. R. Hibbard and R. L. Schalla

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Cleveland, Ohio

FOR REFERENCE

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**NATIONAL ADVISORY COMMITTEE
FOR AERONAUTICS**

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SUMMARY

A study of the literature data on the solubility of water in hydrocarbons has shown that (a) the log of the solubility is inversely proportional to the reciprocal of the absolute temperature, (b) there is an apparent critical solution temperature at about 352° C indicated both by extrapolation of the literature data and by Henry's law, and (c) the solubility increases with decreasing hydrogen-to-carbon ratio H/C. The following equation is proposed for the prediction of the solubility of water at any temperature in nonolefinic hydrocarbons and petroleum fractions:

$$\log x = -(4200 H/C + 1050)(1/T - 0.0016) + 2.00$$

where

x solubility of water in hydrocarbon, mol percent

H/C hydrogen-to-carbon weight ratio for hydrocarbon

T absolute temperature, °K

This equation has been applied to a few petroleum fractions ranging from gasoline to lubricating oil, and a comparison of the calculated and experimentally determined solubilities shows an agreement which is believed to be adequate for most engineering purposes.

INTRODUCTION

The solubility of water in hydrocarbon fuels is of interest in that most fuels are substantially saturated with water at some stage during their processing and storage. This water can be troublesome in applications where a vehicle or aircraft is exposed to temperatures below the refueling temperature and where the water will separate out at the reduced temperatures. This is especially true in aircraft operation where the separated water is likely to freeze and block filters.

A literature search was therefore conducted at the NACA Lewis laboratory for data on the solubility of water in pure hydrocarbons and in hydrocarbon fuels, and an attempt was made to correlate these data in such a way that a reasonably accurate prediction could be made of the solubility of water in any hydrocarbon fuel at any temperature. Presented herein is an analysis of the literature data and an equation which appears useful for the estimation of the solubility of water in hydrocarbon fuels of low olefin content.

ANALYSIS OF PURE HYDROCARBON DATA

Listed in table I are the 22 pure hydrocarbons, the temperature ranges, and the references from which solubility data were compiled together with the number of determinations reported in each reference. In the search some data may have been overlooked but certainly a major portion of that available in the literature was covered; a total of 227 data points is presented. In cases where the solubility of water in a single hydrocarbon was determined at the same temperature by several investigators, considerable disagreement in results was often noted. For example, values for the solubility of water in benzene at 20° C have been reported ranging from 0.185 mole percent (reference 12) to 0.264 mole percent (reference 8). In spite of such inconsistencies as these, an attempt was made to develop an equation for predicting the solubility of water in hydrocarbons which would satisfy the experimental observations within reasonable limits.

In a study of the solubility of water in a naphtha, a kerosene, and a lubricating oil at elevated temperatures, Griswold and Kasch (reference 20) presented a plot of the Henry's law constant for the naphtha as a function of temperature. The equation used was

$$K_w = \frac{P_w}{x_w} \quad (1)$$

where

K_w Henry's law constant

P_w pressure of water vapor over two phase mixtures

x_w water in naphtha, mol fraction

Since the figure presented by Griswold and Kasch was for naphtha alone, Henry's law constants were calculated for all three petroleum fractions by use of the data of reference 20, and the values for these constants are plotted against the temperature in figure 1. Although the scatter

of points is fairly great, there appears to be an increase in the values of these constants with increasing temperature, probably because the solubility of water in the hydrocarbons becomes quite appreciable (of the order of 40 percent) at higher temperatures and the partial pressure of water is no longer approximately equal to the vapor pressure of pure water. The line faired through the Henry's constants is substantially the same as that drawn for naphtha in reference 20 but is extrapolated to higher temperatures. The vapor-pressure curve for pure water, also shown in figure 1, intercepts the line through the Henry's law constants at a pressure of about 2450 pounds per square inch absolute and a temperature of 352° C. If equation (1) is assumed to hold at high temperatures, then this temperature of intercept (352° C) is also the critical solution temperature of water-hydrocarbon systems since $K_w = p_w$ and x_w must be 1.0 at this temperature. The fairing of the curve through the Henry's law constant points is arbitrary; other curves could be drawn which would yield intercepts ranging from 2100 to 2700 pounds per square inch absolute. However, the water-vapor-pressure curve is sufficiently steep in this region that the critical solution temperature would only range between 340° and 360° C.

The data from the references (table I) were tabulated and the solubility of water, in terms of log mole percent, was plotted against the reciprocal of the absolute temperature. It was found that straight lines could be drawn through the data obtained from any single investigation and that the scatter from the line usually was within the expected limits of experimental measurement. This linear relation between the log of the solubility of water and the reciprocal of the absolute temperature agrees with the results presented in reference 4 where the heat of solution of water in benzene was determined by use of the equation

$$\Delta H = RT^2 \left(\frac{d \ln N}{dT} \right) \quad (2)$$

where

ΔH molar heat of solution

R gas constant

T temperature, °K

N solute in solvent at saturation, mol fraction

and which by rearranging and integrating becomes

$$\ln N = - \frac{\Delta H}{RT} + C \quad (3)$$

or

$$\log N = \frac{k}{T} + C \quad (4)$$

where k and C are constants for a given hydrocarbon. It was also found that most of the data justified a straight line passing through 100 mole percent solubility of water at temperatures close to the 352° C that was estimated from figure 1 as being the critical solution temperature for water-hydrocarbon systems. A few illustrative examples of the solubility data referred to in table I are plotted as log concentration against the reciprocal of absolute temperature in figure 2. Figures 2(a) and 2(b), which are for toluene and xylene, respectively, show that a considerable number of data points fall approximately on a straight line passing through 352° C where $1/T$ °K = 0.0016. The data for benzene showed about the same degree of scatter as did those for many of the other hydrocarbons, although in most cases the range of temperatures covered was small. Figure 2(c) is a similar plot for cyclohexane. Figure 2(d) for hexadiene 1,5 and diisobutene is typical of the plots obtained where only a very limited amount of data was available. Methylcyclohexane and propane were the only compounds which gave log solubility against $1/T$ plots in which the best straight line did not clearly indicate a critical solution temperature of near 350° C. The methylcyclohexane data are plotted in figure 2(e), which shows the correlation line that was forced for this hydrocarbon. The propane data are plotted in figure 2(f). It is not known why these two compounds do not follow the trends shown by other hydrocarbons; the data for propane were not used in subsequent correlations.

The equation for straight lines passing through the 352° C critical solution temperature on log concentration against $1/T$ scales is

$$\log x = M (1/T - 0.0016) + 2.00 \quad (5)$$

where

x solubility of water in hydrocarbon, mol percent

M slope of line, constant for each hydrocarbon

T temperature, °K

For most of the data, equation (5) can be used to estimate the solubility of water in a hydrocarbon at any temperature once M has been evaluated from a known solubility at a single temperature.

Since it is also desirable to be able to estimate the solubility of water in hydrocarbons in the absence of any such data, a relation was sought between M and some other hydrocarbon property. The

slope M is always negative and in general it was observed that M had a lower value (solubility of water is higher) for olefins and aromatics than for paraffins and cycloparaffins and, with the exception of the olefins, there appears to be a trend of increasing value for M with increasing H/C ratio. In table II are listed the H/C ratios, the values for M , and the number of data points obtained from the literature for each hydrocarbon. These data are plotted in figure 3; the straight line was faired through the aromatic, cycloparaffin, and paraffin points; the number of determinations represented by each point was taken into account but the olefin and diolefin data were neglected. The points which lie farthest from the line are olefin and diolefin points representing relatively few determinations. This correlation is admittedly poor but should serve to permit a rough estimate of M and of the solubility of water in hydrocarbon fuels, especially if their olefin content is low.

The equation for the line drawn in figure 3 relating M to the H/C ratio is

$$M = -(4200 H/C + 1050) \quad (6)$$

and the equation relating the solubility of water in nonolefinic hydrocarbons at any temperature then becomes

$$\log x = -(4200 H/C + 1050) (1/T - 0.0016) + 2.00 \quad (7)$$

where

x solubility of water, mol percent

H/C hydrogen-to-carbon weight ratio of hydrocarbon

T absolute temperature, $^{\circ}K$

RESULTS ON PETROLEUM FRACTIONS

The literature revealed a considerable amount of data on the solubility of water in petroleum fractions. However, only four sources were found where measurements were made over a range of temperatures, and these data are shown in figure 4 on log concentration against $1/T$ scales. Concentrations are shown in the terms used in the literature sources (either weight or molar percent). The data presented in reference 1 for a kerosene and a paraffin oil show a good linear relation between log concentration and reciprocal temperature (fig. 4(a)). A similar plot of Griswold and Kasch's data (reference 20) is presented in figure 4(b) for naphtha, kerosene, and lubricating oil and similarly the data show excellent agreement with a straight line. Figure 4(c) is for four of the five

gasolines investigated in reference 21 with the fifth fuel omitted for the sake of clarity. Although curved lines have been drawn for three of these fuels, the scatter of data is sufficient in the cases where multiple determinations were made to suggest that the deviation from linearity might be due to experimental inaccuracies. Figure 4(d) presents data from reference 15 for a safety fuel (probably a high-boiling alkylate) and again the deviation from a straight line is probably within the limits of experimental measurement.

Most of the literature data on the solubility of water in petroleum fractions is in weight percent terms; before these data can be compared with the values predicted by use of equation (7), both the molecular weight and the H/C ratio of the petroleum fractions must be known. Both of these properties can be estimated from distillation and gravity by use of the correlations presented in references 22 and 23. For most of the literature on the solubility of water in petroleum fractions, however, insufficient distillation data are presented to allow these correlations to be used. Only the petroleum fractions used in reference 21 and by Griswold and Kasch in reference 20 are sufficiently well described to allow a comparison to be made between the experimentally determined solubilities and those calculated by use of equation (7). This comparison is given in table III, along with the average boiling points, gravities, and estimated molecular weights and H/C ratios for the fractions.

Listed at the bottom of table III are the average differences between calculated and experimental solubilities for various portions of the data given in the table. The average difference for all the data is 56 percent; for the data presented in reference 20, 19 percent; and for the data presented in reference 21, 136 percent. The largest differences are found with the fuels reported in reference 21 at the higher temperatures and the experimental values show a much smaller increase in solubility with increasing temperature than that predicted by equation (7); there is a possibility that these data may be in error as suggested by the fact that the solubility of water in fuels 10 and 19 is quite different even though the fuels have quite similar physical properties. If the five values determined at 50° C in reference 21 are omitted from the 47 values considered, the average difference is then only 30 percent.

DISCUSSION

The critical solution temperature of 352° C proposed herein is above the critical temperature of many of the hydrocarbons studied and has no real physical significance. It is also very close to the 374° C critical temperature of water and the value 374° C ($1/T \text{ } ^\circ\text{K} = 0.00155$)

could have been used in equation (4) without any appreciable change in the predicted solubilities of water in hydrocarbons. However, the assumed miscibility at either of these temperatures is not believed to be of any fundamental significance and the proposed value of 352° C has been used only as an empirical focal point to aid in the fairing of straight lines through a considerable amount of somewhat scattered data.

The validity of using a straight-line relation between log concentration and $1/T$ through 0° C may be in doubt. The Henry's law approach suggests that a straight line should be used only over the temperature range where there is a linear relation between the log of the vapor pressure of water and the reciprocal of the absolute temperature. In the steam-water-ice system this relation does hold down to 0° C, but there is a change of slope at the freezing point. However, in the very dilute solutions present at this and lower temperatures, the great dilution of water molecules in the hydrocarbon environment may yield a condition where the water vapor pressure against temperature curve follows that of supercooled water to temperatures well below the normal freezing point of water. Under this condition the log concentration would be linear with $1/T$ across the 0° C normal freezing point. As indicated, for example, by the water in toluene data plotted in figure 2(a), the determination of the solubility of water at low temperatures is not sufficiently precise to support this point. However, it is of minor practical importance since the concentrations of water in hydrocarbons are extremely low below 0° C in any case.

The comparison between calculated and experimentally determined solubilities which is presented in table III shows the calculated solubilities to be generally higher than those determined by experiment. The constants in equation (7) were derived from pure hydrocarbon data and a better agreement would have been obtained for the predicted solubilities for the petroleum fractions if a somewhat larger value had been used for the first constant of this equation. However, it was felt that there were insufficient data available on the solubility of water in well defined petroleum fractions to justify a change in this constant, which was based on a considerably larger amount of pure compound data.

CONCLUSIONS

A study of the data found in the literature on the solubility of water in hydrocarbons has shown that:

1. For a given hydrocarbon, there is a linear relation between the log of concentration and the reciprocal of the absolute temperature, and the solubility increases with increasing temperature.

2. For most hydrocarbons, this linear relation predicts a critical solution temperature of about 352° C.

3. In general the solubility of water in hydrocarbons increases with decreasing hydrogen-to-carbon weight ratio of the hydrocarbon.

4. The solubility of water in any nonolefinic hydrocarbon at any temperature can be predicted with fair accuracy by use of the equation

$$\log x = -(4200 H/C + 1050)(1/T - 0.0016) + 2.00$$

where

x solubility of water in hydrocarbon, mol percent

H/C hydrogen-to-carbon weight ratio in hydrocarbon

T absolute temperature, °K

5. The foregoing equation may be used to predict the solubility of water in those petroleum fractions for which there are sufficient data in the literature to permit estimates to be made of the H/C ratio. For these fractions the average difference between predicted and experimentally determined solubilities was 56 percent of the amount found by experiment. However, the largest differences were all found in the comparison of the results of one investigator, at one temperature. If these cases are omitted (5 out of 47 cases), the average difference between calculated and experimentally determined solubilities becomes only 30 percent of the amount found by experiment. Accuracies of this order are believed to be adequate for most engineering purposes and are, in any case, comparable with the accuracies which have often been encountered experimentally.

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TABLE I - REFERENCES ON SOLUBILITY OF WATER IN PURE HYDROCARBONS

Fuel	Reference	Determinations reported	Temperature range (°C)
Benzene	1	7	3 - 72
	2	9	5 - 73
	3	6	25 - 71
	4	14	23 - 73
	5	5	20 - 55
	6	6	10 - 60
	7	1	20
	8	7	5 - 70
	9	1	20
	10	5	10 - 50
	11	10	0 - 80
	12	3	10 - 26
	13	5	4 - 40
Styrene	14	5	6 - 51
Toluene	2	12	- 9 - 93
	6	6	0 - 50
	11	6	- 10 - 40
	15	4	- 34 - 43
Xylene	2	1	20
	7	2	10 - 25
	15	5	- 34 - 43
Cyclohexane	2	6	14 - 53
	10	2	20 - 50
	12	1	20
Methylcyclohexane	15	5	- 34 - 43
Propane	16	8	27 - 91
	17	1	38
<u>n</u> -Butane	12	8	5 - 21
	18	8	38 - 144
Isobutane	12	4	7 - 21
Butene-1	12	5	6 - 22
Butene-2	12	5	7 - 21
Isobutene	12	7	6 - 21
Butadiene-1,3	12	6	7 - 21
<u>n</u> -Pentane	12	5	6 - 25
Isopentane	12	10	6 - 22
<u>n</u> -Hexane	12	1	20
	19	1	11
Hexadiene-1, 5	12	2	14 - 20
<u>n</u> -Heptane	12	4	10 - 25
	19	1	11
Heptene-1	12	4	10 - 21
<u>n</u> -Octane	12	1	20
	19	1	11
2,2,4-Trimethylpentane	10	4	- 2 - 40
	15	5	- 34 - 43
Diisobutene	15	2	- 34,43

TABLE II - H/C RATIO AND SLOPE OF SOLUBILITY LINE
FOR PURE HYDROCARBONS

Compound	H/C ratio	Number		Slope M
		Investi- gators	Data points	
Benzene	0.083	12	65	-1420
Styrene	.083	1	5	-1370
Toluene	.092	4	28	-1450
Xylene	.104	3	8	-1450
Cyclohexane	.168	3	9	-1850
Methyl cyclohexane	.168	1	5	-1710
n-Butane	.209	2	16	-1980
Isobutane	.209	1	4	
Butene-1	.168	{ 1 }	5	-1580
Butene-2	.168		5	
Isobutene	.168		7	
Butadiene-1,3	.126	1	6	-1460
n-Pentane	.201	{ 1 }	5	-1880
Isopentane	.201		10	
n-Hexane	.197	2	2	-1680
n-Hexadiene-1,5	.140	1	2	-1310
n-Heptane	.192	2	5	-1720
n-Heptene-1	.168	1	4	-1220
n-Octane	.188	2	2	-1910
2,2,4-Trimethyl pentane	.188	2	9	-1820
Diisobutene	.168	2	2	-1770

TABLE III - COMPARISON OF CALCULATED AND EXPERIMENTALLY DETERMINED
SOLUBILITIES OF WATER IN PETROLEUM FRACTIONS

Petroleum	Reference	Molal average boiling point (°F)	A.P.I. gravity	Estimated		Solubility of water			
				H/C ratio	Molecular weight	T (°C)	Experimental (mol percent)	Calculated (mol percent)	Difference percent of experimental value
Gasoline 10	21	138	77.5	0.185	82	10	0.058	0.029	-50
						30	.073	.078	7
						50	.088	.185	110
Gasoline 12	21	177	72.1	0.184	95	10	0.027	0.030	11
						30	.035	.080	128
						50	.039	.188	382
Gasoline 13	21	187	61.6	0.171	94	10	0.037 ^a	0.038	3
						30	.036	.098	172
						50	.058 ^a	.226	290
Gasoline 15	21	201	65.5	0.176	99	10	0.030 ^a	0.035	17
						30	.095 ^a	.091	-4
						50	.114 ^a	.211	85
Gasoline 19	21	145	76.0	0.184	85	10	0.017	0.030	76
						30	.028 ^a	.080	184
						50	.030	.188	527
Naphtha	20	342	54.3	0.170	147	159	4.97	5.48	10
						186	8.96	9.53	7
						203	11.91	13.1	10
						222	16.18	18.1	12
Kerosene	20	459	42.0	0.158	182	112	1.24	1.95	57
						135	2.18	3.48	60
						169	4.98	7.33	47
						177	5.89	8.59	46
						185	7.39	10.0	35
						191	7.98	11.2	40
						203	9.00	13.9	54
						207	12.14	14.9	23
						216	14.94	17.3	16
						228	19.06	21.0	10
						251	23.04	29.6	29
						264	34.97	35.5	2
						Lubricating oil	20	833	29.3
137	3.19	3.86	21						
151	5.38	5.28	-2						
189	9.53	11.2	17						
226	16.21	20.9	29						
208	16.29	15.6	-4						
215	17.34	17.5	1						
215	18.29	17.5	-4						
250	28.86	29.8	3						
259	30.40	33.8	11						
267	33.42	37.7	13						
272	35.68	40.2	13						
269	37.48	38.7	3						
273	37.77	40.7	8						
274	40.42	41.3	2						
281	43.44	45.2	4						

^aNote - Average of multiple determinations.



Average difference in all data, 56 percent
 Average difference in reference 20 data, 19 percent
 Average difference in reference 21 data, 136 percent
 Average difference in reference 21 10° C data, 31 percent
 Average difference in reference 21 30° C data, 99 percent
 Average difference in reference 21 50° C data, 279 percent
 Average difference in all data except reference 21 at 50° C, 30 percent

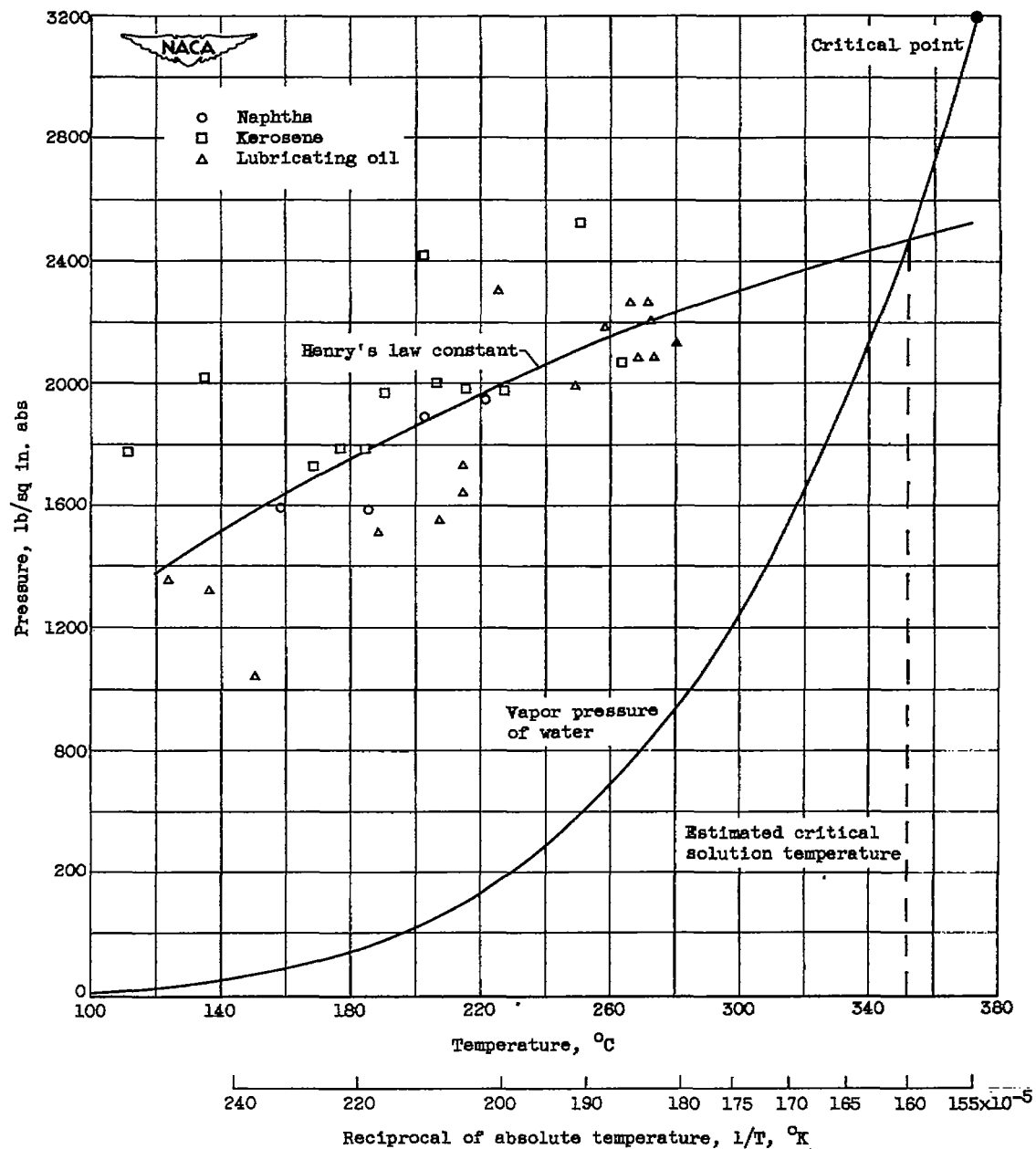
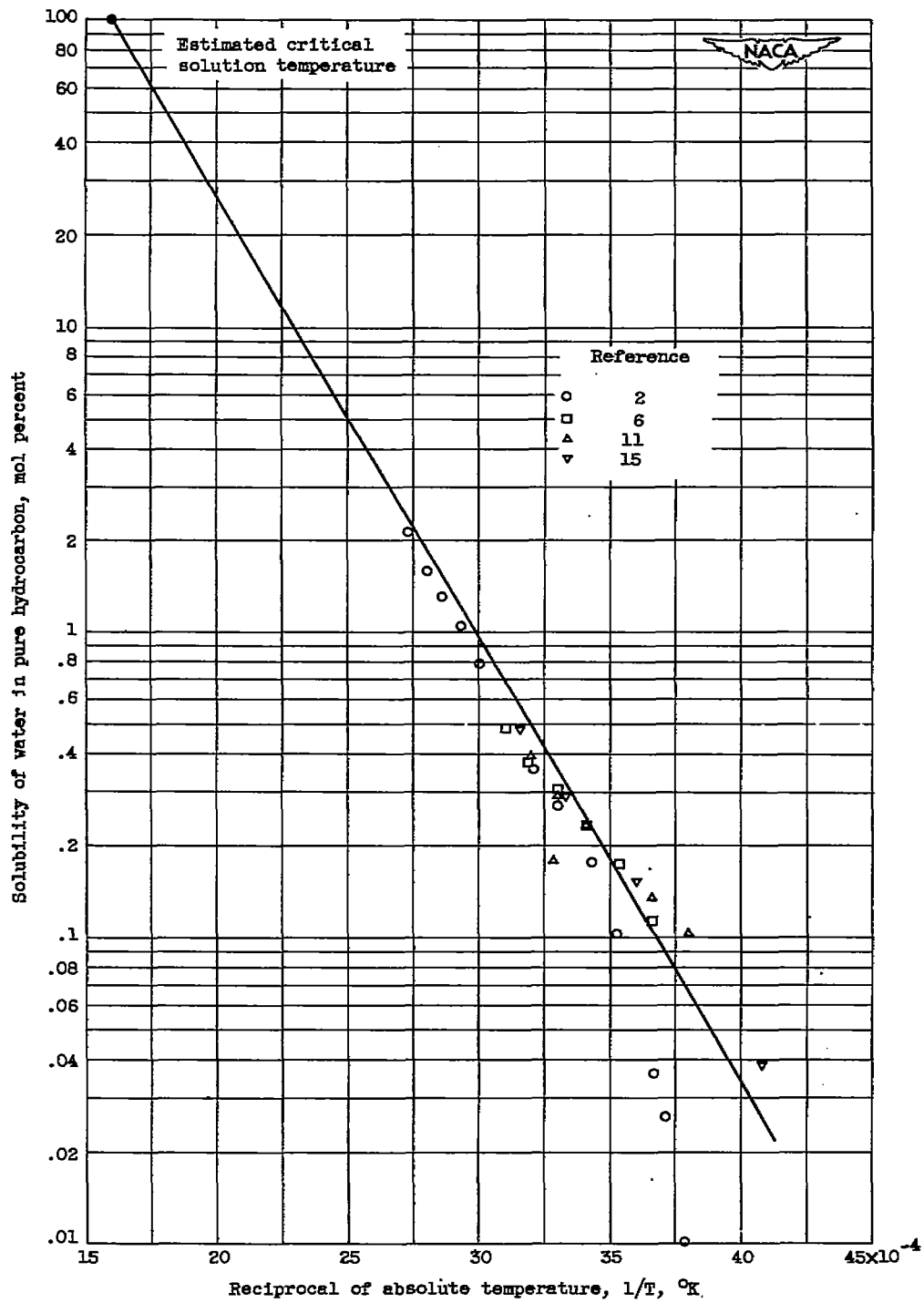
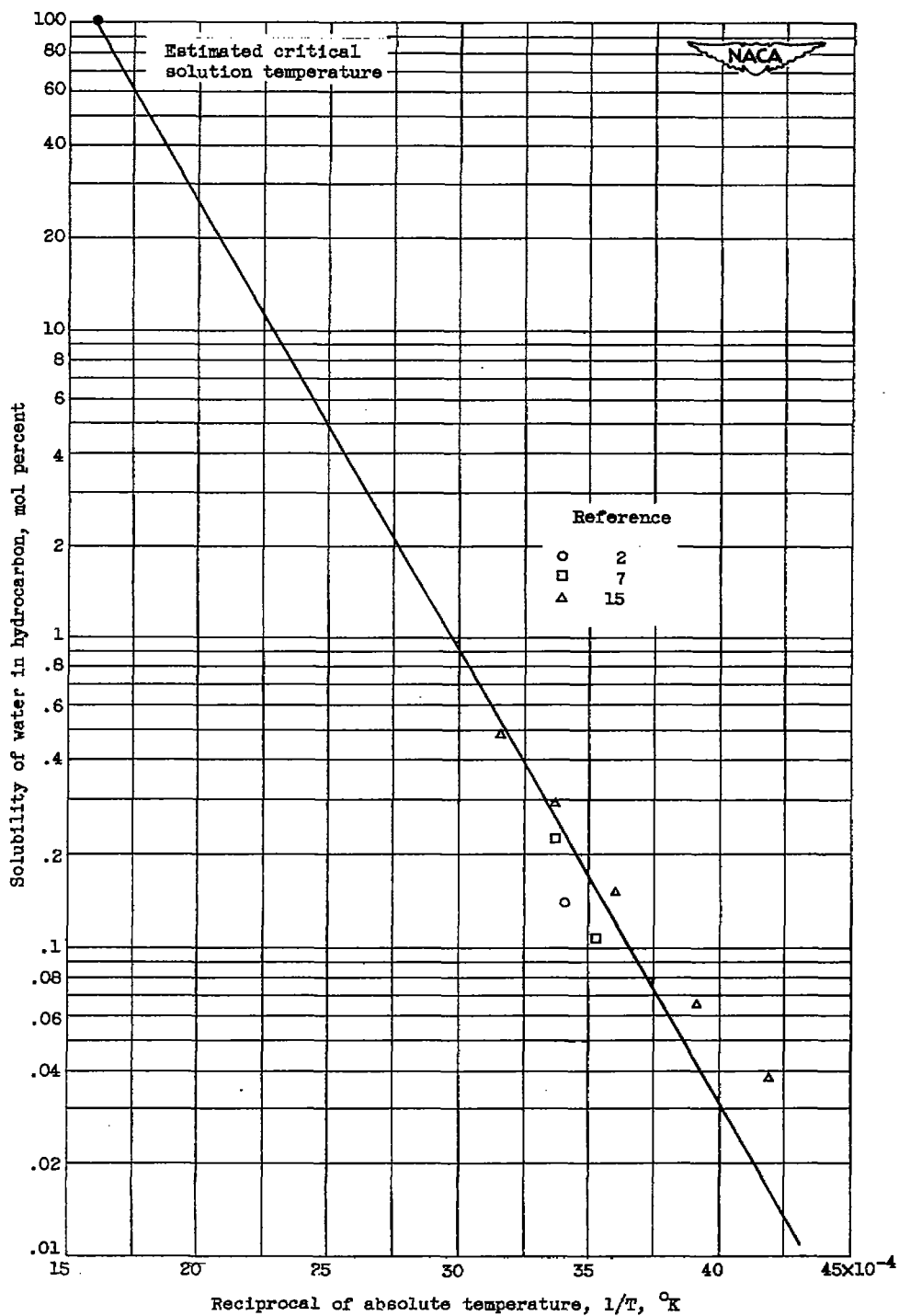


Figure 1. - Henry's law constant and vapor pressure of water as functions of temperature.



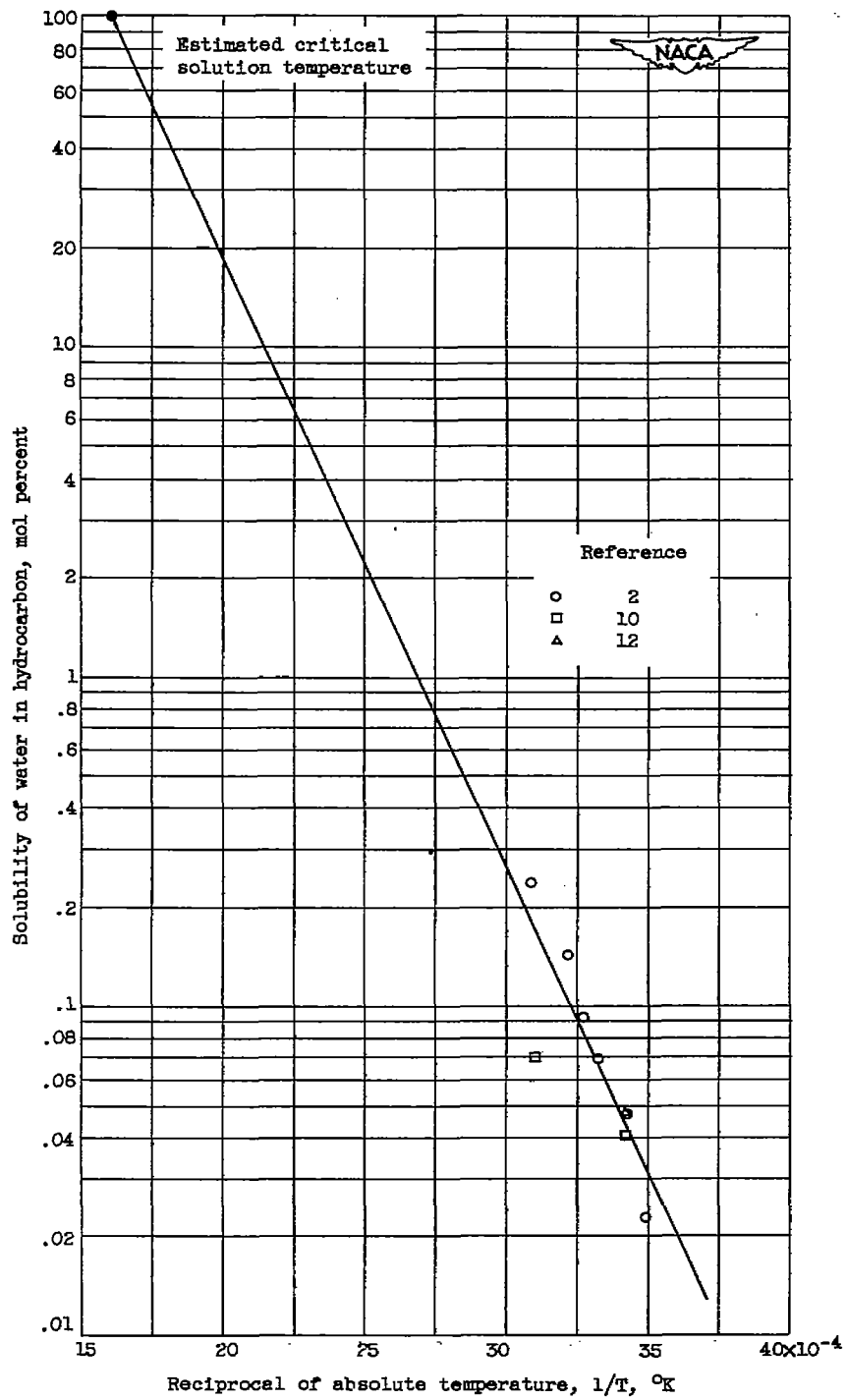
(a) Toluene.

Figure 2. - Solubility of water in pure hydrocarbons as function of temperature.



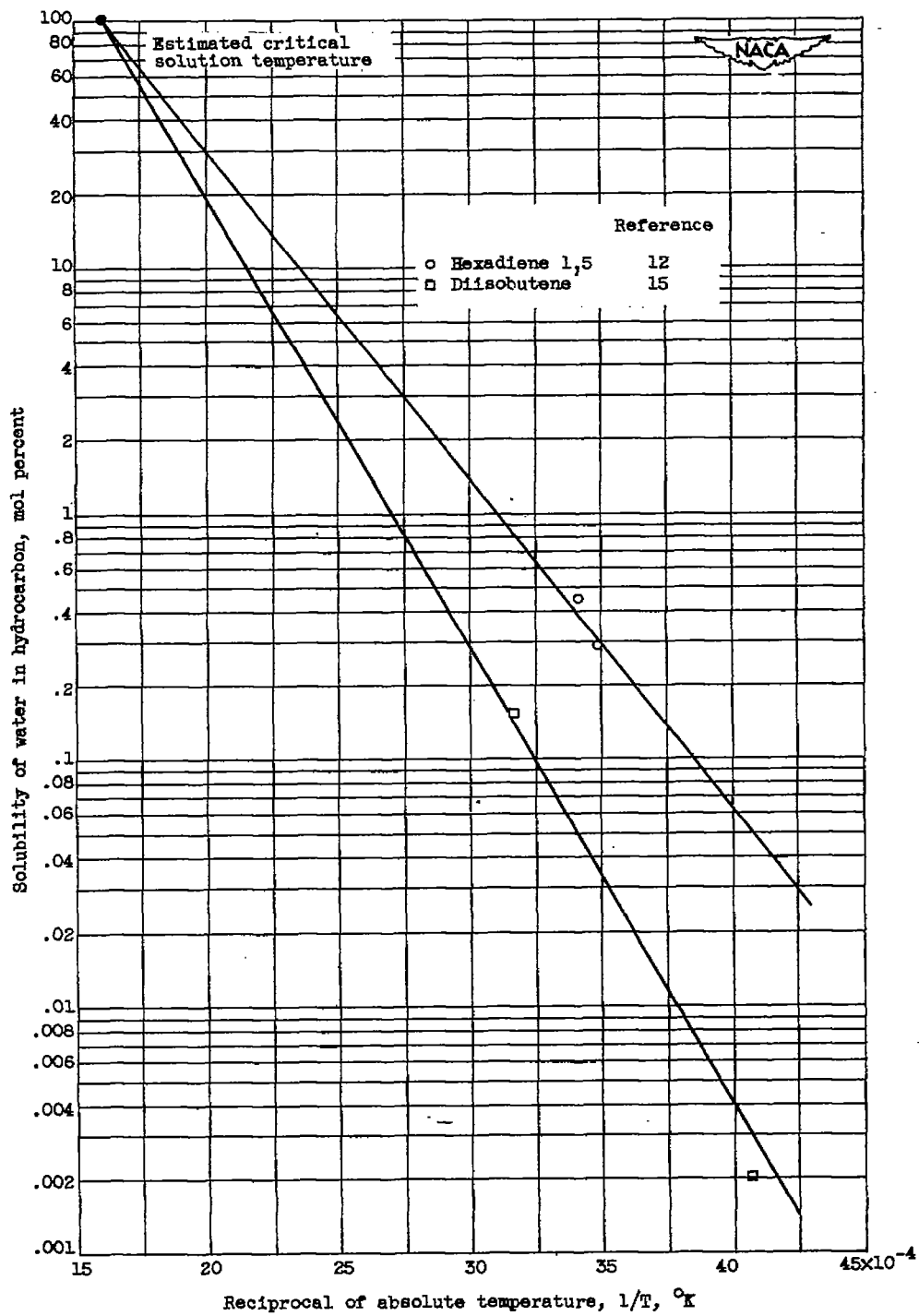
(b) Xylene.

Figure 2. - Continued. Solubility of water in pure hydrocarbons as function of temperature.



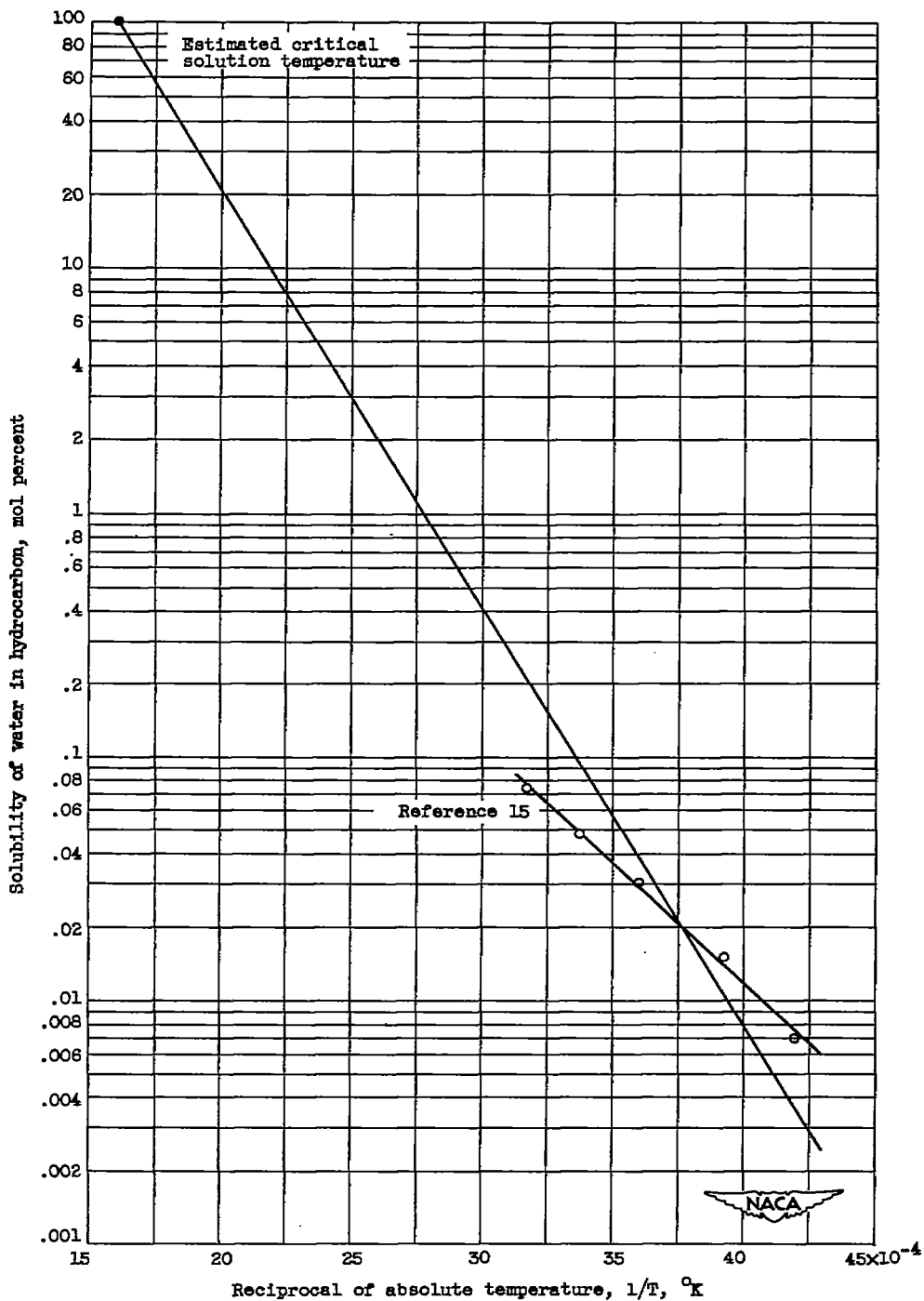
(c) Cyclohexane.

Figure 2. - Continued. Solubility of water in hydrocarbons as function of temperature.



(d) Hexadiene 1,5 and diisobutene.

Figure 2. - Continued. Solubility of water in pure hydrocarbons as function of temperature.



(e) Methylcyclohexane.

Figure 2. - Continued. Solubility of water in pure hydrocarbons as function of temperature.

2336

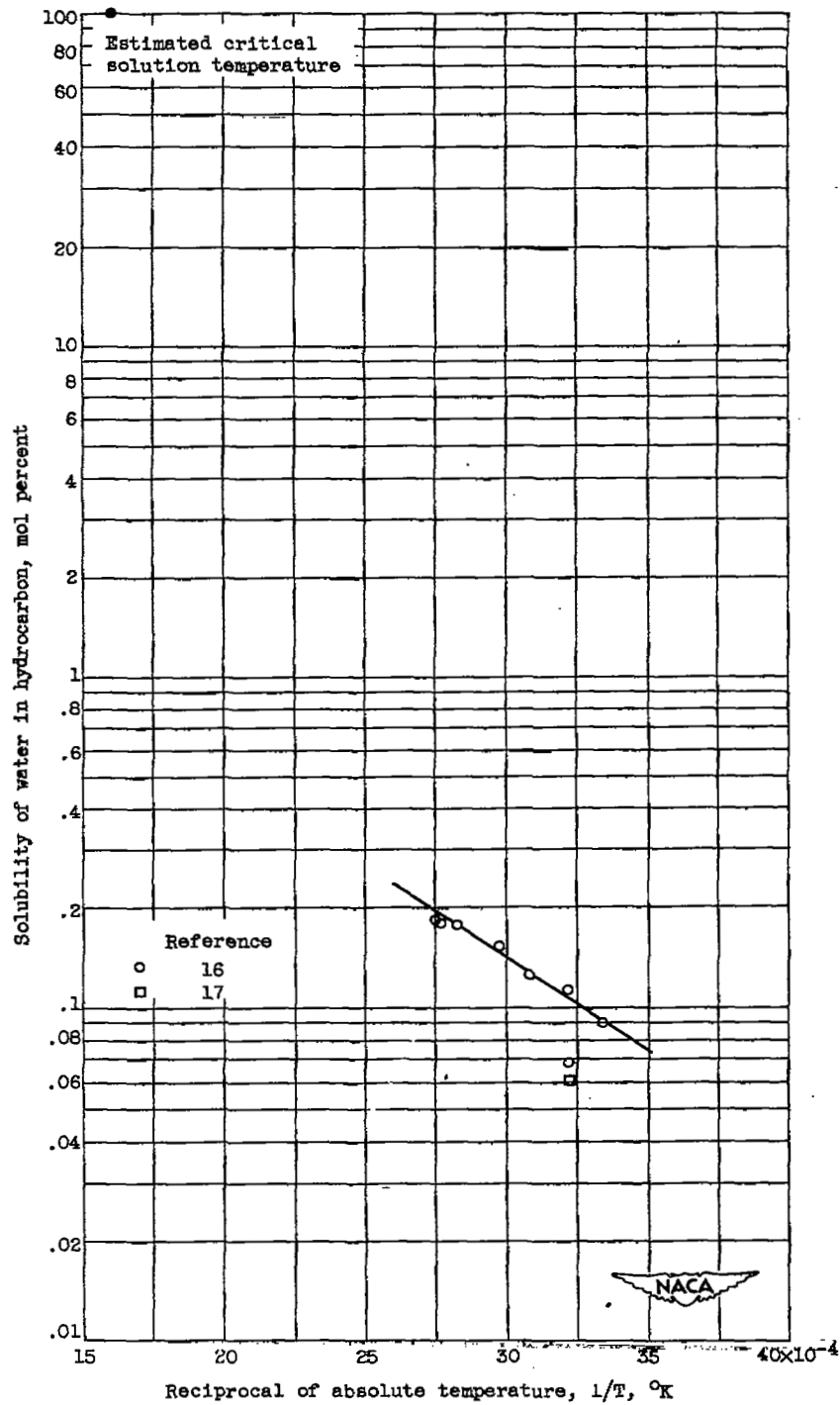


Figure 2. - Concluded. Solubility of water in pure hydrocarbons as function of temperature.

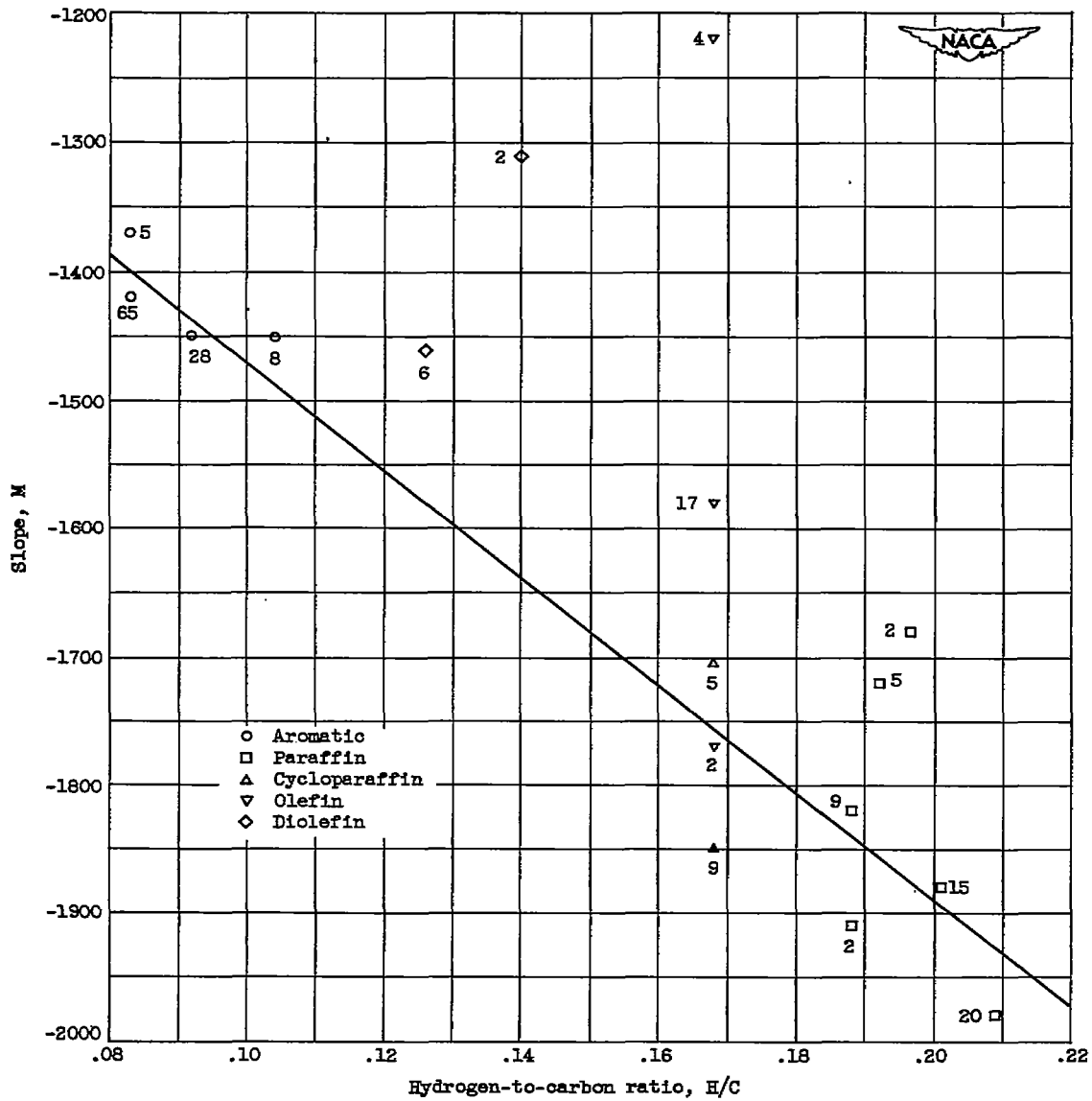
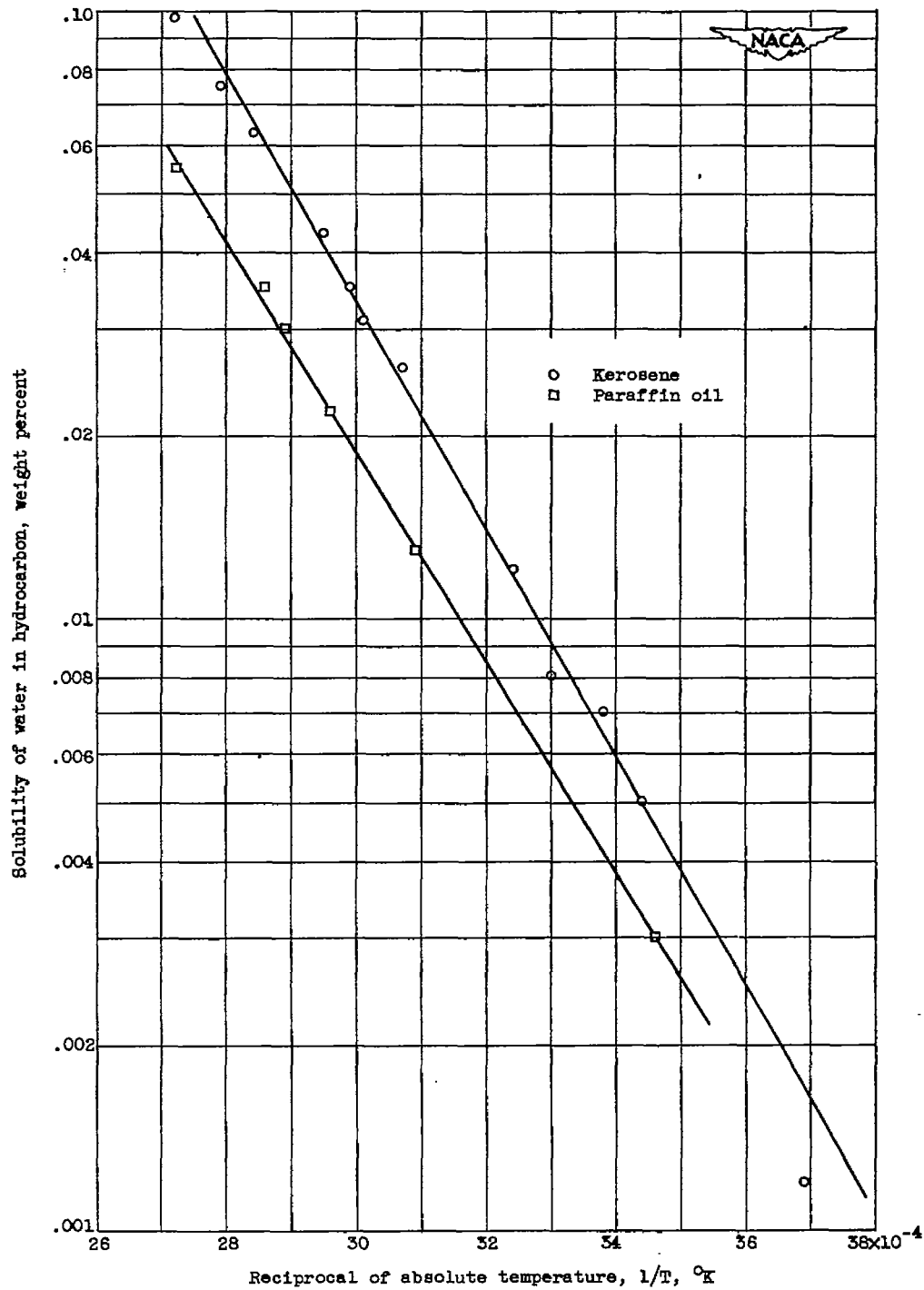
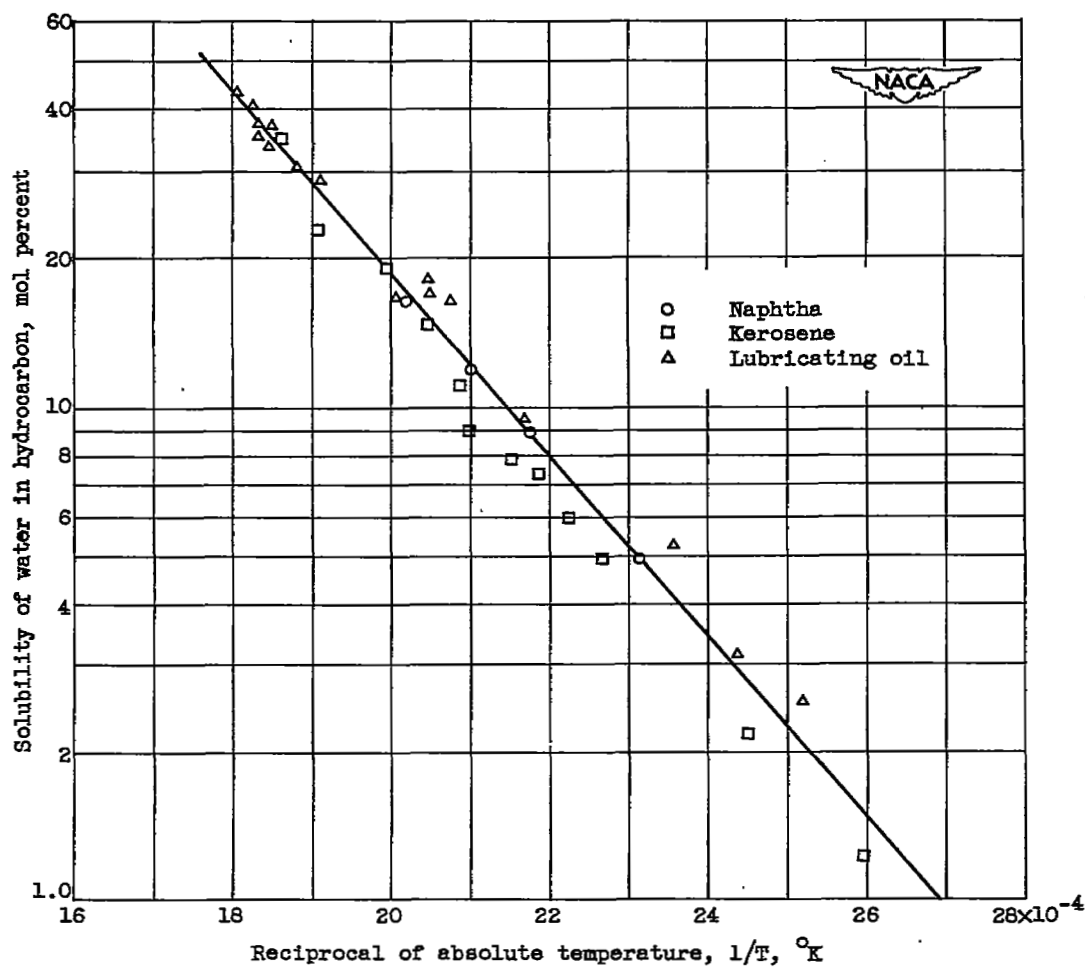


Figure 3. - Slope of solubility correlation line as function of hydrocarbon H/C ratio. Numbers indicate number of determinations represented by each point.



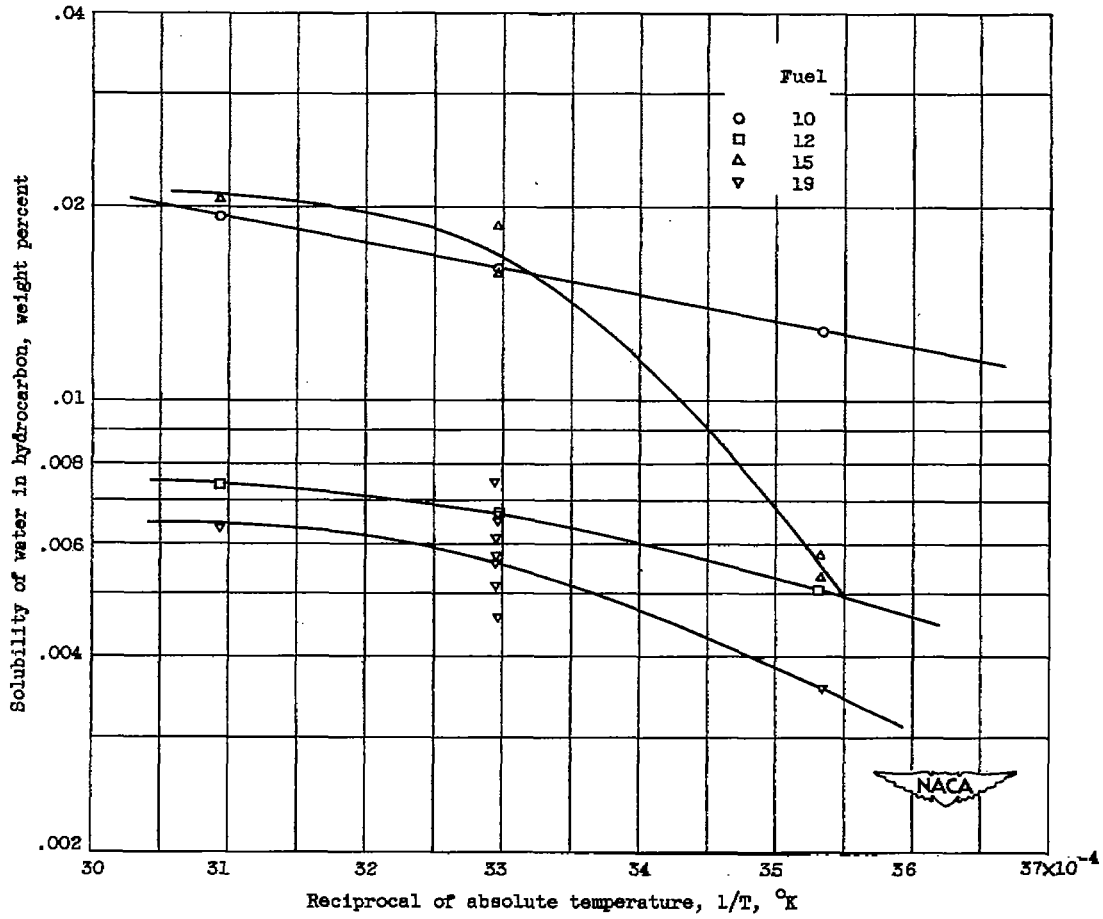
(a) Kerosene and paraffin oil, reference 1.

Figure 4. - Solubility of water in petroleum fractions as function of temperature.



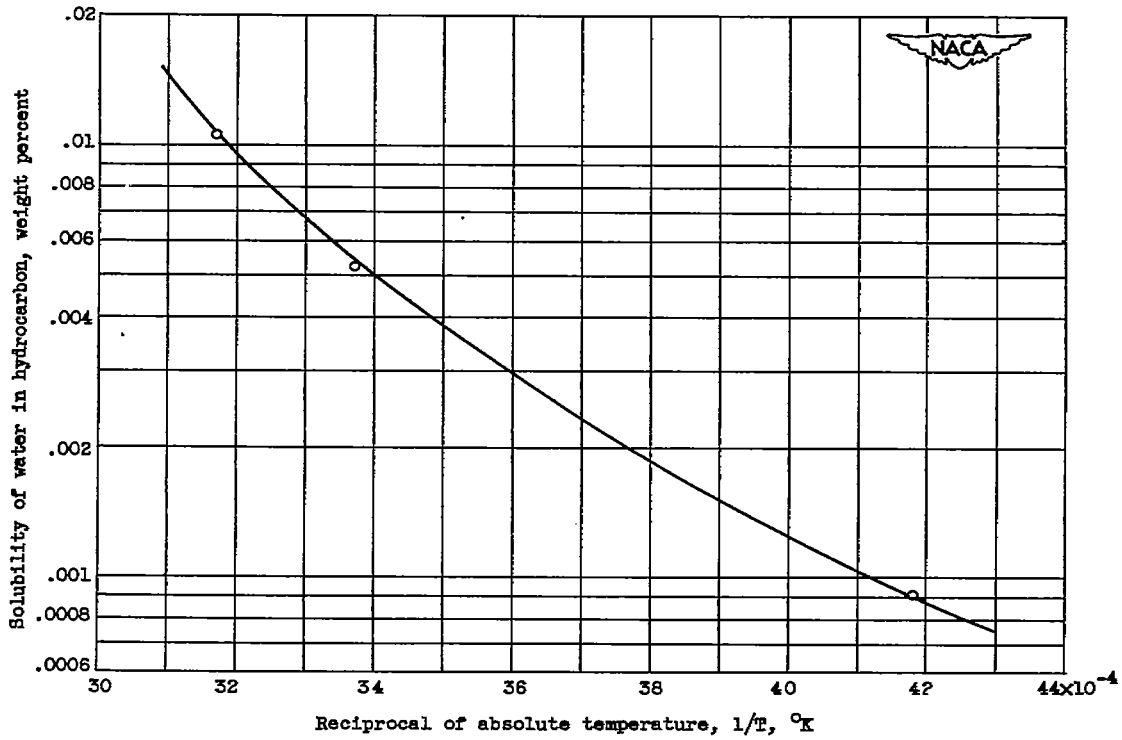
(b) Naphtha, kerosene, and lubricating oil, reference 20.

Figure 4. - Continued. Solubility of water in petroleum fractions as function of temperature.



(c) Four gasolines, reference 21.

Figure 4. - Continued. Solubility of water in petroleum fractions as function of temperature.



(d) Safety fuel, reference 15.

Figure 4. - Concluded. Solubility of water in petroleum fractions as function of temperature.

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