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AROMATIC HYDROCARBONS AS COMPONENTS OF AVIATION GASOLINE

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As a result of research conducted at the Central Institute of Aviation Gasolines and Lubricants (TsIATIM) and the State Red Army Scientific Research Institute of the Red Army Air Force (GKNII VVSKA) to determine the optimum chemical composition for aviation fuels, it was determined that aromatic hydrocarbons were a valuable addition to fuels being used in engines which are repeatedly operated at overload conditions. It was also possible to draw various theoretical conclusions relevant to the role of aromatic hydrocarbons in the knock-free combustion of aviation fuels.

Laboratory and engine performance tests showed that aromatic hydrocarbons have a high octane rating. Thus benzene, toluene, and xylene have 100-plus octane ratings, but are unsuited for the purpose in view, as they will not increase the octane rating of a fuel if added in small quantities. This can be seen from the table below:

Composition (%)		Octane Rating (according to engine method of testing) of Mixtures with Component			
Gasoline with Octane Rating of 70	Component	Isooctane	Benzene	Toluene	Xylenes
100	0	70	70	70	70
90	10	73.5	71.5	72	72
80	20	76	73.5	74.5	74.5
70	30	78.5	76	77	77
50	50	83.5	82	83	83

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It was also observed that aromatic hydrocarbons, when pure or blended with some other substance, were less sensitive to tetraethyl lead than straight-chain paraffin hydrocarbons.

It was generally believed that large amounts of aromatic hydrocarbons must be added to gasoline to improve the latter's performance, but there was never any doubt as to the effect of these hydrocarbons with respect to increasing the antiknock qualities of fuels. Prior to the introduction of ethyl fluid, a mixture of 65 percent aviation benzene and 35 percent Baku gasoline was used as aviation fuel.

Around 1936 the US Air Force first started using air-cooled engines with high-pressure fuel injection. It was then held that with the increased operating temperatures, it would not be advisable to use aromatic hydrocarbons which are more sensitive to temperature changes than paraffin hydrocarbons, and would lose some of their antiknock qualities with temperature increases. In 1936 the Army method of testing was adopted in the US, whereby the octane rating was determined according to the temperature of the (cylinder) head. The use of Midgley's needle was discontinued. The temperature of the cooling sleeve of the cylinder was increased to 160 degrees centigrade, as compared with a temperature of 100 degrees centigrade under the old engine method of testing. A scale was compiled using as a starting point that degree of compression under which benzene and a mixture of 88 percent chemically pure isooctane and 12 percent n-heptane produce the same heating of the (cylinder) head. This temperature is assumed to be the standard intensity of knocking for drawing up the scale and for determining the octane rating of fuels. Benzene was found to have an octane rating of 88.

In 1936, a special fuel conference was held in Moscow. Much of the discussion centered around the value of aromatic hydrocarbons as additives to aviation gasolines. But there were many at TsIATIM and GKNII VVSKA who did not believe in the value of this component. Workers at TsIATIM (Ye. I. Zabryanskiy) showed by fuel tests, according to the Army method, that fuels containing aromatic hydrocarbons actually had a lower octane rating than that found in the engine performance test. However, fuels which had a high aromatic hydrocarbon content were more easily modified by tetraethyl lead under the new procedure than in the engine performance tests. Therefore it can be concluded that the sensitivity of fuels with a high aromatic hydrocarbon content to increased temperatures prior to ignition is largely suppressed by tetraethyl lead.

During 1938 and 1939, a series of tests was conducted with various fuels to evaluate their performance in the M-25A engine. These tests were carried out in connection with the development of a special antiknock alkylbenzene by TsIATIM which was brought up to the stage of industrial production in 1936. The tests themselves were conducted at GKNII VVSKA under the supervision of Army Engineer Filippenko. Ignition qualities were determined with two matters in mind: (1) the permissible degree of leanness and (2) the permissible amount of pressure feed. Prior to the above-mentioned tests, the test motor was operated on B-59, B-70, B-74, and B-78 gasolines with and without tetraethyl lead to determine the octane rating of the latter fuels.

The following fuel mixtures were used:

1. Fifty percent isooctane + 50 percent B-70 gasoline + 2 milliliters of ethyl fluid per kilogram

2. Forty percent alkyl benzene + 60 percent B-70 gasoline + 3 milliliters of ethyl fluid per kilogram

3. B-78 gasoline + 4 milliliters of ethyl fluid per kilogram

According to the engine test method all of the fuel mixtures had octane ratings of 96 to 97, but it was found that fuels with varying chemical compositions behaved differently in a M-25A aircraft engine.

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A mixture of alkylbenzene and straight-run gasoline or B-78 gasoline +4 milliliters per kilogram of ethyl fluid has less tendency to produce knocking than isooctane blends with the same octane rating.

No knocking was observed when the leanness of mixtures of alkylbenzene was lowered to a rate of consumption of 216 grams per horsepower-hour under conditions of rated power output. The maximum temperature of the cylinder heads during the above run remained constant at 222 degrees centigrade.

Infrequent knocking was observed in a run using a mixture of isooctane with straight-run gasoline at a consumption of 212 grams per horsepower-hour. The maximum cylinder head temperature in this case was 230 degrees centigrade.

Tests conducted by raising the amount of pressure feed showed that when 250 grams of fuel were expended per horsepower-hour a mixture of alkylbenzene and gasoline gave the best antiknock performance at the highest pressure feed. No knocking was observed until full throttle, i.e., until P_k = 1,119 millimeters of mercury and N_c = 835. Maximum cylinder head temperature at full throttle operation was 249 degrees centigrade. Knocking started at P_k = 1,110 millimeters of mercury for a mixture of isooctane and gasoline. Maximum cylinder head temperature reached was 248 degrees centigrade.

It was necessary to operate at full throttle before knocking was observed with a mixture of B-78 gasoline containing 4 milliliters of ethyl fluid per kilogram. (P_k = 1,124 millimeters of mercury and N_c = 813). Maximum cylinder head temperature was as high as 250 degrees centigrade. It is of interest that where engine operation was such as to consume fuel at the rate of 280 grams per horse power-hour no knocking was observed even at full throttle with any of the fuels tested.

Full throttle operation on a mixture of alkylbenzene and gasoline gave 850 horsepower while on a mixture of B-78 gasoline and 4 milliliters per kilogram of ethyl fluid the maximum horsepower was only 838.

It can therefore be seen that aromatic hydrocarbons can be valuable high-octane components of aviation gasolines and that they have the characteristics of performing better at higher pressure feeds, also of developing more power than isoparaffins. From 1939 to 1940, associates at TsIATIM conducted tests on an M-105 engine and determined that engine performance was far better with fuels containing large amounts of aromatic hydrocarbons than could be expected merely on the basis of their octane rating. It was proposed that pyrobenzene could be used as an additive to B-78 fuel (at this time it was being used only as an additive to B-70 fuel).

In 1942, a shipment of US aviation gasoline arrived in the USSR, which was classed B-95 and B-100, to correspond to the octane rating. The B-95 contained 76 percent paraffin hydrocarbons, 20 percent naphthenes, and only 4 percent aromatics; while the B-100 contained 90 percent paraffin hydrocarbons, 6 percent naphthenes, and 3 percent aromatics. Thus both fuels possessed predominantly paraffin hydrocarbon characteristics (see Table 1). These two fuels were tested in an aircraft engine which operated under the following conditions: intake pressure (pressure feed = Pa) 1,288 millimeters of mercury at take-off conditions, 2,150 rpm, compression 6.8, and output 34.3 horsepower per liter of fuel. It was first established that the engine performed best on a fuel consisting of ordinary B-78 with 4 milliliters of ethyl fluid per kilogram of gasoline. This fuel had a high naphthene hydrocarbon content, and with the addition of a given amount of tetraethyl lead would have had an octane rating of around 95. It was therefore believed that the engine would operate normally on the US fuels.

Table 1

Constants	B-78 Gaso-line	B-95 Gaso-line	Pyro-benzene	Xylene Fraction	20% Pyro-benzene + 80% B-95	30% Pyro-benzene + 70% B-95	20% Xylene Fraction + 80% B-95	30% Xylene Fraction + B-70
Specific weight d_4^{20}	0.7280	0.7180	0.8509	0.8317	0.740	0.747	0.731	0.745
Octane rating (engine method)	94.8	95	89 (*2	82.2 (*3	95.0	94.7	95.0	93.4
Ethyl fluid (ml/kg)	4.2	4.2 (*1	0	0	4.0	4.0	4.19	4.08
Fractions:								
Boiling begins °C	46.5	49.5	83	136	51	51	66	53
10% boils off at °C	68	70.5	85	140	72.5	77	74	75.5
50% boils off at °C	92	97	100	146.5	97	109	102.5	106
90% boils off at °C	125.5	120	149.5	160	132	149.5	141	147
97% boils off at °C	159	150	169	173.5	164	168.5	168	170
Residue (%)	1.7	1.1	0.9	0.8	1.1	1.3	1.1	1.0
Loss (%)	0.8	1.1	0.9	0.8	1.1	1.3	1.1	1.0
Congelation temperature °C	-60	-60	-20	--	Less than -60	-55	Less than -60	-60
Chemical composition:								
Paraffin hydrocarbons (%)	39	76	20	30	--	--	--	--
Naphthene hydrocarbons (%)	58	20			--	--	--	--
Aromatic hydrocarbons (%)	3	4	80	70	20 (*1	27 (*4	17 (*4	24 (*4

*1. B-95 contains 3.19 ml ethyl fluid per kilogram (1 ml per kilogram was added besides)
*2. With 4 ml ethyl fluid per kg the octane rating is 94
*3. With 4 ml ethyl fluid per kilogram the octane rating is 87
*4. Date on aromatic hydrocarbon content are established by computation

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The fuels were tested by determining engine performance on various fuel mixtures with the maximum permissible lean mixtures set by the altitude mixture control under the following conditions: (1) Take-off conditions at P_a = 1,285 millimeters of mercury (2,150 rpm), (2) Nominal at P_a = 1,180 millimeters of mercury (2,050 and 1,950 rpm).

During these tests the temperature of the water at the exit was maintained at 65 - 75 degrees centigrade. Under operating conditions, the specific expenditure of fuel at a point where knocking starts should not exceed 300 - 325 grams per horsepower-hour at take-off conditions, and 285 - 305 grams per horsepower-hour nominal (at 2,050 rpm). These fuels had a high octane rating, but proved to be inadequate for good performance under overload operating conditions. The B-95 fuel produced very obvious knocking at nominal output where fuel expenditure was $C_e = 291$ grams per horsepower-hour. It was decided that the octane rating of a fuel is no criterion for determining the suitability of a fuel for use in a pressure-feed, low-rpm, large dimensioned engine. However, it was suggested that these US fuels would give very good performance if doctored with aromatic hydrocarbons.

A series of experiments (supervised by Colonel S. P. Kachenovskiy of the Engineers) was conducted to test this theory. Pyrobenzene was added to B-95 and B-100. Another series of tests was conducted using the "xylene fraction" of pyrobenzene (this is the pyrobenzene residue after the 125 - 130 degrees centigrade fraction is distilled). The properties of this fraction are shown in Table 1.

The following mixtures were tested by GKNII VVSKA and TsIATIM during September and October 1942: (1) 80 percent B-95 and 20 percent pyrobenzene, (2) 70 percent B-95 and 30 percent pyrobenzene, (3) 80 percent B-95 and 20 percent xylene fraction, and (4) 70 percent B-95 and 30 percent xylene fraction. (All mixtures contained 4 milliliters per kilogram of ethyl fluid.) An analysis of the mixtures and a group chemical composition of the fractions is shown in Table 1.

The results of the experiments are shown in Table 2. It can be seen that performance was normal with all mixtures notwithstanding the fact that mixtures with 20 percent pyrobenzene and particularly those with 30 percent xylene fraction had a much lower octane rating than conventional B-95 (xylene fraction with 4 milliliters per kilogram of ethyl fluid has an octane rating of 87.)

It was shown that:

1). Specific expenditure of fuel during take-off and at nominal output of an engine operating on a lean mixture until the point of knocking was less with a mixture of 80 percent B-95 and 20 percent xylene fraction than with a mixture of 80 percent B-95 and 20 percent pyrobenzene. This is due to the high qualities of the aromatic hydrocarbons which are contained in the xylene fractions (xylene fractions contain all the pyrobenzene hydrocarbons, in addition to benzene; toluene is contained in minute quantities in pyrobenzene).

2. The relatively lower octane rating (93.4) of a mixture of 70 percent B-95 and 30 percent xylene fraction, as well as the higher specific expenditure of fuel under lean mixture operation until the start of knocking (in comparison to performance with fuel containing 20 percent xylene fraction) is evidence of the poor antiknock qualities of paraffin and naphthene hydrocarbons which are found in the xylene fractions.

B-95 fuel henceforth was improved by adding only small amounts of pyrobenzene. It is also not recommended that more than 20 percent of pyrobenzene be added in view of the additional circumstance that an amount in excess of this will raise the congelation point of gasoline above-60 degrees centigrade. The results of these experiments resulted in a completely new evaluation of aromatic hydrocarbon components of aviation fuels. A similar change of opinion has also taken place in the US and England. Today it is general practice to use aromatic hydrocarbons in gasolines for engines performing under overload conditions, particularly if the principal constituents of the gasoline are paraffin hydrocarbons.

It may be assured that the antiknock action of aromatic hydrocarbons is twofold: (1) oxidation products of aromatics prevent an accumulation of those compounds which result from the oxidation of paraffin hydrocarbons and cause the detonation wave, (2) action consisting in dilution of the paraffin hydrocarbons, thus reducing the number of collisions between paraffin hydrocarbon molecules or free radicals and active oxygen.

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Table 2

Specific Expenditures at Start of Knocking (in gr per hp-hr)

Identification of Fuel	Take-Off Pa = 1,280 mm n = 2,150 rpm	Rated Performance Pa = 1,180 mm n = 2,050 rpm	Rated Performance Pa = 1,180 mm n = 1,950 rpm	Octane Rating
Established maximums	300 - 325	285 - 305	Not established	--
B-95 (containing 4 ml/kg ethyl)	Knocks	Knocks	Knocks	95.0
30% pyrobenzene +70% B-95 (containing 4 ml/kg ethyl)	269	265	247	95.0
20% pyrobenzene +80% B-95 (containing 4 ml/kg ethyl)	292	263	275	94.7
30% xylene fraction +70% B-95 (containing 4 ml/kg ethyl)	295	275	273	93.4
20% xylene fraction +80% B-95 (containing 4 ml/kg ethyl)	271	257	253	95.0
B-78 (containing 4 ml/kg ethyl)	280	241	287	94.8

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According to Rice (1), there is a breaking up of hydrocarbons into low molecular olefins and paraffins as the temperature increases. This occurs over the formation of radicals. The number of free radicals of straight-chain hydrocarbons formed in the engine cylinder is larger than the number of those formed from branched isomers. Rice claims that there is a direct relationship between the number of free radicals which are formed and the tendency of the engine to knock. On formation, the lowest hydrocarbons, and possibly free radicals which collide with the active oxygen, are transformed into that type of oxidized product which in itself is the cause of knocking (for example, peroxides in the second stage of combustion).

It is quite evident that [illegible] higher temperatures and partial pressures of oxygen, a greater quantity of low hydrocarbons will form without preliminary formation of radicals. These factors will also bring about more intensive oxidation of the newly formed products. It follows, therefore, that with an increase in the partial pressure of the oxygen and an increase in the temperature, there will occur knocking during the combustion of hydrocarbons which actually have high knock-resistant qualities.

Studies to determine the octane rating were conducted under conditions not employing pressure feed. Consequently, the temperature of the engines was low notwithstanding the high oxygen pressure in the engines. The results obtained from these studies could not be used for evaluating the performance of fuels in engines performing at overload operating conditions. It must be stated that the action of aromatic hydrocarbons, which would prevent the accumulation of oxidation products causing knocking, leads to the conclusion that they have the same action as aromatic hydrocarbons which prevent the oxidation of oils, i.e., that they act as inhibitors of oxidation. This was shown in studies conducted by N. I. Chernozhukov and S. E. Kreyn(2).

For the purpose of breaking the chain of oxidation effectively, aromatic hydrocarbons must themselves oxidize, but not in the side chains. Oxidation must occur in the nucleus with the resultant formation of phenols in addition to other oxidized compounds. For example, it can be assumed that oxidized aromatic hydrocarbons react with hydroperoxides in a manner similar to that suggested by Egerton (3) for the case of metallo-organic antiknock substances which prevent the formation of peroxides.

Under rigid oxidation conditions, such as are evident in modern engines operating at overload, there is an efficient oxidation of the aromatic nucleus, so that aromatic hydrocarbons are effective antiknock components of aviation fuels. In engines which do not operate at overload conditions (for example, the engines which were used to determine the octane rating of fuels), the aromatic hydrocarbons do not undergo the necessary oxidation and therefore do not serve as effective antioxidizing agents. Instead of breaking the oxidation chain, they merely dilute the fuel. Under the circumstances, it was necessary to add large amounts of aromatic hydrocarbons in order that they be effective.

Due to the antioxidant action of aromatic hydrocarbons, it is sufficient to add them in relatively small quantities (15 percent or so) in order to prevent the knocking set up by isomeric paraffin hydrocarbons.

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Table 3

Specific Expenditures at Start of Knocking

Identification of Fuel	Take-Off Pa = 1,280 mm n = 2,150 rpm	Rated performance = Pa = 1,180 mm n = 2,050 rpm	 n = 1,950 rpm	Octane Rating
Established maximums	300 - 325	285 - 305	Not established	--
B-70 gasoline (60%), isooctane (40%), with 4 ml/kg ethyl	289	246	235	95.4
B-70 gasoline (60%), neohexane (40%), with 4 ml/kg ethyl	327	275	301	95
B-70 gasoline (60%), neohexane (20%), isooctane (20%), with 4 ml/kg ethyl	280	242	253	--
B-70 gasoline (60%), neohexane (10%), isooctane ([illegible]%), with 4 ml/hg ethyl	287	269	275	95.3
B-70 gasoline (60%), isopentane (8%), isooctane -- 40%-industrial mixture -- (32%) with 4 ml/kg ethyl	313 Knocks	291 Knocks	294 Knocks	--

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Nevertheless, it must be kept in mind that aromatic hydrocarbons do not inhibit oxidation and consequently knocking caused by paraffin hydrocarbons in an entirely uniform manner. For the purpose of inhibiting the oxidation of low-octane-rating hydrocarbons which have a normal or weakly branched structure, it is necessary to use larger amounts of aromatic hydrocarbons than for hydrocarbons having several side methyl groups. In the former case the aromatic inhibitor has to break a much larger number of oxidation chains.

It can be seen from Table 3 that the xylene fraction acts with greater effect in 20 percent amounts than in 30 percent amounts. This can be explained by the fact that paraffin hydrocarbons with a low octane rating are components of xylene fractions. Moreover, they are weakly branched, thus explaining the relatively low octane rating of the xylene fraction. Thus the octane rating of 70 percent B-95 and 30 percent xylene fraction mixture is lower than that of a mixture of B-95 with the same amount of ethyl fluid (93.4 - 95). Engine performance with a mixture containing 30 percent xylene fraction is poorer than with a mixture containing only 20 percent xylene fraction, due to the fact that in a 30 percent mixture the paraffin hydrocarbon constituents deteriorate in quality, so that even a large amount of aromatic hydrocarbons will not inhibit oxidation and prevent knocking.

The above has a practical as well as theoretical significance. At present, it cannot be stated conclusively that there is no necessity for the manufacture of isoparaffins. In view of the fact that isoparaffins are most easily protected against knock-producing decomposition, their presence in fuels is desirable. Therefore the continued manufacture of hydrocarbons of this type serves a useful purpose.

Separate studies should be conducted to determine the performance of overload operating engines on naphthene hydrocarbons. From Table 2 it can be seen that the B-78 gasoline with 4 milliliters of ethyl fluid per kilogram (octane rating 95) did not produce knock in overload operating conditions. This gasoline is obtained by straight distillation and contains 60 percent naphthene hydrocarbons, but only 2 - 3 percent of aromatic hydrocarbons. It may be surmised that naphthene hydrocarbons, under severe oxidation conditions, are partially converted into antioxidizing agents and thus protect the rest of the naphthene as well as paraffin hydrocarbons from knock-producing combustion. Both Chernozhukov and Kreyn (2) observed that in the oxidation process, naphthene hydrocarbons change into aromatic hydrocarbons. Consequently, all prerequisites for the conversion of naphthene hydrocarbons into antioxidizing agents are present.

A most interesting observation was made at GKNII VVSKA (reported 25 December 1942): the addition of isooctane to paraffin hydrocarbon gasolines B-95 and B-100 did not bring about knock-free performance of engines at overload operating conditions. A mixture of 40 percent isooctane with B-70 gasoline (having an octane rating of 98 with 4 milliliters of ethyl fluid per kilogram and 60 percent naphthene hydrocarbon content) resulted in knock-free operation. It follows therefore that both components -- naphthene gasoline and isooctane -- mutually prevent knocking. Still, naphthene hydrocarbons are less effective antiknock agents than aromatic hydrocarbons when added to isoparaffins. Thus, the same naphthane gasoline did not prevent knocking in neohexane fuel and was totally ineffective in preventing knocking in a mixture of isooctane and isopentane (Table 3). Obviously the antiknock stability of isopentane and neohexane under overload operating conditions is different from that of isooctane, notwithstanding the fact that their octane rating, when in a 40:60 mixture with B-70 gasoline (with 4 milliliters of ethyl fluid per kilogram) is the same(95).

This final problem must be further studied, as it is possible that poor performance of fuels containing neohexane and isopentane may be due to defective distribution of the fuel among the cylinders.

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Conclusions

1. Aromatic hydrocarbons with low boiling point (benzene, toulene, xylene and ethyl benzene) increase the octane rating of gasoline only when added in large quantities, particularly in the presence of tetraethyl lead. These conclusions were reached on the basis of tests conducted according to the CFR engine method.

2. Under overload operating conditions (with high pressure feed) fuels composed primarily of isoparaffins cause knocking even where the octane rating is high (95 to 100).

3. The introduction of small quantities of aromatic hydrocarbons into paraffin high-octane fuels results in a sharp inhibition of knocking in an engine operating under overload conditions, even through the octane rating of the fuels is not increased.

4. Fuels which are composed of naphthene hydrocarbons, -- for example, straight run B-78 with up to 60 percent of these hydrocarbons (with 4 milliliters of ethyl fluid per kilogram), -- burn in an engine operating at overload conditions without knocking, if the fuels have high octane ratings.

5. The differences in the performance of fuels rich in paraffins, naphthenes and aromatic hydrocarbons in present-day engines which operate at overload conditions and in CFR equipment, as well as in other carburetor engines not working at overload conditions, can be stated as follows:

a. Paraffin hydrocarbons having a branched structure and a high octane rating do not operate without knocking in an engine operating at overload conditions. This results in high operating temperatures and high partial oxygen pressure. Isoparaffin hydrocarbons can be protected by the addition of small quantities of aromatic hydrocarbons. A much larger amount of aromatic hydrocarbons is necessary for knock prevention when normal or weakly branched paraffin hydrocarbons are used in engines operating at overload conditions.

b. The action of aromatic hydrocarbons is twofold: (1) as a diluent of hydrocarbons which have a tendency to knock, and (2) by breaking the chain of oxidation. This latter effect is not brought about directly by the aromatic hydrocarbons, but rather by the products of their oxidation. Under mild oxidation conditions, such as are met in the Waukesha test (engine method) there is no oxidation of aromatics. Under severe conditions of oxidation, in these same engines (high temperatures, high partial oxygen pressure), the aromatic hydrocarbons oxidize and in this form easily break the chain of oxidation of paraffin hydrocarbons.

c. Naphthene hydrocarbons in the oxidation process partially change into aromatic hydrocarbons. Therefore, they can also prevent paraffin hydrocarbons from causing knock, but not as well as aromatic hydrocarbons.

d. The above-stated facts explain why naphthene gasolines with octane ratings of 88 (with 4 milliliters of ethyl fluid per kilogram) blended with isooctane will constitute a knockless fuel for engines operating at overload conditions, while at the same time a fuel consisting of gasoline containing primarily paraffin hydrocarbons and having an octane rating of 95 cannot be improved by adding isooctane so that it will not knock under the same conditions. Low octane naphthene gasoline and isooctane mutually increase each other's antiknock qualities.

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