July 17, 1951 W. A. SCHULZE ET AL 2,560,602

FUEL COMPOSITION

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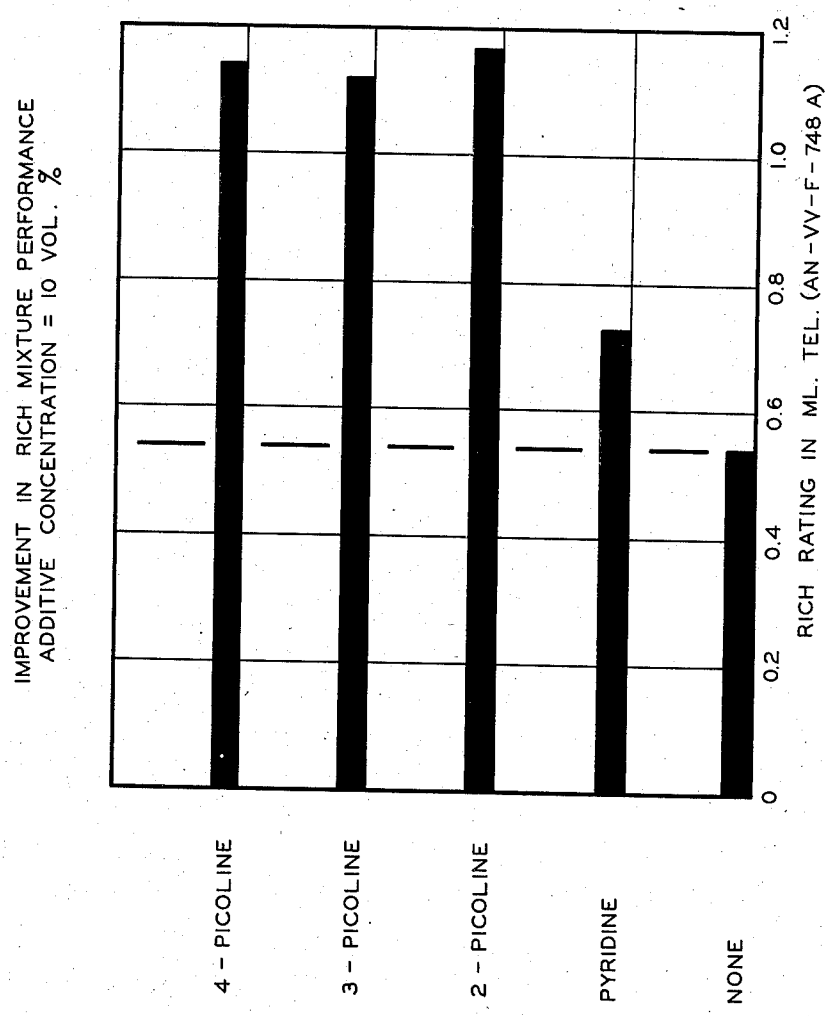



FIG. 1

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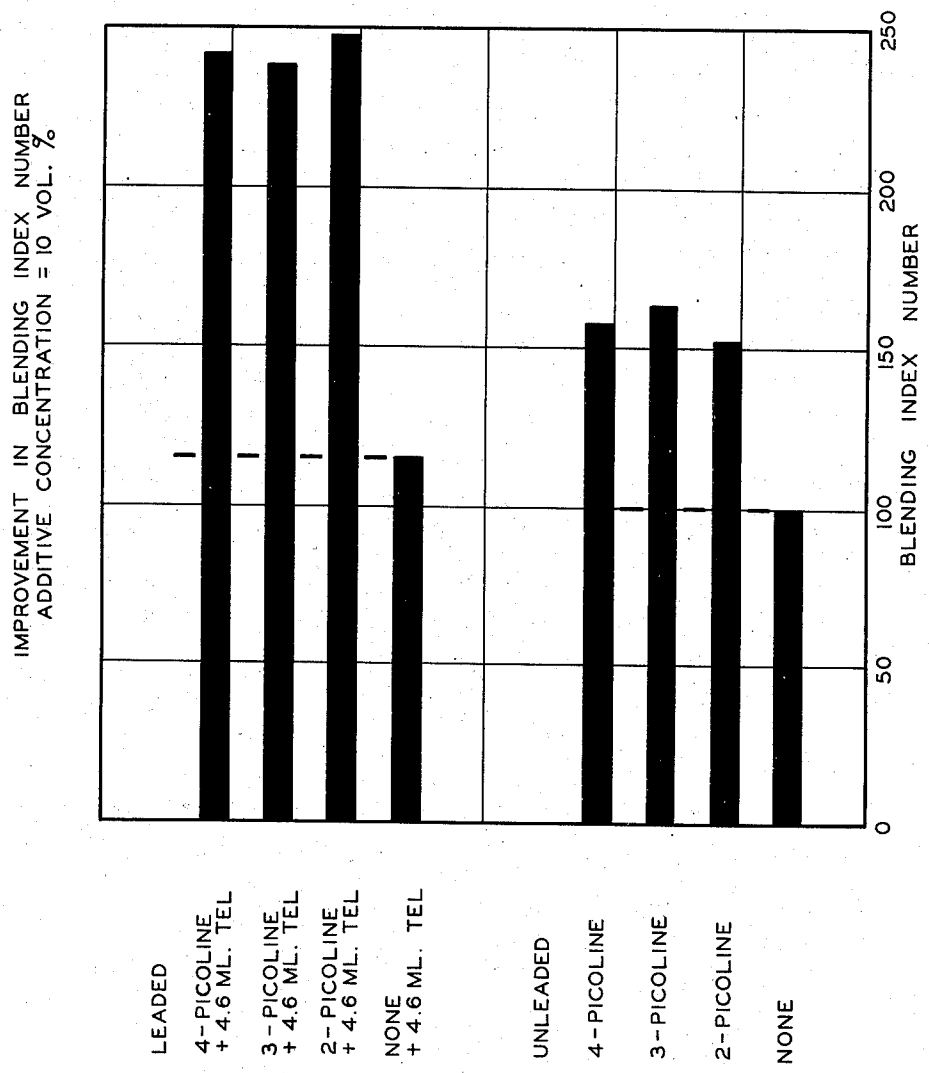



FIG. 2

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# UNITED STATES PATENT OFFICE

2,560,602

## FUEL COMPOSITION

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11 Claims. (Cl. 44—63)

1

This invention relates to an improved motor fuel composition for use in high output aircraft engines. More specifically it relates to an aviation fuel conforming to rigid specifications of antiknock rating, vapor pressure and distillation characteristics, and having, in addition, combustion characteristics which provide superior power output over a broad range of fuel-air ratios and particularly over the range classified as rich mixtures. It relates further to a method of operating an aviation gasoline engine requiring a fuel having an octane number of at least about 100.

High octane number aviation fuels are manufactured and and blended according to specifications so strict that the selection of suitable components is limited in many cases to high purity synthetic blending stocks and naturally occurring base stocks which are segregated with such precision as to approximate the purity of the synthetic hydrocarbons. Such fuels must have high octane number ratings and this in turn means that the components must have high octane number ratings and/or excellent response to the addition of antidetonants. Further, the unsaturation of the components must be exceedingly low in order that the fuels be substantially free of gum and of susceptibility to gum formation. These qualifications together with rigid requirements for vapor pressure, end point, and distillation characteristics, sharply limit the choice of fuel components to the relatively low-boiling, predominantly paraffinic hydrocarbons, say of 5 to 9 carbon atoms, and preferably to the higher octane number branched-chain or isoparaffins.

In the manufacture of aviation fuels of 90 to 100 octane number or of those fuels having antiknock ratings beyond the conventional octane scale, the procedure usually includes the manufacture of synthetic isoparaffins as one blending component. For example, processes, such as selective polymerization, thermal or catalytic alkylation, or the like, may be utilized to prepare concentrates of isooctanes together with usually much smaller amounts of higher and lower homologues. Thermal alkylation may produce such stocks as neohexane, which are highly desirable blending stocks. As a second component, base stocks comprising isohexanes, isoheptanes, isooctanes, etc., may in many instances be prepared by precise fractionation schemes from crude oil and/or natural gasolines.

A third component, which is ordinarily considered separately, is isopentane, which can be prepared in substantially pure form by fractionation of hydrocarbon mixtures containing it. This last named material is ordinarily the lowest boiling stock included in aviation fuels, and it functions as a source of volatility to adjust the vapor pressure of the blend, and to produce desirable distillation characteristics, especially in the initial portion of the distillation curve.

2

Since the synthetic isoparaffin blending stocks are ordinarily available in somewhat smaller volume than base stocks prepared from naturally occurring distillates, blending formulas which require minimum volumes of such synthetic stocks are preferred. Assuming that isopentane is plentiful and used to the maximum extent possible within vapor pressure and front-end volatility limits, it then remains to select and prepare other components commonly termed base-stocks of both high octane number and low vapor pressure. These characteristics are most conveniently obtained by segregation of isoparaffins, such as the isohexanes, etc., as being more valuable than a full boiling range naphtha, unless the naphtha is deficient in normal (low octane number) paraffins or has unusually high octane number as a result of its content of naphthene hydrocarbons.

The net result of the above described blending procedure is the production of fuels of suitable distillation characteristics, vapor pressure, gum stability, and octane number rating comprising largely isoparaffins of 5 to about 8 or 9 carbon atoms, substantially free of $C_4$ hydrocarbons, and preferably containing only minor amounts of $C_5$ to $C_9$ normal paraffins. The content of naphthenes is often small, since these compounds are usually present in only small quantities or in many cases are removed more or less completely by the precise fractionation which separates the normal $C_5$, $C_6$, and $C_7$ paraffins from the isoparaffins in naphtha base stocks.

The complex and strenuous requirements of military aviation under war conditions have emphasized some deficiencies of aviation fuels under certain conditions. Such conditions, for example, are those which require increased or emergency power for improved and rapid take-off, particularly with heavy loads, rapid acceleration and climb during combat. The production of maximum power output under these conditions is of primary importance and the development of fuels which will meet these severe requirements under all conditions is an essential military requirement. It is also obvious that fuels which meet rigid military requirements will be of great value in the development of com-

mercial aviation. The present invention, therefore, provides a novel fuel composition as well as mode of operating aviation engines requiring high octane fuels, particularly of 100 octane or higher. In the practice of this invention, as described herein, a gasoline essentially comprising a mixture of isoparaffin hydrocarbons having 5–9 carbon atoms per molecule, containing TEL, usually at least about 3 ml. per gallon, together with a minor proportion of the additive in question, preferably 1–10 per cent, and a Reid vapor pressure not greater than about seven pounds, is supplied to an aviation engine during an operating period, and particularly when rich mixture performance is required.

The term "rich mixture performance" as now used by the art, and as referred to herein, describes the power output of aviation engines under rich mixture conditions, such, for example, as would be obtained by substantially increasing the fuel concentration in an air-fuel mixture at the intake of an aviation engine. This performance is usually defined in terms related to a standard 100 octane number reference fuel, the relative improvement being stated in ml. of tetraethyl lead.

While laboratory tests and octane ratings by the conventional methods employed for aviation fuels indicate satisfactory performance, more recent test procedures involving fuel performance in supercharged test engines (for example Method AN–VV–F–748*a*) have indicated a deficiency in the power output of said predominantly isoparaffinic fuels. This deficiency has been shown to occur most markedly in rich mixture ratings of the fuel corresponding to take-off or emergency power requirements in aircraft at fuel-air ratios very much higher than the lean mixture ratios ordinarily employed for efficient cruising operation. This discrepancy in the incremental power output with greatly increased fuel-air ratio and fuel consumption has introduced a new consideration into the previous blending formulas to deal with the rich mixture rating of the finished fuels.

Since the predominantly isoparaffinic fuel compositions are very satisfactory from the standpoint of most specifications not involving rich mixture requirements and permit the production of larger volumes of finished fuel per volume of synthetic blending stocks than are possible with other blending formulas, it is ordinarily most advantageous to retain the isoparaffin blending formulas insofar as possible. This procedure requires that such special performance characteristics as lean and rich mixture ratings be obtained through the inclusion of minor proportions of substantially pure chemical additives. These additive compounds must be carefully selected so that the desired improvements are obtained with such small quantities that other fuel characteristics are not impaired and blend specifications are not infringed.

It is, therefore, a principal object of the present invention to provide an improved fuel composition for use in aircraft engines whereby the effective operation and power output of the engines are improved. It is a further object of the present invention to provide an improvement in the blending formula for preparing predominantly isoparaffinic aviation fuels whereby the rich mixture characteristics of said isoparaffinic aviation fuels are greatly improved. It is another object of the present invention to provide an isoparaffinic aviation fuel of 100 octane number rating or better containing a relatively minor proportion of an added compound providing greatly improved rich mixture characteristics without undesirably affecting the other characteristics of the fuel. It is an additional object of the present invention to provide an isoparaffinic aviation fuel having an improved lead response and enhanced blending value. It is a still further object of the present invention to provide an improved method for operating aviation engines under rich mixture conditions by the addition to the isoparaffinic aviation fuel utilized therein of a minor proportion of a picoline. Further objects will be apparent from the herein disclosure.

We have now found that aviation fuels of the type described and comprising essentially isoparaffin hydrocarbons have greatly improved octane number ratings at high fuel-air ratios when minor proportions of alkyl pyridines are added. Particularly useful are the methyl pyridines, or picolines. The picolines are employed as the substantially pure compounds, since their efficiency is highly specific and definitely superior to certain other nitrogen bases and the like which might be present in crude picoline fractions. A further purpose served by the use of substantially pure compounds is the elimination of associated impurities which may have deteriorative effects on the fuel and/or on aircraft fuel systems in which it is used.

This use of the pyridine bases to improve fuel performance at rich mixture conditions is wholly new and unexpected. While their presence in petroleum from certain fields has long been known, they have always been regarded as harmful to fuel performance. The complex and expensive operations to which petroleum is subjected in refining distillates have either deliberately or incidentally removed all traces of nitrogen bases from the resulting gasolines. Thus, petroleum from certain California fields has been shown to contain small amounts of a wide variety of heterocyclic nitrogen bases, including the picolines, and a number of other pyridine derivatives. In cracked distillates from these California oils the pyridine bases are the only group which occurs in appreciable amount and whose presence has been definitely established. Extraction and treating processes to which the cracked distillates are subjected remove these nitrogen compounds and they are not present in the finished products. The highly napthenic gasolines produced from these petroleum stocks are in any event radically different from the synthetic isoparaffin aviation fuels used in our process and the presence of mixed nitrogen bases of many types in impure form therein is in no way equivalent to our process. There has been no suspicion that any desirable properties of improved rich mixture performance reside in the methyl pyridines or indeed anything but undesirable properties.

It has been shown that some nitrogen compounds of the aromatic amine group, represented by aniline as the most common member, do have properties as antidetonants when added to aviation fuels. In this respect, however, they are analogous to the high octane number paraffins, or aromatic compounds such as benzene which have high value as blending agents. The improvement effected in fuel performance when minor amounts of aniline are added is similar to the effect of the addition of tetraethyl lead. That is to say, the octane number rating in the usual test method AN–VV–F–746 shows an im-

provement. But as has been clearly pointed out in the preceding paragraphs this effect is not equivalent to the improvement in rich mixture performance as determined in the supercharged engine test (AN–VV–F–748*a*).

The use of mixtures of the isomeric picolines, or of other alkyl pyridines, is also possible in many instances with very satisfactory results and should not be understood to be excluded by the statements in the earlier paragraphs. Thus, for instance, we have found that 2-methyl, 3-methyl, and 4-methyl pyridines may each be utilized in our process. Mixtures of these in varying proportions will obviously also be useful. Impure preparations which contain other nitrogen bases, and other than nitrogen compounds, where such compounds are detrimental to the rating of the fuel, or the fuel injection system of the engine, or adversely affect other properties of the fuel such as its volatility characteristics, are what is meant to be excluded by the expressions "substantially pure."

While specific embodiments of the present invention may be invoked in a great variety of blending operations involving isoparaffinic blending and base stocks, one satisfactory procedure may be outlined in the following operations. A fuel is to be prepared according to a blending formula, from isooctane, isopentane and a naphtha comprising $C_6$ and $C_7$ isoparaffins in proportions which produce 100 octane number with 4 ml. of tetraethyl lead per gallon. This blending formula is altered according to the present invention to include 2-picoline (or in an exactly similar manner to include 3-picoline, 4-picoline, or other alkyl pyridines) by use of a predetermined volume per cent of a 2-picoline-isopentane mixture having a vapor pressure substantially equal to that of the finished fuel (usually 7 pound Reid vapor pressure). The volume of the mixture used replaces a corresponding volume of isoparaffin base stock and synthetic blending stock with the blending proportions of the latter usually being readjusted to produce the same octane number rating as before.

The picoline may be added alone to fuel blends, but it is often more convenient to employ the isopentanized mixture. The advantages lie in the maximum utilization of isopentane and less difficulty in blending to meet vapor pressure specifications. The amount of picoline thus included in the finished fuel is further limited by the relatively high boiling points (290° F. for 3-picoline, 289.5 for 4-picoline and 264° F. for 2-picoline) which restricts the volume permissible in a fuel having a maximum 90 per cent evaporated temperature as determined by the ASTM distillation procedure, of 275° F.

The picolines or other alkyl pyridines may be derived from any suitable source provided a relatively pure product, as explained above, is obtained. In many cases it is difficult to separate picolines from complex mixtures of alkyl pyridines and other types of nitrogen bases, cresols, etc. such as may result from coal coking operations. For this reason, the preferred source of picolines for the present invention is the synthetic process by which acetaldehyde and ammonia are reacted to produce picolines, as disclosed in copending application Serial No. 567,780 filed December 11, 1944, by one of us and issued as Patent 2,523,580. The picolines which may be produced in high yields are substantially free of undesired impurities and are readily separated from unreacted ammonia and aldehyde.



The amounts of alkyl pyridines added will obviously be dependent on the other fuel components and on the rich mixture octane rating which is desired in the final blend. In most cases the picolines will vary between about one and about 20 volume per cent of the blend, with a somewhat narrower range of about one to about 10 volume per cent usually preferred. The picolines in substantially pure form are relatively expensive, and hence, are not ordinarily used to replace conventional blending ingredients.

The use of certain aromatic compounds especially isopropylbenzene (cumene) and butylbenzene, to improve the rich mixture performance of aviation fuels of the type described in this disclosure has been proposed. In copending applications Serial No. 436,714 and 436,715 filed March 28, 1942, issued as Patents 2,409,156 and 2,409,157, respectively, of which one of us is a coinventor, the use of cumene and of butylbenzene for this purpose is described in detail. While these materials perform in a very satisfactory manner their use is limited by the available supply, and the discovery that the improved performance can be obtained by the use of picolines constitutes a further advance in the art.

It is an advantage of the use of picolines over these aromatic substances when used as components of aviation fuels that they result in much reduced swelling of and diffusion through rubber or rubber-like materials with which the fuel compositions are in contact. Such materials are customarily used as fuel tanks in military aviation, and the notoriously bad effect of aromatics thereon has been a serious detriment to the use of fuels containing them.

Test method AN–VV–F–746 as referred to herein is the method identified as Army-Navy Aeronautical Specification Fuel: Aircraft Engine General Specification (Method for Knock-Test AN–VV–F–746), dated October 5, 1940. This method is utilized for determining ordinary octane number ratings of 100 octane number aviation fuels. Test method AN–VV–F–748*a*, as referred to herein is identified as Army-Navy Aeronautical Specification Fuel; Aircraft Engine, General Specification (Method for Supercharged Knock-Test) AN–VV–F–748*a* dated June 17, 1942. The test method described therein is used for determining lean and rich mixture ratings of 100 octane number aviation fuels. In this latter specification (AN–VV–F–748*a*) a lean mixture is shown to be about 0.06 pound of fuel per pound of air and a rich mixture at least about 0.09 pound of fuel per pound of air.

More recently, however, it has been shown that while this latter method (AN–VV–F–748*a*) can be used to give an excellent measure of the performance of fuels in rich mixtures, lean mixture ratings obtained in this supercharged engine test are erratic and unreliable. At the present time the ratings obtained in the ordinary aircraft engine test (AN–VV–F–746) which is carried out under lean mixture conditions, are considered a more reliable measure of lean mixture performance and the lean mixture ratings obtained in the supercharged test have fallen into disuse.

It has been found that it is generally not possible to add directly the rich mixture ratings of the components of an aviation fuel blend in their volumetric proportions and arrive at a correct value for the rating of the blend, or conversely, from the rating of the blend to determine the value of the additive. This is particularly true where the components differ widely in their rat-

ing, and results in part from the conventions used in expressing rich ratings. While rich ratings superior to the performance of iso-octane are given in terms of the value of tetraethyl lead that must be added to reference fuel S (essentially pure iso-octane) to reproduce the same engine performance, rich ratings inferior to iso-octane are expressed in terms of the volume per cent of fuel S in a blend with reference fuel M (low octane straight run gasoline) to reproduce the engine performance. To reconcile these two methods of expression and the non-linearity of the concentration-performance relationship, the procedure has been adopted of assigning a number known as the "rich mixture blending index" number to each measurable performance level. These numbers are so assigned and tabulated that they may be used directly with the volumetric percentage of the constituents to give the performance rating of the blend. Thus, a fuel of very poor performance, which is equivalent to a blend of 83.0 per cent fuel S in fuel M has a blending index of 4. Pure iso-octane has a blending index number of 100, while a superior fuel whose performance is equal to that of iso-octane with 6.0 cc. of lead added has an index number of 162.

Products which are useful as rich mixture additives should have high index numbers, preferably above 150 when tested in unleaded fuels. It is also important, however, that the additive is not detrimental to the lead response of the fuel, and is particularly advantageous if the effect of lead on the additive is to increase its rich mixture index number, i. e., to make it more effective for the purpose for which it is designed. Thus, benzene is known to be useful as an additive, and has been found to have an index number of 218.0 when tested at 10 per cent concentration in an isoparaffinic fuel blend with 3.0 ml. of TEL per gallon. When this is increased to 4.0 ml./gal. the index number has decreased to 216.6 and at 6.0 ml./gal. to 213.3. Benzene is adversely affected by TEL, and its usefulness as an additive is limited by this inverse lead response.

It is an advantage of the use of our picoline additives that they display an excellent lead response. Thus, when tested in an unleaded fuel, the rich mixture blending index numbers are generally found to be about 150 to 160, but when tested in the same fuel and at the same concentrations, blending index numbers of about 240 are obtained when 4.6 ml. TEL per gallon is present. This excellent response to the addition of tetraethyl lead makes their use in aviation blends particularly advantageous.

As illustrations of the improved performance characteristics obtained by the addition of picolines to isoparaffinic aviation fuels, the following examples are cited:

## EXAMPLE I

A 100 octane number aviation fuel was prepared according to blending formula A, given below, plus 4.6 ml. tetraethyl lead per gallon. The alkylate used was a cut from the product of a commercial hydrogen fluoride alkylation unit in which isobutane is alkylated with butylenes to produce a mixture comprising largely iso-octanes. This formula was then modified to permit the inclusion of ten volume per cent of each of the following additives (1) benzene, (2) 2-picoline, (3) 3-picoline, and (4) 4-picoline. These four modified blends, noted below as B, C, D, and E conformed to fuel specifications and rated approximately 100 octane number with 4.6 ml. of tetraethyl lead per gallon.

*Isoparaffin components (parts by volume)*

| Formula | Alkylate | Isoheptane | Isohexane | Isopentane | Additive, 10 Parts by Vol. |
|---|---|---|---|---|---|
| A | 21.3 | 41.0 | 22.6 | 15.1 | None. |
| B | 21.3 | 41.0 | 22.6 | 15.1 | benzene. |
| C | 21.3 | 41.0 | 22.6 | 15.1 | 2-picoline. |
| D | 21.3 | 41.0 | 22.6 | 15.1 | 3-picoline. |
| E | 21.3 | 41.0 | 22.6 | 15.1 | 4-picoline. |

These blends were rated each with 4.6 ml. tetraethyl lead per gallon Army-Navy test method AN–VV–F–746 and also according to the supercharged engine method AN–VV–F–748*a* with 4.6 ml. tetraethyl lead per gallon. Octane numbers above 100 are recorded as ml. of tetraethyl lead in 100 octane number iso-octane in the following table. The rich mixture index numbers were read from the standard tables corresponding to these ratings, and the blending index numbers calculated for the additive from the known composition of the fuel.

| Blend Formula | Antiknock Rating | | | Calculated-Rich Mixture Blending Index No. of Additive |
|---|---|---|---|---|
| | AN-VV-F-746 | AN-VV-F-748*a* | | |
| | | Rating—ml. TEL | Index No. | |
| A | 100 | 0.54 | 115.2 | |
| B | 100 | 1.01 | 125.2 | 215.0 |
| C | 100 | 1.17 | 128.4 | 247.2 |
| D | 100 | 1.12 | 127.4 | 237.2 |
| E | 100 | 1.14 | 127.8 | 241.2 |

These results indicate the substantially improved rich mixture rating provided by the picolines as compared to fuel A alone and to formula B using benzene. The effect of the heterocyclic nitrogen base additives is very slight in the conventional (F–746) rating, which is also a measure of the lean mixture performance. While a certain small improvement is indicated for benzene, the magnitude of the improvement is in most instances not sufficient to justify the use of this additive. The superior qualities of the picolines are clearly indicated since the rich mixture ratings of fuels C, D, and E are approximately twice that of fuel A when considered in terms of ml. of TEL in iso-octane.

The three picolines, when rated in blends with reference fuel A without addition of lead by the ordinary engine test (AN–VV–F–746) gave blending octane numbers of 93.0, 89.0, and 91.0 for 2, 3, and 4-picoline, respectively. These ratings in themselves gave no indication of the unexpectedly good rich mixture performance of the picolines. Likewise ratings in motor fuel base blends by the usual motor fuel test method (ASTM–D357–43T) gave octane numbers of only about 72 to 83 for these nitrogen compounds.

## EXAMPLE II

The rich mixture ratings of 2-methyl, 3-methyl, and 4-methyl pyridines were determined in blends in an isoparaffinic fuel without the addition of tetraethyl lead, by the supercharged engine test AN–VV–F–748a. The fuel used designated below as fuel F comprised essentially iso-octane, and rated 100 octane number in the conventional aviation engine test procedure (AN–VV–F–746) without lead. Rich mixture blending index numbers

were calculated for the three heterocyclic nitrogen compounds.

The blends were then duplicated in the isoparaffinic aviation fuel of Example I which rated 100 octane number (by method AN–VV–F–746) when 4.6 ml. TEL per gallon were added. Rich mixture blending index numbers were thus obtained in the presence of lead in a fuel of similar type. Results of the ratings obtained by method AN–VV–F–748a in the two series of blends are tabulated below.

| Additive | Volume Per Cent | Ratings by Method AN–VV–F–748a | | | | | |
|---|---|---|---|---|---|---|---|
| | | Unleaded | | | +4.6 ml. TEL/gal. | | |
| | | Fuel | Rating, ml. TEL | Blending Index No. | Fuel | Rating, ml. TEL | Blending Index No. |
| None | | F | 0.0 | 100 | A | 0.54 | 115.2 |
| 2-picoline | 10 | F | 0.17 | 115 | A | 1.17 | 247.2 |
| 3-picoline | 10 | F | 0.21 | 163 | A | 1.12 | 237.2 |
| 4-picoline | 10 | F | 0.19 | 157 | A | 1.14 | 241.2 |

The blending index numbers, which represent the performance of the additive in the fuel blend, show the very markedly greater effect of the additive in the presence of the tetraethyl lead. Thus, while the increase in blending index number is 51 to 63 numbers in the unleaded fuel, addition of the same quantities of picolines to the leaded blend, which in itself had a higher number, produced an increase of 122 to 132 units. Stated in another way, the increased effect of the addition in the presence of lead may be seen in the case of 2-picoline for instance by comparing the blending index number of 151 obtained for this addition in unleaded fuel with the value of 247.2 obtained in the leaded base.

In the accompanying drawings, Figure 1 is a graphical comparison, based on the examples, of the relative improvement in rich mixture performance achieved by the incorporation of minor proportions of the picolines of the present invention as compared with a base fuel having a rich mixture rating of 0.54 ml. TEL and with such a base fuel containing corresponding proportions of pyridine as an additive. As the graph shows, the incorporation of the picolines results in an unexpected improvement in rich mixture performance by comparison with pyridine. The picolines are roughly three times as effective in improving rich mixture performance as pyridine showing that the presence of the alkyl group is of definite and unexpected value in achieving this result.

Figure 2 is a graphical comparison illustrating the effect of the incorporation of the picolines on improving the blending value of the fuel as described in Example II. The improved lead response is strikingly illustrated in the graph which shows that when about 10 per cent of 2-picoline is added to leaded and unleaded fuels, the blending value of the leaded fuel is increased from about 157 to about 265, or about 108 numbers.

We claim:

1. A fuel composition having gasoline characteristics, improved rich mixture performance and lead response which consists essentially of branched chain paraffin hydrocarbons having between five and nine carbon atoms per molecule; and from one to twenty volume per cent of at least one picoline.

2. The fuel composition of claim 1, wherein the picoline content is between one and ten volume per cent.

3. An aviation fuel according to claim 1 wherein the picoline is 2-picoline.

4. An aviation fuel according to claim 1 wherein the picoline is 3-picoline.

5. An aviation fuel according to claim 1 wherein the picoline is 4-picoline.

6. An aviation engine fuel having improved lead response and rich mixture performance when used in aviation engines, which consists essentially of a mixture of isoparaffinic hydrocarbons of five to nine carbon atoms having gasoline characteristics of vapor pressure and distillation range, and from one to twenty per cent by volume of a picoline, said fuel having a Reid vapor pressure not greater than about seven pounds, containing tetraethyl lead, having a maximum 90 per cent ASTM distillation procedure evaporated temperature of 275° F., and having an octane rating of at least 100.

7. An aviation fuel according to claim 6 wherein the picoline is 2-picoline.

8. An aviation fuel according to claim 6 wherein the picoline is 3-picoline.

9. An aviation fuel according to claim 6 wherein the picoline is 4-picoline.

10. A fuel composition having gasoline characteristics, improved rich mixture performance and lead response when used in aviation engines, which consists essentially of a mixture of isoparaffinic hydrocarbons having five, six, seven, eight, and nine carbon atoms per molecule, respectively; and from one to twenty volume per cent of a picoline, said fuel having a maximum Reid vapor pressure of seven pounds and a maximum 90 per cent ASTM distillation procedure evaporated temperature of 275° F.

11. A fuel composition having gasoline characteristics, improved rich mixture performance and lead response which consists essentially of isoparaffinic hydrocarbons having between five and nine carbon atoms per molecule; and from one to twenty volume per cent of a mixture of at least two picolines.

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