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(54) DIESEL OIL PRODUCTION

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SPECIFICATION

TO WHOM IT MAY CONCERN:

Be it known that we, Friedrich Martin, whose permanent address is 38 Uhlenhorstweg, Mülheim-Speldorf, Germany, Otto Roelen, whose permanent address is 30 Bruchsteg, Oberhausen-Holten, Germany, and Paul Schaller, whose permanent address is 57 Wilhelmstrasse, Oberhausen-Sterkrade, Germany, having invented certain new and useful "Process for producing Diesel oils", do hereby declare that the following is a full, clear and exact description of the same:-

Up to the present no successful attempt has been made to replace the gas oils used for driving Diesel engines, by distillation or extraction products of carbonaceous substances such as pit-coal or brown coal, as no one has succeeded in producing the ignition of such substances in a Diesel engine. Only in Diesel engines working under particularly high compression is it possible to make use of the products of the distillation of brown coal or pit-coal tar. However it has been proposed to mix tar oils, such as pit-coal tar oil with gas oil, but here again there has been no satisfactory On the one hand the asphaltic and resinous result. constituents dissolved in the pit-coal tar oil produce in the combustion chamber of the Diesel engine deposits of carbon, and on the other hand the ignition properties of such a mixture of fuels are unsatisfactory. The gas oil added favourably affects the readiness of the mixtures to ignite to such an extent that there is ignition in the Diesel engine, but the delay in ignition peculiar to gas oil, when gas oil is used in admixture with tar oils,

is insufficient, so that the mixture injected into the Diesel engine gives an extremely short burning period. The sharp increase of pressure produced by the high speed of combustion of the mixture is the cause of the considerable knocking when such difficultly ignitable fuels are used in Diesel engines.

It is known that those middle and heavy oils boiling at temperatures lying between 200 - 320°C. which are obtained by the catalytic conversion of carbon monoxide and hydrogen into synthetic hydrocarbons, are suitable for use in Diesel engines. These oils excel by reason of their readiness to ignite, and in their behaviour in the engine cylinder resemble very closely the gas oils used up to the present in Diesel engines. It would therefore be logical to assume that the use in Diesel engines of mixtures of tar oils and the synthetic oil hereinbefore referred to would lead to the same unsatisfactory results.

It has been found that the tar oils, such as pit-coal tar oils, brown coal tar oils, low temperature distillation tars, shale oils, wood tar and the like, which are obtained during the high or low temperature coking of carbonaceous bituminous substances such as pit-coal, brown coal, shale, wood, peat and the like, are so improved by the addition of synthetic oil that by such addition to them a fuel is produced that is suitable for use in Diesel engines. Those constituents of paraffin hydrocarbons obtained by the catalytic hydrogenation of carbon monoxide at ordinary or slightly altered pressure which boil at over 200°C. have proved particularly suitable for this purpose.

By the use of the hydrocarbon oils obtained by

the catalytic hydrogenation of carbon monoxide it is possible, as with the tar oils before referred to, to improve other hydrocarbons poor in hydrogen, such for example as the extraction products of pit coal and brown coal, or also products of a mild hydrogenation - in fact solid hydrocarbons can be treated in this manner even under normal conditions.

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If these substances are insoluble in the paraffinic cils, dissolving agents are also used, the effect of which is to dissolve the solid bodies or to distribute them in a very fine state of division. The paraffinic cils are added to the products poor in hydrogen in such quantity that the resulting mixture is one which will readily ignite in the Diesel engine. The proportions which are suitable depend upon the nature of the substances used, and the best conditions under which the mixing is effected are in every case determined by simple experiment.

whereas hitherto for the purpose of converting an hydrogenation oil into a usable Diesel oil it has been essential to increase by expensive means the hydrogen content to about 12 - 13%, according to the present invention the simple mixing together of the hydrocarbon oils rich in hydrogen and a hydrogenation product with a considerably smaller content of hydrogen produces a very good usable Diesel oil. The possibility of eliminating large additions of hydrogen is of very great practical importance because it is precisely the addition of hydrogen in large quantities - that is to say the carrying out of considerable hydrogenation - that necessitates the use of particular agents such as catalysts, or requires high pressures or considerable

losses of gaseous constituents. When using the products poor in hydrogen which are referred to above, it is necessary however to ensure that the content of hydrogen is at least so high that the added hydrocarbon oils rich in hydrogen such as are obtained by the catalytic hydrogenation of carbon monoxide, do not cause excessive quantities of hydrocarbons to be separated out as a flocculent precipitate from the Occasionally it has proved products poor in hydrogen. to be advantageous to precipitate from the hydrocarbons poor in hydrogen the substances which are capable of precipitation by means of the hydrocarbons rich in hydrogen, with a small quantity of another hydrocarbon rich in hydrogen, such as gas oil. The oil from which the precipitated substances have been removed by filtration gives an extremely good Diesel oil by having mixed with it those conversion products of a mixture of carbon monoxide and hydrogen which are rich in By this means substances such as extracts hydrogen. from coal to which only a little hydrogen has been added can be employed in a very useful manner. insoluble constituents obtained by previous precipitation can if required be converted by a further addition of hydrogen into a substance which likewise gives a very valuable Diesel fuel when mixed with hydrocarbons rich If conditions are suitable, the hydroin hydrogen. carbon oils which have a relatively very low content of hydrogen and which have been separated out from the insoluble substances are not subjected to a further weak hydrogenation until they are mixed with the pro-Hydrogenation can be carried ducts rich in hydrogen. out under particularly simple reaction conditions.

It is of particular advantage to the present process that when the two components are mixed together there is not only an improvement in the quality of the mixture for engines but also a refining of the components poorer in hydrogen. Thus it is already known that the added synthetic paraffin hydrocarbons separate out from the hydrocarbons poor in hydrogen the constituents which in the Diesel engine lead to the deposition of carbon. The separated substances may be very simply removed from the mixture of oils, it being preferable for the refining agent, namely the synthetic paraffin hydrocarbons, to remain in the hydrocarbons poor in hydrogen, and to be burnt in the Diesel engine. Thus the addition of these synthetic paraffin hydrocarbons renders possible the use of the crude tar oils directly instead of being obliged to commence with the much more expensive distillation products.

The mixture of oils together with the substances separated out may be treated first with lye and then with Fuller's earth or alternatively it may be treated with Fuller's earth alone,

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By mixing an oil whose specific gravity is near to 1, with a synthetic hydrocarbon oil which has a specific gravity of approximately 0.75, a mixture is obtained which has a specific gravity of about 0.84 to 0.86 just as have the Diesel oils hitherto used.

The important point is favourably to influence the solidification point. Whereas the tar oils have a very low solidification point and the synthetic paraffin hydrocarbons employed according to the process of the invention have a high solidification point, the Diesel oil produced by mixing together the two components has

a solidification point, normaly for Diesel oils, of for example -20° C.

The process is hereinafter explained by means of examples.

75 parts by weight of crude heating Example 1. oil obtained from pit-coal tar and having a specific gravity of 1.09, which oil will not ignite in a Diesel engine, are intimately mixed at ordinary temperature with 100 parts by weight of a paraffin oil of a specific gravity of 0.76, an initial boiling point of about 200°C., and a solidification point of ± 0°C. obtained from car-On admixture black insoluble bon monoxide and hydrogen. substances and liquid impurities are removed from the The mixture of oils is treated at a pit-coal tar oil. slightly raised temperature first with a 30% solution of caustic soda and then with Fuller's earth. After filtration 170 parts by weight of a clear unvarying Diesel oil having a specific gravity of 0.85 is obtained. This Diesel oil ignites very readily and has an excellent solidification point, which lies at -20°C.

Example 2. 70 parts of brown coal tar boiling at temperatures lying between 200 and 300°C. whose specific gravity is 0.907 and which has a solidification point of -23.5°C., are mixed with 30 parts by weight of a paraffin oil obtained from carbon monoxide and hydrogen whose specific gravity is 0.77 and which boils at temperatures lying between 200 and 310°C. After the removal of the constituents separated out, a Diesel oil is obtained which has a specific gravity of 0.866 and a solidification point of -16.5°C. This oil was found to be very satisfactory when tested in a Diesel engine.

Example 3. 65 parts of a pit-coal distillate

boiling at temperatures lying between 215 and 360°C., having a specific gravity of 1.07 and a solidification point of -29°C., are mixed with 35 parts of the same synthetic oil as used in Example 2. After the removal of the constituents separated out and after treatment of the mixture of oils with caustic soda and Fuller's earth, a Diesel oil is obtained which has a specific gravity of 0.96, and a solidification point of -18°C. and which ignites very readily.

Example 4. 65 parts of a fraction, boiling at between 200 and 320°C. of a tar obtained from pitcoal by low temperature distillation and which has a specific gravity of 0.991 and a solidification point of -27.5°C. is mixed with 35 parts of the same synthetic oil as used in Example 2. After the removal of the constituents separated out by treatment of the mixture of oils with Fuller's earth, a Diesel oil is obtained which has a specific gravity of 0.912 and a solidification point of -20.5°C.

Example 5. 70 parts of a fraction, boiling at between 200 and 320°C. of a dephenolated tar obtained from pit-coal by low temperature distillation, which fraction has a specific gravity of 0.966 and a solidification point lying below -50°C. are mixed with 30 parts of the same synthetic hydrocarbon oil as used in Example 2. After the constituents separated out have been removed, a Diesel oil is obtained which has a specific gravity of 0.707 and a solidification point of -23°C.

Example 6. 70 parts of a hydrogenation oil obtained from brown coal in the first hydrogenation stage, having thus been only incompletely hydrogenated, whose solidification point is 17.5°C. having a specific

gravity of 0.901 and a boiling range of 200 to 320°C. is mixed with 30 parts of the same synthetic oil as used in Example 1. After the removal of the separated-off constituents by treatment of the mixture of oils with Fuller's earth, a Diesel oil is obtained whose specific gravity is 0.861 and which has a solidification point of -15°C. This oil when tested on an engine proved to be excellent Diesel oil which can be stored indefinitely without deterioration.

Example 7. A hydrogenation oil obtained from pit-coal in the first hydrogenation stage, and which has a boiling range of from 200 to 320°C., a specific gravity of 0.974 and a solidification point lying below -40°C. was used as the component poor in hydrogen. The content of acidic constituents of this hydrogenation oil which had been incompletely saturated with hydrogen, amounted to 16.0%. 60 parts of this hydrogenation oil were mixed with 40 parts of the same synthetic oil as used in Example 2. After the removal of the separated-off constituents a Diesel oil was obtained having a specific gravity of 0.895 and a solidification point of -20°C. This mixture likewise can be stored without deterioration.

Example 8. 70 parts of a liquefied extract of pit-coal which has been obtained by mild hydrogenation and which boils at between 200 and 320°C. While having a specific gravity of 1.028 and a solidification point of -43.5°C., are mixed with 30 parts of the same synthetic oil as used in Example 2. When the constituents separated-off are removed a Diesel oil is obtained which has a specific gravity of 0.946 and a solidification point of -22.5°C.

Example 9. 80 litres of a heavy anthracene oil are mixed at ordinary temperature with 20 litres of a gas oil having a content of hydrogen of about 8%. There is at once a heavy precipitation of asphaltic substances, which are filtered off. The filtrate is mixed with the same volume of a mixture of hydrocarbons which boil at between 200 and 310°C., and which have been obtained by the catalytic hydrogenation of carbon monoxide, the product being a mixture which is excellent as a fuel for Diesel engines.

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We claim:

- 1. A process for producing fuels suitable for use in Diesel engines, consisting in effecting at a high temperature the catalytic conversion of a mixture of carbon monoxide and hydrogen to produce paraffinic oils and mixing the said paraffinic oils with carbonaceous products poorer in hydrogen.
- 2. A process according to claim 1, wherein the catalytic conversion is effected under a pressure which falls within a range extending from slightly reduced pressure to slightly elevated pressure by reference to normal pressure.
- 3. A process according to claim 1, wherein the carbonaceous products are liquid.
- 4. A process according to claim 1, wherein the carbonaceous products are solid.
- 5. A process according to claim 1, wherein the carbonaceous products are of the character of tar oils obtained by the dry distillation of carbonaceous material.
- 6. A process according to claim 1, wherein the carbonaceous products are extraction products of carbonaceous materials.
- 7. A process according to claim 1, wherein the carbonaceous products are hydrogenation products of carbonaceous material.
- 8. A process according to claim 1, wherein the paraffinic oils are mixed with the carbonaceous products poorer in hydrogen in such quantity that the resulting fuel has the same specific gravity as the Diesel oils customarily used.
- 9. A process according to claim 1, wherein the carbonaceous products are solid and if insoluble in

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the paraffinic oils are dissolved by the use of dissolving agents.

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- 10. A process according to claim 1, wherein the carbonaceous products are solid and are distributed in the paraffinic oils in a fine state of division.
- 11. A process according to chim 1, wherein the carbonaceous products are used in the form of a hydrogenation product of carbonaceous material that has been hydrogenated at least to such an extent that when added to the paraffinic oils excessive quantities of hydrocarbons are not separated out as a flocculent precipitate.
- adding to the products poor in hydrogen and which have an insufficient hydrogen contents small quantity of a hydrocarbon oil rich in hydrogen such as gas oil for the purpose of precipitating as a flocculent precipitate the substances insoluble in paraffin hydrocarbons, removing the flocculent precipitate from the oil and mixing the oil with the hydrocarbons produced by conversion of carbon monoxide with hydrogen and which are rich in hydrogen.
- adding to the carbonaceous products poor in hydrogen and which have an insufficient hydrogen content a small quantity of a hydrocarbon oil rich in hydrogen such as gas oil for the purpose of precipitating as a flocculent precipitate the substances insoluble in paraffin hydrocarbons, removing the flocculent precipitate and subjecting the freed carbonaceous products poor in hydrogen to a further hydrogenating reaction under mild reaction conditions and mixing the resulting product with the hydrocarbons produced by the conversion of carbon monoxide with hydrogen and which are rich in hydrogen.

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