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64 Method for preparing a synthetic fuel and/or sythetic components for fuels, and the product obtained thereby.

(5) A synthetic fuel and/or synthetic components for fuels are prepared from a mixture of polycarboxylic acids of suitable elementary composition, having a mean molecular weight of between 120 and 800 and an acidity of between 5 and 18 meq/g, by decarboxylating them by heat treatment and subjecting the decarboxylated product to esterification with one or more alcohols or one or more olefins.

This invention relates to a method for preparing a synthetic fuel and/or synthetic components for fuels, and the product obtained by said method.

Various methods are known in the art for preparing synthetic fuels, in particular starting from coal by hydroliquefaction or pyrolysis, or starting from synthesis gases.

However, all the known methods suffer from the drawback of involving high plant and/or operating costs.

Numerous methods exist for oxidising coal with oxygen, oxygencontaining gases or oxidising substances in an aqueous alkaline medium.

A large percentage of the product of this oxidation consists of polycarboxylic acid mixtures.

Examples of methods for producing carboxylic acids by oxidising coal include those described in French patent 1,347,213 and in German patents 841,140, 864,992 and 879,103.

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The polycarboxylic acids obtained by known methods cannot be used as fuels because of their acidic nature, so that their use is practically limited to the chemical field.

It has been surprisingly found that polycarboxylic acid mixtures can be converted into fuels and/or components for fuels by transforming them into oleophilic compounds in a simple and economical manner.

Besides polycarboxylic acid mixtures obtained by oxidising coal, polycarboxylic acid mixtures of any origin can obviously be used for transformation into oleophilic compounds provided that their

elementary compositions, molecular weight and acidity lie within the

limits indicated hereinafter.

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A first subject matter of the present invention is a method for transforming polycarboxylic acid mixtures into oleophilic compounds which can be used as synthetic fuel and/or synthetic components for fuels.

The method for preparing a synthetic fuel and/or synthetic components for fuels according to the present invention starts with a polycarboxylic acid mixture obtained in any manner, and is characterised by the polycarboxylic acids having the following elementary composition:

10 - carbon from 42% to 70% by weight hydrogen from 1.5% to 6% by weight oxygen from 14.0% to 52.7% by weight,

a mean molecular weight of between 120 and 800, and an acidity of between 5 and 18 meq/g, and preferably between 8 and 14 meq/g, the mixture of polycarboxylic acids or their salts being subjected to decarboxylation which in the case of acids is carried out by heat treatment over catalysts constituted by transition metal oxides, and in the case of salts by simple heat treatment, the decarboxylated product being finally subjected to esterification with one or more alcohols or one or more olefins.

The decarboxylation can be effected by heating, in aqueous solution, for a time of between 10 and 30 minutes, preferably 20 minutes, to a temperature of between 300° and 350°C, preferably 315°C, in the presence of one or more alkaline and/or alkaline earth carbonates.

Alternatively, the decarboxylation can be effected in the absence of water at a temperature of between 350° and 550°C, preferably 500°C, for a time of between 10 and 30 minutes, preferably 20 minutes.

The esterification is preferably effected with alcohols comprising 1 to 10 carbon atoms, the alcohols being in H<sub>2</sub>SO<sub>4</sub> solution at a concentration of between 5% and 20% by weight, or with olefins comprising between 2 and 10 carbon atoms.

A second subject matter of the present invention is the synthetic fuel and/or the synthetic components for fuels, when obtained by the aforesaid method.

A third subject matter of the present invention is the fuel obtained by mixing the synthetic fuel and/or synthetic components according to the invention with products chosen from a medium petroleum distillate, a medium distillate from coal hydroliquefaction, the product of coal pyrolysis and particularly the mixture of benzene, toluene and xylenes, a fuel oil of petroleum origin, a mixture of aromatic hydrocarbons or a mixture of C<sub>1</sub>-C<sub>10</sub> aliphatic alcohols.

## **ELAIMS:**

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- 1. A method for preparing a synthetic fuel and/or synthetic components for fuels from a polycarboxylic acid mixture obtained in any manner, characterised by the polycarboxylic acid mixture
- having the following elementary composition: carbon from 42% to 70% by weight hydrogen from 1.5% to 6% by weight oxygen from 14.0% to 52.7% by weight,
- a mean molecular weight of between 120 and 800, and an acidity of

  10 between 5 and 18 meq/g, the mixture of polycarboxylic acids or their

  salts being subjected to decarboxylation, the decarboxylated product

  being finally subjected to esterification with one or more alcohols

  or one or more olefins.
- 2. A method for preparing a synthetic fuel and/or synthetic
  15 components for fuels as claimed in claim 1, characterised in that
  the polycarboxylic acid mixture has an acidity of between 8 and
  14 meg/g.
  - 3. A method for preparing a synthetic fuel and/or synthetic components for fuels as claimed in claim 1, characterised in that the decarboxylation is effected by heating in aqueous solution for a time of between 10 and 30 minutes to a temperature of between 300° and 350°C in the presence of one or more alkaline and/or alkaline earth carbonates.
  - 4. A method as claimed in claim 3, characterised in that the heating is effected to a temperature of 315°C.
    - 5. A method as claimed in claim 3, characterised in that the decarboxylation time is 20 minutes.

- 6. A method for preparing a synthetic fuel and/or synthetic components for fuels as claimed in claim 1, characterised in that the decarboxylation takes place in the absence of water at a temperature of between 350° and 550°C for a time of between 10 and 30 minutes.
- 7. A method as claimed in claim 6, characterised in that the decarboxylation time is 20 minutes.
- 8. A method as claimed in claim 6, characterised in that the decarboxylation temperature is 500°C.
- 9. A method for preparing a synthetic fuel and/or synthetic components for fuels as claimed in claim 1, characterised in that the esterification is effected with alcohols comprising from 1 to 10 carbon atoms, said alcohols being used in H<sub>2</sub>SO<sub>4</sub> solution at a concentration of between 5% and 20% by weight.
- 15 10. A method for preparing a synthetic fuel and/or synthetic components for fuels as claimed in claim 1, characterised in that the esterification is effected with olefins comprising between 2 and 10 carbon atoms.
- 11. A synthetic fuel and/or synthetic component for fuels when20 obtained by the method of the preceding claims.
  - 12. A fuel obtained by mixing synthetic fuel and/or the synthetic components for fuels, when obtained in accordance with claims 1 to 10, with a product chosen from:
  - a) medium petroleum distillate;

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- 25 b) medium distillate from coal hydroliquefaction;
  - c) coal pyrolysis product, and in particular the mixture of benzene, toluene and xylenes;

- d) fuel oil of petroleum origin;
- e) mixture of aromatic hydrocarbons;
- f) mixture of  $C_1-C_{10}$  aliphatic alcohols.