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**JAPANESE PATENTS GAZETTE, vol. 78, no. 33, "Petroleum", page 2, Derwent Publications, London, GB;**

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## Description

This invention relates to the use of a reformed pitch prepared by specific procedures for lowering the softening point of a mesophase containing pitch to 270 °C or less. Since the reformed pitch has the characteristic property that it has a high fixed carbon content in spite of its low softening point and low quinoline insoluble content and that it can be converted easily to mesophase by heat treatment, the reformed pitch is used as a mixing material for modifying various kinds of pitch.

High grade carbon materials such as carbon-carbon composite materials, artificial graphite electrodes, carbon-graphite shaped articles, etc. are generally produced by shaping, carbonizing and graphitizing a mixture of a basic material such as shaped cokes and a binder pitch. In the case of producing a high density, high strength material, it is necessary to repeat pitch impregnation and carbonization process several times before graphitization. The impregnant is indispensable for producing a high grade carbon material, because the pitch impregnation of a carbonized material is effective to bind the basic materials with each other, to decrease a porosity and increase a density, strength, electric conductivity and a thermal conductivity of the produced carbon material.

Pitch based impregnants are generally produced from petroleum or coal based pitch by heat-treating to cause a condensation polymerization reaction and to remove a low boiling point fraction.

Pitch based impregnants are required to have properties as follows for various purposes.

- (1) Low quinoline insoluble (QI) content
- (2) Low softening point
- (3) High fixed carbon content
- (4) High resin content (difference between benzene insoluble content and QI content)
- (5) Low ash content
- (6) Low content of low boiling point fraction

Among these properties low QI, low softening point and high fixed carbon content are very important.

According to the conventional production process of pitch based impregnants, if the softening point were lowered to improve an impregnation efficiency, the fixed carbon content would extremely decrease, therefore, a pitch impregnation and carbonization process would have to be repeated several times. While if the fixed carbon content were raised to decrease the number of repeating times of pitch impregnation and carbonisation, the impregnation efficiency would extremely decrease because of the increase of the QI content and the raise of the softening point, therefore, a solvent extraction process would be necessary to remove quinoline insoluble components.

Some documents disclose the modification of product quality by coexistence of a Lewis acid in converting pitches to mesophase. JP-A-53-7533 discloses a process for producing a mesophase pitch having a softening point of 200-300 °C which comprises directly adding a Lewis acid such as  $AlCl_3$  or the like to a petroleum based tar or pitch having a softening point of 120 °C or lower, subjecting the resulting mixture to heat treatment at a temperature higher than the softening point of the said mixture, usually at 200-300 °C, and after removing the catalyst, subjecting to the second heat treatment at a temperature of 350-500 °C.

Since the flow characteristic of non-mesophase component is close to that of mesophase component in this mesophase pitch, spinnability is excellent even when mesophase content is low and it is said that this mesophase pitch is preferable as a raw material of carbon fibers. However, the temperature of the first heat treatment should be kept high to perfectly melt the solid Lewis acid.

In US-A-4,457,828, there is disclosed a mesophase pitch having molecules of ellipsoidal shape which are produced by the polymerization of aromatic hydrocarbon containing two or more condensed rings. The molecules of this mesophase pitch have been polymerized as 60% or more of bonds which connects condensed rings are coupling (bonding which does not form ring closure) and have a long and slender shape as a whole and considered to be close to ellipsoid.

This polymerization reaction is carried out using a catalyst of a weak Lewis acid such as an anhydrous  $AlCl_3$  accompanied by a second component such as  $CuCl_2$  which has a function of reducing the activity of  $AlCl_3$ . As a solvent, orthodichlorobenzene, nitro benzene and trichlorobenzene are preferable. It is said that the mesophase pitch which is obtained by subjecting a pitch, from which a catalyst has been removed, to heat treatment is preferable for spinning probably due to its slender molecule. Further, this mesophase pitch has a low softening point and good shaping property at a low temperature. It is said that this mesophase pitch has a thin laminate layer of molecule compared with conventional mesophase pitches in spite of high completeness of crystal. Further, it is said that this mesophase pitch has characteristic properties different from the mesophase pitch produced by using a strong Lewis acid which is not accompanied by the second component.

EP-A-0 016 661 describes a process of treating an aromatic carbonaceous pitch with a Lewis acid and a co-solvent at a temperature up to 250 °C and separating the resulting reaction product.

The present invention provides the use of a reformed pitch prepared by mixing a pitch having an aromatic carbon ratio  $f_a$  of more than 0.6 with a Lewis acid and a co-solvent, reacting said mixture, then removing said Lewis acid and the co-solvent from the resulting reaction product, said Lewis acid being  $\text{BF}_3$ ,  $\text{HF} \cdot \text{BF}_3$ , anhydrous  $\text{AlCl}_3$ , anhydrous  $\text{CuCl}_2$ , anhydrous  $\text{ZnCl}_2$  or anhydrous  $\text{SnCl}_2$  and said co-solvent being selected from the group consisting of dichlorobenzene, nitrobenzene and trichlorobenzene, which dissolve both said pitch and said Lewis acid to contact in the liquid state, and the mol ratio of said Lewis acid to said pitch being in the range of 0.3 to 5.0 and the mol ratio of said co-solvent to said pitch being in the range of 2.5 to 50 and the reaction temperature being 100 to 300 °C or by mixing a pitch having an aromatic carbon ratio  $f_a$  of more than 0.6 with a Lewis acid and a co-solvent, reacting said mixture, then removing said Lewis acid and the co-solvent from the resulting reaction product, said Lewis acid being  $\text{HF} \cdot \text{BF}_3$  and said co-solvent being selected from the group consisting of dichlorobenzene, nitrobenzene and trichlorobenzene, which dissolve both said pitch and said Lewis acid to contact in the liquid state, and the mol ratio of said  $\text{HF}$ ,  $\text{BF}_3$  and the co-solvent to said pitch being in the range of 1 to 5, 0.3 to 1 and 2.5 to 50, respectively, and the reaction temperature being 100 to 300 °C, for lowering the softening point of a mesophase containing pitch to 270 °C or less.

The pitch is selected from those having an aromatic carbon ratio  $f_a$  (ratio of carbon forming aromatic rings to the total carbon) of more than 0.6 or more. It is preferable that the pitch is a petroleum or coal based high boiling point fraction, but the use of a low boiling point raw material is also allowable.

There is no particular problem about the use of a raw material having a boiling point, e.g., in the extent of gas oil or kerosene. As a raw material, a single or mixed use of a pure material having a high aromatic carbon ratio, e.g. naphthalene, anthracene, phenanthrene, etc. is possible.

The Lewis acid catalysts used in the present invention are those such as  $\text{BF}_3$ , or  $\text{HF} \cdot \text{BF}_3$ , anhydrous  $\text{AlCl}_3$ , anhydrous  $\text{CuCl}_2$ , anhydrous  $\text{ZnCl}_2$  or anhydrous  $\text{SnCl}_2$ .

From the point of forming velocity of microbeads, anhydrous  $\text{AlCl}_3$  is preferable, but in case where complete removal of a catalyst from a reaction product is necessary, a vaporizable catalyst is preferable. Particularly,  $\text{HF} \cdot \text{BF}_3$  is preferable since  $\text{HF}$  increases the function of the catalyst, and effectiveness as a solvent can be expected and recovery and re-use are easy.

The co-solvents used in the present invention are those compounds which have a boiling point preferably in the range of 100-350 °C and most preferably in the range of 150-250 °C and which do not cause a reaction as decomposition of Lewis acid and are easily separable from the reaction product. The co-solvents are preferably aromatic compounds having a neutral or an acidic substituent and most preferably, compounds in which one or more compounds selected from the group consisting of dichlorobenzene, nitrobenzene, trichlorobenzene are principal components. Even basic compounds such as pyridine, quinoline or the like, which react with the Lewis acid, but does not destroy the structure of the Lewis acid and does not form water by neutralization, are usable because they only weakens the catalytic effect.

When a co-solvent is present various kinds of effect can be obtained.

First of all, since the co-solvent dissolves both a pitch and a Lewis acid, both contact in the liquid state, an efficiency of reaction is increased and uniform reformed pitch can be obtained.

Further since even a pitch having a high softening point such as 250 °C can be used as a raw material if the pitch dissolves in a co-solvent, selection of a raw material can be varied flexibly depending upon the required quality of product. Particularly in case of  $\text{HF} \cdot \text{BF}_3$  without a co-solvent, a raw material having a high softening point cannot be used because of limitative dissolving power of  $\text{HF}$ . Further, as the amount of  $\text{HF}$  becomes greater, the reaction system turns to high pressure and separation and recovery of  $\text{HF}$  becomes difficult, but if a co-solvent is present, the amount of  $\text{HF}$  can be greatly reduced and reaction temperature can be lowered. Namely, even a high softening point pitch can be turned to a liquid state at a lower temperature by dissolving in a co-solvent. In case of  $\text{AlCl}_3$ , since melting point is about 190 °C, if it is to be turned to a complete liquid state, the reaction temperature must be higher than this, but if a co-solvent is present, it is possible to turn to liquid state at a lower temperature.

A mixing ratio of a pitch, a Lewis acid and a co-solvent is preferably in the range of 1:0.3-5:2.5-50. In case of  $\text{HF} \cdot \text{BF}_3$ , the amount of  $\text{HF}$  should be removed in the above mentioned ratio. Reaction temperature is in the range of 100-300 °C, preferably 120-250 °C. Reaction time is preferably in the range of 1-30 hours. In case of  $\text{HF} \cdot \text{BF}_3$ , 0.3-1.0 mol of  $\text{BF}_3$  and 1-5 mol of  $\text{HF}$  are present relative to 1 mol of pitch. The reaction time is preferably 1-5 hours. Even at a reaction temperature of 100 °C a uniform reformed pitch can be obtained. Reduction of the ratio of Lewis acid to less than 0.3 is not preferable because reaction yield is reduced. If the ratio of Lewis acid is more than 5.0, the increase of reaction velocity becomes small

and on the one hand the time necessary to remove Lewis acid from a reaction product becomes longer and this causes increase of cost and hence is not preferable. Reaction temperature lower than 100 °C is not preferable because reaction yield from a pitch is extremely reduced.

Further if reaction temperature is elevated over 300 °C, local rapid reactions tend to occur and the uniformity of the reformed pitch is lowered,

A reaction time less than one hour provides generally low yield from a pitch. On one hand even when reaction is carried out over 30 hours, the reaction yield from the pitch scarcely increases

After the reaction of the pitch in the presence of a co-solvent and a Lewis acid, the co-solvent and the Lewis acid are removed from the reaction system. In case of the solid Lewis acid, removal of the co-solvent is preferably carried out by vacuum distillation. It is preferable to carry out the operation in the inert gas atmosphere. The removal of the Lewis acid is preferably carried out by extraction with an aqueous solvent. Particularly, repetition of washing with a dilute hydrochloric acid is effective. In case of a vaporizable Lewis acid, purging by an inert gas or vacuum distillation is preferable to remove the co-solvent and the Lewis acid from the reaction system, followed by catching thereof. It is preferable to re-use the co-solvent or the Lewis acid.

The properties of the produced reformed pitch fairly depend on the raw pitch. In the case of a pitch having a softening point of about 250 °C, produced reformed pitch has a softening point of about 270 °C, a quinoline insoluble content of about 5 wt % and a fixed carbon content of about 90 wt%. In the case of a pitch having a softening point of about 100 °C, produced reformed pitch has a softening point of about 140 °C, a quinoline insoluble content of less than 1 wt% and a fixed carbon content of about 70 wt%

The reformed pitch is used to modify a conventional petroleum or coal based pitch. When a mesophase containing pitch produced from a pitch having an aromatic carbon ratio  $f_a$  of more than 0.6 is heat treated, the mesophase content can be increased, but at the same time the softening point is also raised to the extent which is unsuitable for spinning to make a carbon fiber. It is found out that when the mesophase containing pitch is mixed with the reformed pitch and the mixture is heat treated, the resulting pitch has a low softening point and a high mesophase content and is useful for spinning to make a carbon fiber because the conversion rate to mesophase is increased while the increase of the softening point is suppressed. Further, when essentially 100% mesophase pitch is mixed with the reformed pitch and the mixture is heat treated, the resulting pitch is a 100% mesophase pitch having a lowered softening point.

Following examples are presented to illustrate the process of the present invention, but they are not intended to limit the scope of the invention.

#### Example 1

A petroleum based pitch (having an initial distillation fraction of 460 °C and a final distillation fraction of 560 °C) produced as a by-product of Fluid catalytic cracking (F.C.C.) of desulfurized vacuum gas oil (DVGO) and having a softening point of 72 °C (Mettler softening point measuring apparatus is used) and a number average molecular weight of 400 was taken in an amount of 200 g and put into glass round bottom flask, 90 g of anhydrous  $AlCl_3$  as a Lewis acid catalyst, 1000 ml of o-dichlorobenzene as a solvent were added and reaction was carried out at a temperature of 180 °C under reflux of a solvent for 26 hours. (Mol ratio of the pitch, Lewis acid and compatible co-solvent were 1 : 1.35 : 17.65).

After completion of the reaction, the solvent was removed by vacuum distillation under nitrogen atmosphere, whereby a solid residual product was obtained. This solid residual product was washed with distilled water and 1N dilute hydrochloric acid and anhydrous  $AlCl_3$  was removed by hydrolysis, whereby a reformed pitch was obtained. This pitch form product was obtained in nearly the same amount as that of raw material pitch before reaction. The softening point of this reformed pitch was 176 °C.

#### Example 2

A petroleum based pitch produced as a by-produce of F.C.C. of DVGO and having a softening point of 130 °C and a number average molecular weight of 500 was reacted in the presence of anhydrous  $AlCl_3$  and o-dichlorobenzene (mole ratios were same as in Example 1) at 180 °C under reflux of a solvent for 26 hours.

After completion of the reaction, anhydrous  $AlCl_3$  and o-dichlorobenzene were removed as in Example 1 and a reformed pitch having a softening point of 195 °C was obtained at a yield of about 100%.

## Example 3

A coal based pitch having a softening point of 86.3 °C (Mettler softening point measuring apparatus is used), toluene insoluble content of 20.9 wt%, quinoline insoluble content of 0.3 wt%, and a mean molecular weight of 450, was taken in an amount of 200 g and put into a glass, round bottom flask (capacity 2000 ml), 90 g of anhydrous  $\text{AlCl}_3$  as a Lewis acid catalyst, and 1000 ml of o-dichlorobenzene as a co-solvent were added and reaction was carried out at 180 °C under reflux of the solvent for 25 hours. (Mol ratio of the pitch, Lewis acid and co-solvent was 1 : 1.52 : 20).

After completion of the reaction, the solvent was removed by vacuum distillation in nitrogen atmosphere and a solid residual product was obtained. This solid residual product was washed with water and 1N dilute hydrochloric acid. The anhydrous  $\text{AlCl}_3$  was removed by hydrolysis and a reformed pitch form product was obtained. The softening point of this reformed pitch was 180 °C.

## Example 4

By using the petroleum based pitch of Example 1 and changing the kinds, ratios of Lewis acid and co-solvent, reactions were carried out. The reaction conditions are shown in Table 1.

Table 1

No.	Reaction condition					
	Lewis acid		co-solvent		reaction	
	kind	mol ratio	kind	mol ratio	temperature (°C)	time hr.
1	$\text{AlCl}_3$	0.5	OCB	3.0	180	26
2	$\text{AlCl}_3$	0.5	OCB	3.0	180	26
3	$\text{AlCl}_3$	0.5	OCB	6.8	180	26
4	$\text{AlCl}_3$	0.5	OCB	6.8	180	26
5	$\text{AlCl}_3$	1.0	OCB	6.8	180	26
6	$\text{AlCl}_3$	1.0	OCB	6.8	180	26
7	$\text{AlCl}_3$	2.0	OCB	13.6	180	26
8	$\text{AlCl}_3$	2.0	OCB	13.6	180	26
9	$\text{AlCl}_3$	0.5	NB	8.1	211	10
10	$\text{AlCl}_3$	0.5	NB	8.1	211	10
11	$\text{AlCl}_3$	0.5	NB	8.0	211	15
12	$\text{AlCl}_3$	0.5	NB	8.1	211	15
13	$\text{AlCl}_3$	0.5	NB	8.1	211	15
14	$\text{AlCl}_3$	0.5	NB	8.1	211	15
15	$\text{CuCl}_2$	1.0	OCB	6.8	180	26
16	$\text{CuCl}_2$	1.0	OCB	6.8	180	26
(symbol) OCB: dichlorobenzene, NB: nitrobenzene						

## Example 5

A petroleum pitch (initial distillation fraction of 460 °C to final distillation fraction of 560 °C), produced as a by-product of fluid catalytic cracking process (F.C.C.) of desulfurized vacuum gas oil, having a softening point of 72 °C (Mettler softening point measuring apparatus was used) and an average molecular weight of 400, in an amount of 0.5 mols was introduced into a 500 ml stainless steel autoclave, 1.25 mols o-

dichlorobenzene was added, after dissolving, the content was cooled till 5 °C. Then, under cooled state 2.5 mols HF was introduced and after replacement of the inside with nitrogen, 0.5 mols BF<sub>3</sub> was blown in. The temperature was elevated at a heating rate of 3 °C/min. and reaction was carried out at 180 °C for 2 hours. After completion of the reaction, cooling was carried out till room temperature. While purging with N<sub>2</sub>, temperature was elevated till 200 °C, o-dichlorobenzene and HF•BF<sub>3</sub> were removed to the outside of the system at the same time, and were caught. A reformed pitch was taken out after removal operation of o-dichlorobenzene and HF•BF<sub>3</sub>. The yield of the reformed pitch was 100%. The softening point of the reformed pitch was 114 °C.

#### Example 6

By using a petroleum based pitch having a softening point of 130 °C (Mettler softening point measuring apparatus was used) and a mean molecular weight of 500, which was a by-product of Fluid catalytic cracking (F.C.C.) of desulfurized vacuum gas oil and changing the ratios of Lewis acid and co-solvent, reactions were carried out. The reaction conditions are shown in Table 2.

Table 2

Reaction condition of pitch by HF BF <sub>3</sub>						
No.	Lewis acid		co-solvent		reaction	
	kind	mol ratio	kind	mol ratio	temperature °C	time hr
1	BF <sub>3</sub>	0.9	OCB	2.5	120	3
	HF	5.0				
2	BF <sub>3</sub>	0.9	OCB	2.5	160	3
	HF	5.0				
3	BF <sub>3</sub>	0.9	OCB	2.5	180	3
	HF	5.0				
4	BF <sub>3</sub>	0.5	OCB	4.0	180	3
	HF	3.0				
5	BF <sub>3</sub>	0.5	OCB	4.0	180	3
	HF	3.0				
(symbol) OCB: dichlorobenzene						

#### Example 7

A petroleum based pitch, as a by-product of Fluid catalytic cracking process (F.C.C.) of desulfurized vacuum gas oil, having a softening point of 130 °C, (Mettler softening point measuring apparatus was used) a number average molecular weight of 500, in an amount of 6 mols was introduced into a stainless steel autoclave. 17.8 mols o-dichlorobenzene was added and after dissolving, the content was cooled to 5 °C. Then, under cooled state, 12 mols HF was introduced. After the inside was replaced by nitrogen, 6 mols BF<sub>3</sub> was blown in. Temperature was elevated at a heating rate of 1.5 °C/min. and reaction was carried out at 160 °C for 3 hours.

While purging with N<sub>2</sub>, temperature was elevated up to 200 °C, o-dichlorobenzene and HF•BF<sub>3</sub> were simultaneously removed to the outside of the system and caught there. After the removing operation of the o-dichlorobenzene and HF•BF<sub>3</sub>, a reformed pitch was taken out. The yield of the reformed pitch was 100%. Resulting reformed pitch had a softening point of 151 °C.

## Example 8

A petroleum based pitch, as a by-product of Fluid catalytic cracking process (F.C.C.) of desulfurized vacuum gas oil, having a softening point of 200 °C, (Mettler softening point measuring apparatus was used) a number average molecular weight of 598, in an amount of 5 mols was introduced into a stainless steel autoclave. 17.8 mols o-dichlorobenzene was added and after dissolving, the content was cooled to 5 °C. Then, under cooled state, 25 mols HF was introduced. After the inside was replaced by nitrogen, 5 mols BF<sub>3</sub> was blown in. Temperature was elevated at a heating rate of 1.5 °C/min. and reaction was carried out at 160 °C for 3 hours.

After completion of the reaction, cooling to a room temperature was carried out. while purging with N<sub>2</sub>, temperature was elevated up to 200 °C, o-dichlorobenzene and HF•BF<sub>3</sub> were simultaneously removed at a reduced pressure to the outside of the system and caught there. After the removing operation of the o-dichlorobenzene and HF•BF<sub>3</sub>, a reformed pitch was taken out. The yield of the reformed pitch was 100%. Resulting reformed pitch had a softening point of 232 °C.

## Example 9

A petroleum based pitch, as a by-product of F.C.C. of DVGO, having a softening point of 72 °C, was heat treated in the nitrogen atmosphere to obtain Pitch A having a mesophase content of 10% and a softening point of 190 °C. The heat treatment was further continued to obtain Pitch B having a mesophase content of 100% and a softening point of 278 °C.

The reformed pitch of Example 7 (a softening point of 151 °C) was blended to the Pitch A at a ratio of 20 wt% relative to Pitch A. The mixture was heat treated at 400 °C for 2 hours, whereby the resulting pitch had a mesophase content of 90% and a softening point of 262 °C.

While, the reformed pitch of Example 7 (a softening point of 151 °C) was blended to the Pitch B at a ratio of 20 wt% relative to Pitch B. The mixture was heat treated at 380 °C for 0.5 hours, whereby the resulting pitch had a mesophase content of 100% and a softening point of 270 °C.

## Function and Effectiveness of the Invention

This invention relates to the use of a reformed pitch as a mixing material for modifying various kinds of pitch.

## Claims

1. Use of a reformed pitch for lowering the softening point of a conventional mesophase containing pitch to 270 °C or less, wherein the reformed pitch is prepared by mixing a pitch having an aromatic carbon ratio fa of more than 0.6 with a Lewis acid and a co-solvent, reacting said mixture, then removing said Lewis acid and the co-solvent from the resulting reaction product, said Lewis acid being BF<sub>3</sub>, HF•BF<sub>3</sub>, anhydrous AlCl<sub>3</sub>, anhydrous CuCl<sub>2</sub>, anhydrous ZnCl<sub>2</sub> or anhydrous SnCl<sub>2</sub> and said co-solvent being selected from the group consisting of dichlorobenzene, nitrobenzene and trichlorobenzene, which dissolve both said pitch and said Lewis acid to contact in the liquid state, and the mol ratio of said Lewis acid to said pitch being in the range of 0.3 to 5.0 and the mol ratio of said co-solvent to said pitch being in the range of 2.5 to 50 and the reaction temperature being 100 to 300 °C.
2. Use as in claim 1, wherein the reformed pitch is prepared by mixing a pitch having an aromatic carbon ratio fa of more than 0.6 with a Lewis acid and a co-solvent, reacting said mixture, then removing said Lewis acid and the co-solvent from the resulting reaction product, said Lewis acid being HF•BF<sub>3</sub> and said co-solvent being selected from the group consisting of dichlorobenzene, nitrobenzene and trichlorobenzene, which dissolve both said pitch and said Lewis acid to contact in the liquid state, and the mol ratio of said HF, BF<sub>3</sub> and the co-solvent to said pitch being in the range of 1 to 5, 0.3 to 1 and 2.5 to 50, respectively, and the reaction temperature being 100 to 300 °C.

## Patentansprüche

1. Verwendung eines reformierten Pechs zur Erniedrigung des Erweichungspunkts eines herkömmlichen Mesophase-enhaltenden Pechs auf 270 °C oder weniger, wobei das reformierte Pech dadurch hergestellt worden ist, daß ein Pech mit einem aromatischen Kohlenstoffverhältnis fa von mehr als 0,6 mit

einer Lewis-Säure und einem Co-Lösungsmittel vermischt worden ist, daß das genannte Gemisch zur Umsetzung gebracht worden ist, daß sodann die Lewis-Säure und das Co-Lösungsmittel aus dem resultierenden Reaktionsprodukt entfernt worden sind, wobei die Lewis-Säure  $\text{BF}_3$ ,  $\text{HF} \cdot \text{BF}_3$ , wasserfreies  $\text{AlCl}_3$ , wasserfreies  $\text{CuCl}_2$ , wasserfreies  $\text{ZnCl}_2$  oder wasserfreies  $\text{SnCl}_2$  ist und das Co-Lösungsmittel aus der Gruppe, bestehend aus Dichlorbenzol, Nitrobenzol und Trichlorbenzol, die sowohl das Pech und die Lewis-Säure zur Kontaktierung im flüssigen Zustand auflösen, ausgewählt ist, und wobei das Molverhältnis der Lewis-Säure zu dem Pech im Bereich von 0,3 bis 5,0 und das Molverhältnis des Co-Lösungsmittels zu dem Pech im Bereich von 2,5 bis 50 liegt und die Reaktionstemperatur 100 bis 300 °C beträgt.

2. Verwendung nach Anspruch 1, dadurch **gekennzeichnet**, daß das reformierte Pech dadurch hergestellt worden ist, daß ein Pech mit einem aromatischen Kohlenstoffverhältnis  $\text{fa}$  von mehr als 0,6 mit einer Lewis-Säure und einem Co-Lösungsmittel vermischt worden ist, daß das genannte Gemisch zur Reaktion gebracht worden ist, daß sodann die Lewis-Säure und das Co-Lösungsmittel aus dem resultierenden Reaktionsprodukt entfernt worden sind, wobei die Lewis-Säure  $\text{HF} \cdot \text{BF}_3$  ist und das Co-Lösungsmittel aus der Gruppe, bestehend aus Dichlorbenzol, Nitrobenzol und Trichlorbenzol, die sowohl das Pech als auch die Lewis-Säure zur Kontaktierung im flüssigen Zustand auflösen, ausgewählt ist, und wobei das Molverhältnis von  $\text{HF}$ ,  $\text{BF}_3$  und dem Co-Lösungsmittel zu dem Pech im Bereich von 1 bis 5, 0,3 bis 1 bzw. 2,5 bis 50 liegt und die Reaktionstemperatur 100 bis 300 °C beträgt.

## Revendications

1. Utilisation d'un brai reformé pour abaisser le point de ramollissement d'un brai contenant une mésophase classique à 270 °C ou moins, dans laquelle le brai reformé est préparé par mélange d'un brai ayant un rapport de carbone aromatique  $\text{fa}$  de plus de 0,6 avec un acide de Lewis et un co-solvant, réaction de ce mélange, puis élimination de cet acide de Lewis et du co-solvant du mélange réactionnel résultant, cet acide de Lewis étant  $\text{BF}_3$ ,  $\text{HF} \cdot \text{BF}_3$ ,  $\text{AlCl}_3$  anhydre,  $\text{CuCl}_2$  anhydre,  $\text{ZnCl}_2$  anhydre ou  $\text{SnCl}_2$  anhydre et ce co-solvant étant choisi dans le groupe consistant en dichlorobenzène, nitrobenzène et trichlorobenzène, qui dissolvent à la fois ce brai et cet acide de Lewis pour les mettre en contact à l'état liquide, et le rapport molaire de cet acide de Lewis à ce brai étant dans la gamme de 0,3 à 5,0 et le rapport molaire de ce co-solvant à ce-brai étant dans la gamme de 2,5 à 50 et la température de réaction étant de 100 à 300 °C.
2. Utilisation suivant la revendication 1, dans laquelle le brai reformé est préparé par mélange d'un brai ayant un rapport de carbone aromatique  $\text{fa}$  de plus de 0,6 avec un acide de Lewis et un co-solvant, réaction de ce mélange, puis élimination de cet acide de Lewis et du co-solvant du mélange réactionnel résultant, cet acide de Lewis étant  $\text{HF} \cdot \text{BF}_3$  et ce co-solvant étant choisi dans le groupe consistant en dichlorobenzène, nitrobenzène et trichlorobenzène, qui dissolvent à la fois ce brai et cet acide de Lewis pour les mettre en contact à l'état liquide, et les rapports molaires de  $\text{HF}$ , de  $\text{BF}_3$  et du co-solvant à ce brai étant dans la gamme de 1 à 5, 0,3 à 1 et 2,5 à 50, respectivement, et la température de réaction étant de 100 à 300 °C.