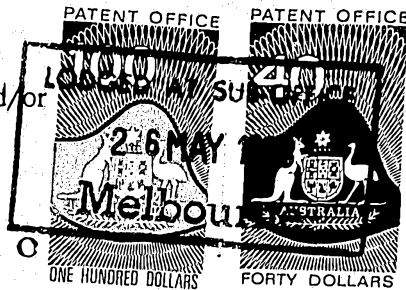


16683/88
(NON-CONVENTION. By one or more persons and/or



Form 1.

COMMONWEALTH OF AUSTRALIA

Patents Act 1952-1969

APPLICATION FOR A PATENT

602408

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26 MAY 1988
Melbourne

(1) Here insert (in full) Name or Names of Applicant or Applicants, followed by Address (es).

X We (1) THE BRITISH PETROLEUM COMPANY P.L.C.
of Britannic House, Moor Lane, London EC2Y 9BU,
United Kingdom

(2) Here insert Title of Invention.

hereby apply for the grant of a Patent for an invention entitled: (2)
PROCESS FOR PREPARING A BINDER BASED ON BITUMEN AND
POLYMERS

APPLICATION ACCEPTED AND AMENDMENTS

ALLOWED 27-7-90

which is described in the accompanying ~~PROVISIONAL~~ COMPLETE specification.

XXV Our address for service is Messrs. Edwd. Waters & Sons, Patent Attorneys,
50 Queen Street, Melbourne, Victoria, Australia.

DATED this 25th day of May 19 88

THE BRITISH PETROLEUM COMPANY

P.L.C.

by

Louis C. Gebhardt

Registered Patent Attorney

(3) Signature (s) of Applicant (s) or Seal of Company and Signatures of its Officers as prescribed by its Articles of Association.

(3)

DECLARATION IN SUPPORT OF AN APPLICATION FOR A PATENT OR PATENT OF ADDITION

(a) Insert full name(s) of applicant(s).

In support of the Application made by (a) THE BRITISH PETROLEUM COMPANY P.L.C.

for a patent for an invention entitled: - (b) Process for preparing a binder based on bitumen and polymers.

(b) Insert title of invention.

(c) Insert full name(s) of declarant(s).

I, (c) Richard David Crack

(d) Insert address(es) of declarant(s).

of (d) the Applicant's address (Britannic House, Moor Lane, London EC2Y 9 BU, United Kingdom

do solemnly and sincerely declare as follows: -

I am the applicant for the patent of addition

(Or in the case of an application by a body corporate.)

(e) Insert name of body corporate.

1. I am authorized by (e) The British Petroleum Company p.l.c.

the applicant for the patent to make this declaration on its behalf.

I am the actual inventor of the invention

(Or where a person other than the inventor is the applicant.)

(f) Insert full name(s) of actual inventor(s).

2. (f) JEAN LESAGE, 31 Rue Georges Sand 95170 Deuil-la-Barre, France

(g) Insert address(es) of actual inventor(s).

of (g)

is the actual inventor of the invention and the facts upon which the Applicant

is entitled to make the application are as follows: - (h)

The Applicant is the assignee of the said actual inventor

(h) Set out how applicant(s) derive(s) title from actual inventor(s), i.e. assignee of the invention from the actual inventor(s).

Declared at London, England this 26th day of May 1988

THE BRITISH PETROLEUM COMPANY P.L.C.

Handwritten signature of R.D. Crack

(Signature of Declarant)

R.D. Crack - by Power of Attorney

To:

The Commissioner of Patents, COMMONWEALTH OF AUSTRALIA

(12) PATENT ABRIDGMENT (11) Document No. AU-B-16683/88
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(54) Title
PROCESS FOR PREPARING A BINDER BASED ON BITUMEN AND POLYMERS

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(56) Prior Art Documents
AU 41096/85 C08L 95/00
AU 50009/79 C08L 95/00
AU 37348/68 C08L 95/00

(57) Claim

i. Process for preparing a bituminous binder which comprises mixing with a bitumen 1 to 35% by weight based on the total weight of the mixture of a composition of an ethylene/vinyl acetate copolymer and a polyisobutylene having a molecular weight of 400 to 1,000, wherein the ratio of polyisobutylene to ethylene/vinyl acetate is from 90:10 to 50:50.

COMPLETE SPECIFICATION

(ORIGINAL)

Class

Int. Class

Application Number:

Lodged:

602408

Complete Specification Lodged:

Accepted:

Published:

Priority:

Related Art:

This document is the
amendment to the original
application and is not for
publication

Name of Applicant: THE BRITISH PETROLEUM COMPANY P.L.C.

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50 QUEEN STREET, MELBOURNE, AUSTRALIA, 3000.

Complete Specification for the invention entitled:

PROCESS FOR PREPARING A BINDER BASED ON BITUMEN AND POLYMERS

The following statement is a full description of this invention, including the best method of performing it known to : : US

Process for preparing a binder based on bitumen and polymers.

The invention relates to a process for preparing compositions based on bitumen and polymers intended, in particular, for use as a road surfacing or an impermeable coating.

It is known that polybutenes such as polybutylene and polyisobutylene are compatible with polyethylenes, polyethylene waxes, polyisobutylenes such as those sold under the name VISTANEX, butyl rubbers, regenerated rubbers, butadiene/styrene copolymers (GRS), and petroleum resins such as diene resins.

The Applicant has discovered that, by combining a polybutene with a copolymer of ethylene and vinyl acetate, and by introducing this mixture, prepared beforehand, into bitumen, it was possible, surprisingly, to boost the consistency and softening properties of the bituminous binder.

The subject of the invention is hence a process for preparing a bitumen/polymer mixture comprising, in a first stage, the preparation of a single additive based on polybutene and an ethylene/vinyl acetate copolymer and the addition of this additive to bitumen. This process is especially advantageous since it suffices simply to mix, for example in a static mixer, the additive with the bitumen, whereas the addition of ethylene/vinyl acetate copolymer or polybutene separately necessitates several hours' mixing using a paddle malaxator, which leads to less homogeneity in the properties of the mixture of bitumen and additive based on polybutene and the ethylene/vinyl acetate copolymer.

Another subject of the invention consists of the additive prepared in the above manner.

The object of the invention is also the production of surfacings or sealings employing the product thereby obtained.

Other subjects of the invention will become apparent

on reading the description and the examples which follow.

The process according to the invention, ~~consists~~ ^{comprises} essentially in adding to a bitumen a mixture, prepared beforehand, comprising an ethylene/vinyl acetate copolymer and a polyisobutylene having a molecular weight of
5 between 400 and 1,000.

The composition comprising the mixture of the ethylene/vinyl acetate copolymer and polyisobutylene constitutes another subject of the invention. It is preferably prepared by the gradual addition of granules of
10 ethylene/vinyl acetate copolymer to a liquid polyisobutylene at a temperature slightly above the ~~temperature of dissolution~~ of the ethylene/vinyl acetate copolymer, by mixing in a slow paddle malaxator or injection by pump
15 and stirring.

The ethylene/vinyl acetate copolymer is preferably a copolymer containing 5 to 45% by weight of vinyl acetate units, and preferably a proportion smaller than 20%, and a melt flow index of between 0.1 and 600, and ~~more~~
20 preferably between 100 and 200. The polyisobutylene preferably has a molecular weight of less than 1,000 and greater than 400. The mixing of polymers is performed in ratios of polyisobutylene to ethylene/vinyl acetate of between 90:10 and 50:50, and preferably of the order
25 of 80:20.

The bitumen is preferably a blown or direct road bitumen, which is well known in the prior art.

The composition based on the mixture of polymers is added to this bitumen, preferably in proportions of
30 1 to 35% with respect to the total weight of the composition, and preferably in proportions from 5 to 30%.

The Applicant has found that it was possible to prepare a bituminous binder directly on site, by mixing the composition based on polymers, as defined above, and
35 bitumen in times of less than 1/4 hour, with, for example, a paddle malaxator, or almost immediately with a static mixer.



The Applicant has found that the addition of the composition containing the mixture of polymers, as defined above, enables a synergistic effect to be obtained as a result of an increase in the softening point and a parallel lowering of the low-temperature breaking point. Thus, as a result of the combination, it is possible to improve the plastic range and the penetration index. This is especially surprising in the light of the properties of the separate polymers.

Another advantage of the invention is that the composition based on the mixture of polymers, designed to be added to bitumen, may be stored without any problem for a long period without degradation and with retention of the initial viscosity.

The road surfacings thus produced behave particularly well in strong heat; the Applicant has, in effect, found that the binder retains sufficient consistency and viscosity to prevent any irreversible deformation due to flow of the surfacing, in particular under the action of heavy traffic.

Moreover, in cold weather, the binder thus applied displays resistance to cracking in the cold, which enables the surfacing to withstand winter stresses such as the use of spiked tyres. The bituminous binders prepared in this manner possess, moreover, exceptional properties of elasticity, tenacity and adhesiveness to the aggregates.

The examples which follow are designed to illustrate the invention without, however, being limiting in nature.

EXAMPLE 1

23 parts of an ethylene/vinyl acetate copolymer, having a vinyl acetate content of 18% and a melt flow index of 140, and 77 parts of a polyisobutylene having a molecular mass of 900 are mixed at a temperature of 140°C in a paddle malaxator, the ethylene/vinyl acetate copolymer being introduced in the form of granules. With 3 kg

of polymers at 300 rpm, fusion of the ethylene/vinyl acetate granules in the polyisobutylene is obtained in less than 1/4 hour.

Keeping this polymer mixture for 15 days at a temperature of 135°C shows that the mixture is completely stable and retains its initial viscosity. No settling is observed.

This mixture is added to road bitumen (40/50) in proportions of 30% of the polymer mixture with respect to the total weight of the composition. The polymer/bitumen mixture is produced in less than 1/4 hour by means of a paddle malaxator.

EXAMPLE 2 (comparative)

2% of ethylene/vinyl acetate copolymer containing 18% of vinyl acetate and having a melt flow index of 140 is added to a grade 40/50 bitumen (penetrability 41, softening point 56, Fraass point -9°C, penetration index +0.46). The softening point is brought to 60.5°C and the penetration index to 0.85 without the Fraass point being modified. The plastic range has risen from 65 to 69.5°C.

The addition of 5% of polyisobutylene of molecular mass 900 to the same bitumen does not modify the softening point, lowers the Fraass point to -16°C and brings the penetration index to +0.5 and the plastic range to 70.5°C.

The addition to the same bitumen of a composition as defined in Example 1, based on ethylene/vinyl acetate copolymer and polyisobutylene and containing 28% of ethylene/vinyl acetate copolymer and 72% of polyisobutylene, this composition being added in the proportion of 7% to the bitumen composition, brings the softening point to 58°C, the Fraass point to -17°C, the penetration index to +1.2 and the plastic range to 75°C.

The addition of the composition hence makes it possible to improve the bituminous binder in respect of all these properties, without increasing its service stresses.



EXAMPLE 3

A formulation is prepared containing 10 parts of polyisobutylene having a molecular mass of 900 and 3 parts of ethylene/vinyl acetate copolymer having a vinyl acetate content of 18% and a melt flow index of 140, and this is introduced into 87 parts of 40/50 bitumen as defined above.

The following viscosities are observed:

at 140°C: 722 mPa.s; at 160°C: 297 mPa.s; at 180°C: 142 mPa.s; whereas the base bitumen has the following viscosities:

at 140°C: 600 mPa.s; at 160°C: 263 mPa.s; at 180°C: 142 mPa.s.

It is found that the combination according to the invention enables the viscosity when hot to be increased slightly, and also improves the ease of handling of the mix. This is observed in the Laboratoire des Ponts de Chaussées rotary shearing press test, on a discontinuous 0/14 mix of formula:

10/14 porphyry: 26%

6/10 porphyry: 31%

0/2 porphyry: 39%

calcareous fines: 4%

binder content: 7.7 pph (parts by weight per hundred parts of aggregates).

The 0/14 mix with the polymer binder according to the invention achieves

89.5 compactness at 10 rotations

92.0 compactness at 20 rotations

95.0 compactness at 50 rotations.

The binder prepared in this manner possesses, moreover, properties of resistance in the fatigue test and of resistance to stripping by water or adhesiveness which are also improved.

A fatigue test is performed, according to the LCPC (Laboratoire Central des Ponts et Chaussées) method in sinusoidal bending to constant deformation of trapezoid

test pieces, the test being performed using plates of mixes manufactured and compacted according to the Laboratoire des Ponts et Chaussées procedures, on the mixes of the following formula:

5 6/10 La Noubleau 33%
 2/6.3 La Noubleau 24%
 0/2 La Noubleau 40.7%
 calcareous filler: 2.3%
 binder 5.75 pph.

10 The following figures are obtained at a frequency of 25 hertz and at 10°C:

 at 10⁶ cycles, the relative deformation is:

 1.40 x 10⁻⁴ for the reference mix (40/50 binder),
 but 2.20 x 10⁻⁴ for the mix with the bitumen/polymer
15 binder defined above.

 at 10⁵ cycles, the relative deformation is:

 2 x 10⁻⁴ for the reference mix; and

 3 x 10⁻⁴ for the mix with the bitumen/polymer
binder.

20 It is hence observed that there is a larger permissible deformation for the same fatigue test time for the mix containing the polymer binder according to the invention. In the water stripping test, this property is demonstrated by the ratio of the resistances at 18°C in the DURIEZ (or LCPC) unconfined compression test, with retention on immersion in water for 7 days with respect to the concentration in the dry state also for 7 days.

 Taking the following mix formula:

 10/14 La Noubleau 69%

30 0/2 La Noubleau 27.5%

 calcareous filler: 3.5%

 binder content 5.5 pph

 the ratio of the resistances is 0.88 for the 40/50 reference mix, compared with 0.96 for the mix with the bitumen/polymer binder as defined above.

35

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:

1. Process for preparing a bituminous binder which comprises mixing with a bitumen 1 to 35% by weight based on the total weight of the mixture of a composition of an ethylene/vinyl acetate copolymer and a polyisobutylene having a molecular weight of 400 to 1,000, wherein the ratio of polyisobutylene to ethylene/vinyl acetate is from 90:10 to 50:50.

2. Process according to claim 1 in which the ethylene/vinyl acetate copolymer is a copolymer containing 5 to 45% by weight of vinyl acetate units and having a melt flow index of 0.1 to 600.

3. Process according to claim 1 or 2 which is carried out at the site of use, the mixing time not exceeding 1/4 hour.

4. Process according to claim 3 in which, after mixing is complete, the resulting mixture is applied as a road surfacing.

5. Process for preparing a composition of an ethylene/vinyl acetate copolymer and a polyisobutylene having a molecular weight of 400 to 1,000, wherein the ratio of polyisobutylene to ethylene vinyl acetate is from 90:10 to 50:50 which comprises gradually adding granules of the ethylene/vinyl acetate copolymer to the polyisobutylene at a temperature slightly above the melting point of the ethylene/vinyl acetate copolymer in the liquid polyisobutylene and mixing.



6. Process according to claim 5 substantially as hereinbefore described.

7. A composition whenever prepared by a process as claimed in claim 5.

8. Process according to claim 1 or 2 which comprises mixing with a bitumen 1 to 35% by weight based on the total weight of the mixture of a composition according to claim 7.

9. Process according to claim 1 substantially as described in Example 1 or 3.

10. A bituminous binder whenever prepared by a process as claimed in any one of claims 1 to 4, 8 and 9.

DATED this 4th day of July, 1990.

THE BRITISH PETROLEUM COMPANY Plc

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