

The invention relates to a process for the production of a bituminous binder modified with synthetic thermoplastic material for construction materials, in particular road building material, wherein for the formation of the binder, molten bitumen together with a thermoplast or with a mixture of thermoplasts, preferably with an olefin polymer, is subjected to a homogenization treatment under the influence of mechanical shearing forces, wherein a disintegration of molecules takes place in the thermoplastic material.

A number of processes of the above-indicated type are known, by which bituminous binders can be produced, whose properties have been improved relative to ordinary bitumen by the addition of the thermoplast. Often such a modification of bitumen requires a relatively great labor expenditure because a number of different thermoplasts whose addition to bitumen can considerably improve the properties of the latter, have a very poor solubility in bitumen; other thermoplasts which might be considered for the modification of bitumen and which are more readily soluble in bitumen, often are expensive to produce, and construction materials produced with such a binder often are not as strong as construction materials whose binder is a bitumen admixed with thermoplasts less readily soluble in bitumen. In various processes a highly heated mixture of bitumen and a thermoplast was mixed more or less thoroughly, yet this had the disadvantage that the entire energy supplied (thermal and mechanic) could not be dosed correctly, thus resulting either in end products having poor physical properties, if the energy supplied was too little, or in unnecessary energy expenditures or too pronounced decomposition, particularly of the bitumen, and also to low quality products, if the energy supplied was too much.

In a process known from AT-A-370 126, a highly heated mixture of bitumen and thermoplastic material is subjected to a forced mixing procedure exerting high shearing forces on that mixture for homogenization thereof, mixing means being used for this forced mixing procedure, in which the mixture is relatively slowly guided through a gap formed between mixing tools moved relative to each other, in which it is subjected to high shearing forces; in particular, mixing pumps, i.e. screw-type

pumps, spiral pumps or gear pumps, or kneading means are considered as the mixing means.

It is an object of the present invention to provide a process of the initially defined kind by which not only the homogenization procedure can be effected rapidly when using thermoplasts usually poorly soluble in bitumen, such as e.g., polyethylene, but which also yields a bituminous binder having properties clearly better than those of bituminous binders of the same or a similar composition produced by known processes.

The process according to the invention of the initially defined kind is characterised in that at the homogenization treatment, to the mixture containing the bitumen and a thermoplast or a mixture of thermoplasts, an amount of energy in the form of kinetic energy is supplied sufficient to cause the disintegration of molecules of the thermoplastic material, by imparting to this mixture a velocity corresponding to this amount of energy and then abruptly braking the mixture on an impact surface, wherein processing of said mixture is continued until a change of structure of the mixture is noticeable, e.g. by a change of the X-ray spectrum lines.

By this mode of procedure the above-indicated aim may be met well, and thus it is possible to produce binders on the basis of bitumen and modifying thermoplasts, even if the latter as such are poorly soluble in bitumen, within a relatively short period of time, and furthermore, on account of this modification, to achieve a substantial improvement of the binder properties as manifested e.g. in the adhesion of the binder to rock material and in the strength of the construction materials produced with such a binder.

By bringing the mixture comprised of bitumen and thermoplastic material to a high velocity, kinetic energy is supplied thereto. By subsequently abruptly braking this mixture on an impact surface, the kinetic energy previously supplied to this mixture is converted in a very short span of time, and thereby the molecules of the thermoplastic material are disintegrated under formation of highly reactive molecule fragments without the disintegration causing a detrimental material-damaging disintegration of the synthetic thermoplastic material; the molecules of the bitumen whose molecular mass is

much smaller than that of the molecules of the thermoplastic material, are hardly disintegrated by the abrupt braking of the mixture on the impact surface. Thus an effect is achieved which equals the effect of a very short intensive heating of merely the synthetic thermoplastic material present in the mixture, during which no detrimental disintegration occurs because of the very short duration of heating. With the method according to the invention, the energy supplied is utilized in a differentiated manner, being aimed at the desired controlled disintegration of the thermoplastic material contained in this mixture, while treating the material, in particular the bitumen used, as gently as possible.

The disintegration of the molecules of the synthetic material under formation of particularly reactive fragments may be promoted by the action of high centrifugal forces on the mixture of bitumen and synthetic thermoplastic material to be processed to a binder. To this end it is suitable to impart to the mixture as high a circulation speed as possible and to brake the velocity of this mixture within a short span of time on the impact surface.

A preferred embodiment of the process according to the invention consists in that the mixture containing the bitumen and a thermoplast or a mixture of thermoplasts is forced through a narrow gap between two oppositely arranged mixing tools rotating relative to each other, the gap on an average having a width of less than 3 mm, preferably a width of less than 0.3 mm, and the abrupt braking of the mixture is effected in the gap region, preferably immediately after emergence of the mixture from the gap. The mixing tools rotating relative to each other are preferably arranged coaxially to each other. Advantageously, processing of the mixture may frequently be interrupted after each gap passage by pauses.

The statements made above in respect of the width of the gap between the oppositely arranged mixing tools rotating relative to each other are to be considered as average values. The gap may be interrupted by recesses, grooves or the like; the larger distance provided on the site of such recesses or grooves is not taken into consideration when calculating the average value of the gap width.

If the molten mixture of bitumen and synthetic thermoplastic material is forced through a narrow gap, preferably a gap having a width of less than 0.3 mm, between two mixing tools rotating relative to each other, in addition to the thermal influence, there results a particular wear of the mixture passing the gap by the kinetic energy being transformed at the impact on the impact surface, this wear particularly acting on the thermoplast component which has a larger molecular mass and a higher molecular weight, respectively, than bitumen; the disintegration of the molecules present in the mixture and occurring under these stresses, resulting in shorter molecule fragments (i.e. molecule fragments having a smaller molecular mass or a lower molecular weight, respectively,) which have a pronounced tendency to combine with each other. While when stirring such a mixture, e.g. a mixture of bitumen and polyethylene, at common intensities, during which this mixture or the substances contained therein, respectively, are subjected to combined thermal and mechanical stresses, after termination of the treatment merely a weakening or a complete disappearance of the spectral lines of the X-ray spectrum characteristic of the crystal structure of the synthetic material portion can be observed, if the treatment has been continued sufficiently long at the disintegration temperature of the constituent(s) subjected to molecular disintegration, the examination of a mixture forced, according to the process of the invention, through a narrow gap between rotating mixing tools, clearly shows X-ray lines indicating the presence of a new crystal structure. Corresponding to this occurrence of X-ray lines characteristic of a crystal structure, construction materials produced with a binder obtained in a manner according to the invention clearly exhibit better mechanical properties than construction materials formed with binders in which bitumen and a synthetic thermoplastic material have been mixed in the molten state merely by intensive stirring. A further advantage of the mode of procedure of the invention mentioned before in which a mixture of molten bitumen and synthetic thermoplastic material is forced through a narrow gap between rotating mixing tools is that in this manner the production of the bituminous binder modified by synthetic thermoplastic material can be effected in

a much shorter period of time than with the known intensive stirring of such a mixture. Moreover, and this is a substantial advantage, it is possible to proceed at a much lower mean temperature or treatment temperature of the mixture of synthetic thermoplastic material and bitumen to be treated than with the known homogenization of such a mixture by intensive stirring, and it is obvious that this is made possible by the fact that the molecules of the synthetic material experience a mechanical stretching when the mixture is forced through the narrow gap, on the one hand, and that the molecules of the synthetic thermoplastic material at the conversion of the kinetic energy supplied to the mixture are partitioned at the impact surface into highly reactive fragments. Such fragments unite later on to form at least a partially crystalline structure. This also appears from the change of the X-ray lines as compared to the state prevailing before the mixture has been forced through the gap. By being formed in surrounding bitumen, this at least partially crystalline structure also becomes tightly interlaced with the bitumen.

The tight interlacing of the thermoplast with the bitumen is also visible from the behaviour of a binder produced according to the invention, during an extended hot storage. A mixture of bitumen and thermoplast treated according to the process of the invention immediately upon its production is visually homogenous when illuminated with light of the visible wave length, and when illuminated with ultraviolet light, it exhibits a dense pattern of light spots on a darker base; after hot storage for many hours at a temperature clearly above the melting temperature of the thermoplast, the particles previously visible as spots unite to form a light cover layer whose volume is about four to eight times the volume of the thermoplast portion of the mixture; by a simple, short-time stirring this cover layer can again be united with the remaining portion of the binder into a visually homogenous substance. This behaviour differs clearly from the behaviour of a homogenized product obtained in a known manner by extended stirring of a molten mixture of bitumen and thermoplast; when hot-storing such a product for several hours, a light cover layer forms whose volume approximately corresponds to the volume of the thermoplast portion of the mixture, and for

a renewed homogenization of the cover layer with the main portion of the material from which the cover layer has separated during hot storage, practically the same stirring work is required as has been applied during the first homogenizing stirring.

The two mixing tools between which the gap is present through which the mixture is forced to form the binder shall rotate relative to each other. This may simply be realized by one mixing tool standing still and the other one rotating. Yet also both mixing tools may rotate, either in opposite rotational directions or in equal rotational directions, yet at different rotational speeds.

Within the scope of the process according to the invention it is particularly suitable if the mixture containing bitumen and a synthetic thermoplastic material is forced through a gap present between mixing tools coaxially rotating relative to each other, which gap on an average has a width of approximately 0.1 mm or less. By further developing this measure preferably provided in the process according to the invention, of treating the mixture to be homogenized in a very narrow gap, the disintegrating effect caused according to the process of the invention in the molecules of the synthetic material, which leads to the formation of highly reactive molecule fragments having a particular tendency to re-crystallize, may further be intensified, it being possible to obtain this effect with decreasing gap widths at ever lower mean temperatures of the mixture. It has furthermore proved to be advantageous to form the gap such that in the gap, the mixture is subjected to a pressure built up by hydrodynamic action.

For the above-mentioned embodiment of the process according to the invention in which the mixture is forced through a gap it is advantageous that the mixture is forced through a gap extending radially at least at its rim of emergence, from which it emerges in the radial direction. A particularly good effect can be obtained if the abrupt braking of the mixture is carried out in the gap region, preferably immediately after the emergence of the mixture from the gap, since thus the high velocity imparted to the molecules of the synthetic material by the centrifugal force and the kinetic energy associated

therewith is particularly well utilized for partitioning the synthetic molecules into reactive fragments.

As regards the velocity which is imparted to the mixture before it is abruptly braked on an impact surface, a value of approximately 30 m/s has proved to be suitable for various applications.

The invention will now be further explained by way of examples and with supplementary reference to the drawings, wherein

Fig. 1 are X-ray spectra of various substances, and

Fig. 2 are sectional views of bodies made up of various substances.

E x a m p l e 1 : A molten mixture comprised of 94 % by mass of bitumen B 100 and 6 % by mass of high-density polyethylene at first was stirred slightly in a storage container, and this mixture was supplied from the storage container to a throughput mixing device which had two disc-shaped oppositely arranged mixing tools, one mixing tool standing still and the other mixing tool being rotatably mounted coaxial to the first one and connected with a drive. Both, the mixing tool standing still and the rotating mixing tool were provided with ribs at the sides facing each other. At the sides of the two mixing tools facing each other a gap of approximately 0.1 mm was present between the ribs of the one mixing tool and the ribs of the other mixing tool, through which gap the mixture to be treated was forced. The peripheral velocity of the rotating mixing tool at the outer rim of the gap was approximately 30 m/s. Likewise, the flow velocity of the mixture through the gap was approximately 30 m/s. In the gap region, impact surfaces were provided which were hit by the mixture, the latter being abruptly braked thereby. The mixture was fed six times through the throughput-mixing device. This required approximately 20 minutes. Then the bituminous binder thus obtained was mixed with pre-heated rock material and mineral powder filler of standardized composition for asphalt concrete 08, and from the construction material thus obtained, Marshall test bodies were produced in a standardized procedure, which then were examined for their mechanical properties. The measured values obtained are listed in column 1 of the table.

E x a m p l e 2 : It was proceeded analogous to Example 1, yet the mixture intended for the production of the binder being composed of 94 % by mass of bitumen B 100 and 6 % by mass of low density polyethylene. The measured values obtained are listed in column 2 of the table.

E x a m p l e 3 : It was proceeded analogous to Example 1, yet the mixture intended for the production of the binder being composed of 94 % by mass of bitumen B 100 and 6 % by mass of ethylene-propylene-diene copolymer. The measured values obtained are listed in column 3 of the table.

E x a m p l e 4 : It was proceeded analogous to Example 1, yet the mixture intended for the production of the binder being composed of 94 % by mass of bitumen B 100 and 6 % by mass of styrene-butadiene-styrene block polymer. The measured values obtained are listed in column 4 of the table.

E x a m p l e 5 : It was proceeded analogous to Example 1, yet the mixture intended for the production of the binder being composed of 94 % by mass of bitumen B 100 and 2 % by mass of ethylene-propylene-diene copolymer and 4 % by mass of low density polyethylene. The measured values obtained are listed in column 5 of the table.

E x a m p l e 6 : It was proceeded analogous to Example 1, yet the mixture intended for the production of the binder being composed of 94 % by mass of bitumen B 100 and 2 % by mass of ethylene-propylene-diene copolymer and 4 % by mass of high density polyethylene. The measured values obtained are listed in column 6 of the table.

E x a m p l e 7 : For a comparison, the properties of the bitumen used in Examples 1 to 6 and of a construction material produced analogously therewith were examined. The measured values obtained are listed in column 7 of the table.

E x a m p l e 8 : The finished binder obtained according to Example 1 in the cooled-off state was subjected to an X-ray spectral examination. The spectrum denoted by a in Fig. 1 of the drawings was obtained. Furthermore, a visually homogenous mixture obtained merely by stirring and having the same composition as the mixture according to Example 1 in the cooled-off state was subjected to the same X-ray spectral examination, the spectrum denoted by b in Fig. 1 being obtained, and also the

polyethylene used in Example 1, in the cold state, was subjected to this X-ray spectral examination, the spectrum denoted by c in Fig. 1 being obtained. From these X-ray spectra it is apparent that the strong spectral lines present in spectrum c and caused by the crystalline structure of the polyethylene do no longer appear in spectrum b. Hence follows that that mixture of bitumen and polyethylene mixed merely by stirring, also in the cooled-off state practically exhibits no crystalline structure. Spectrum a shows spectral lines which correspond to the strong spectral lines of spectrum c caused by the crystalline structure of the polyethylene. Hence follows that the binder produced according to the process of the invention according to Example 1 in the cooled-off state at least partially exhibits a crystalline structure formed by the molecule fragments of the polyethylene.

E x a m p l e 9 : A sample of a binder produced according to Example 2 of 94 % by mass of bitumen B.100 and 6 % by mass of low-density polyethylene according to the process of the invention was allowed to solidify in a cup, whereupon the body thus obtained was cleaved vertically. When irradiated with visible light, the cleavage surface looked homogenous; when irradiated with ultraviolet light, a dense pattern of uniformly distributed light spots was visible on a darker base. A view of this cleavage surface is indicated by a in Fig. 2. A further sample of this binder was stored in a cup, at 160°C for 24 hours, and then was allowed to cool in this cup, whereupon the body thus formed was cleaved. When irradiating with ultraviolet light, a light layer could be recognized which approximately covered that third of this body that lay on top during the previous storage, while the two thirds of the body that lay therebelow had a dark colour. A view of the cleavage surface of this body is denoted by b in Fig. 2. Thereupon, this body was melted again, and this melt was stirred at approximately 170°C, the melt attaining a completely homogenous appearance after a few minutes.

E x a m p l e 10 : A mixture was formed of the substances also used in Example 2 with a composition of 94 % by mass of bitumen B 100 and 6 % by mass of low-density polyethylene. This mixture was stirred for 1 1/2 hours at 280°C to form a

homogenous product. Then a sample of this product was stored in a cup, at 160°C for 24 hours, and subsequently was allowed to cool in this cup, whereupon the body thus formed was cleaved. When irradiated with ultraviolet light, a light layer could be recognized whose volume approximately corresponded to the volume of the polyethylene portion of the product and was located at the side which had been on top during the previous storage. A view of the cleavage surface of this body is indicated by c in Fig. 2. This body was then melted again, and the melt thus obtained was stirred again at approximately 280°C. This stirring had to be continued for more than 1 hour to attain a completely homogenous appearance of the melt.

EXAMPLE	7	1	2	3	4	5	6
	B 100 100%	B 100 94% HDPE 6 %	B 100 94% LDPE 6 %	B 100 94% EPDM 6 %	B 100 94% SBS 6 %	B 100 94% LDPE 4 % EPDM 2 %	B 100 94% HDPE 4 % EPDM 2 %
Softening point ring and ball °C	48.0	57.0	54.0	86.0	72.0	66.0	74.0
Penetration at 25°C 1/10 mm	101.0	75.0	61.0	47.0	82.0	40.3	32.3
Brittle point °C	-9	-10	-10	-21	-15	-15	-14
Binder content M%	5.2	5.2	5.2	5.2	5.2	5.2	5.2
Crude density g/cm ³	2.60	2.60	2.60	2.60	2.58	2.58	2.59
Volumetric density g/cm ³	2.55	2.54	2.52	2.53	2.54	2.53	2.54
Void content vol%	1.9	2.3	3.1	2.7	1.6	1.8	1.9
Stability "T" KN	11.0	12.4	15.4	13.9	13.1	15.3	14.5
Flow value "F" mm	5.07	4.47	2.93	4.47	5.00	4.50	4.57
Rigidity T/F KN/mm	2.1	2.7	5.2	3.1	2.6	3.4	3.1
Crushing strength/25°C/N/mm ²	0.81	1.04	1.43	1.27	0.78	1.41	1.39
Crushing strength/40°C/N/mm ²	0.18	0.31	0.48	0.45	0.28	0.47	0.46
E-modulus /25°C N/mm ²	461	708	1197	909	461	1104	1096
E-modulus /40°C N/mm ²	110	202	456	327	189	320	304
Density overall g/cm ³	1.016	1.013	1.017	1.018	1.022	1.029	1.025

TABLE



Claims:

1. A process for the production of a bituminous binder modified with synthetic thermoplastic material for construction materials, in particular road building material, wherein for the formation of the binder, molten bitumen together with a thermoplast or with a mixture of thermoplasts, preferably with an olefin polymer, is subjected to a homogenization treatment under the influence of mechanical shearing forces, wherein a disintegration of molecules takes place in the thermoplast, characterised in that at the homogenization treatment, to the mixture containing the bitumen and a thermoplast or a mixture of thermoplasts an amount of energy in the form of kinetic energy is supplied sufficient to cause the disintegration of molecules of the thermoplastic material, by imparting to this mixture a velocity corresponding to this amount of energy and then abruptly braking the mixture on an impact surface, wherein processing of said mixture is continued until a change of structure of the mixture is noticeable, e.g. by a change of the X-ray spectrum lines.
2. A process according to claim 1, characterised in that the mixture containing the bitumen and a thermoplast or a mixture of thermoplasts is forced through a narrow gap between two oppositely arranged mixing tools rotating relative to each other, the gap on an average having a width of less than 3 mm, preferably a width of less than 0.3 mm, and the abrupt braking of the mixture is effected in the gap region, preferably immediately after emergence of the mixture from the gap.
3. A process according to claim 2, characterised in that the processing of the mixture is interrupted after each gap passage by pauses.
4. A process according to claim 2 or 3, characterised in that the mixture is forced through a gap present between mixing tools rotating relative to each other, the gap on an average having a width of approximately 0.1 mm or less.



5. A process according to any one of claims 2 to 4, characterised in that the mixture is forced through a gap extending radially at least at the rim of emergence, the mixture emerging from the gap in the radial direction.
6. A process according to any one of the preceding claims, characterised in that a velocity of approximately 30 m/s is imparted to the mixture.
7. A process according to any one of the preceding claims, characterised in that polyethylene is used as the thermoplastic material.