

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
11 October 2007 (11.10.2007)

PCT

(10) International Publication Number
WO 2007/113221 A1

(51) International Patent Classification:
C08L 95/00 (2006.01)

(21) International Application Number:
PCT/EP2007/053055

(22) International Filing Date: 29 March 2007 (29.03.2007)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
2006-099782 31 March 2006 (31.03.2006) JP

(71) Applicant (*for all designated States except US*): **SHELL
INTERNATIONALE RESEARCH MAATSCHAPPIJ
B.V.** [NL/NL]; NL-2596 HR The Hague (NL).

(72) Inventors; and

(75) Inventors/Applicants (*for US only*): **HIRAYAMA,
Hayato** [JP/JP]; Showa Shell Sekiyu K.K., 2-3-2 Daiba,
Minato-ku, 135-8074 Tokyo (JP). **SEO, Akira** [JP/JP];
Showa Shell Sekiyu K.K., 2-3-2 Daiba, Minato-ku,
135-8074 Tokyo (JP). **MORIKUBO, Michio** [JP/JP];
Showa Shell Sekiyu K.K., 2-3-2 Daiba, Minato-ku,
135-8074 Tokyo (JP).

(74) Agent: **ZEESTRATEN, Albertus, Wilhelmus, J.**; C30,
NL-2501 CJ The Hague (NL).

(81) Designated States (*unless otherwise indicated, for every
kind of national protection available*): AE, AG, AL, AM,

AT, AU, AZ, BA, BB, BG, BH, BR, BW, BY, BZ, CA, CH,
CN, CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, EG, ES,
FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN,
IS, KE, KG, KM, KN, KP, KR, KZ, LA, LC, LK, LR, LS,
LT, LU, LY, MA, MD, MG, MK, MN, MW, MX, MY, MZ,
NA, NG, NI, NO, NZ, OM, PG, PH, PL, PT, RO, RS, RU,
SC, SD, SE, SG, SK, SL, SM, SV, SY, TJ, TM, TN, TR,
TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) Designated States (*unless otherwise indicated, for every
kind of regional protection available*): ARIPO (BW, GH,
GM, KE, LS, MW, MZ, NA, SD, SL, SZ, TZ, UG, ZM,
ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM),
European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI,
FR, GB, GR, HU, IE, IS, IT, LT, LU, LV, MC, MT, NL, PL,
PT, RO, SE, SI, SK, TR), OAPI (BF, BJ, CF, CG, CI, CM,
GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Declaration under Rule 4.17:

— *as to applicant's entitlement to apply for and be granted a
patent (Rule 4.17(ii))*

Published:

— *with international search report*
— *before the expiration of the time limit for amending the
claims and to be republished in the event of receipt of
amendments*

*For two-letter codes and other abbreviations, refer to the "Guid-
ance Notes on Codes and Abbreviations" appearing at the begin-
ning of each regular issue of the PCT Gazette.*

(54) Title: ASPHALT COMPOSITION

(57) Abstract: An asphalt composition is made by incorporating 4-18 weight %, based on the total weight of the composition, of a hydrogenated thermoplastic elastomer having a bromine number of 10 g Br₂/100 g or less, a molecular weight of 50 000 to 200 000 and a styrene content of 15-40 weight %, and wherein 5-35 weight % of block copolymers of the aforesaid styrene is present at each of the two ends of the elastomer molecules, into an asphalt such as straight asphalt, blown asphalt or the like. The asphalt composition has excellent thermal stability and high temperature storage stability, and excellent durability in the temperature range from -20°C to 600°C.

WO 2007/113221 A1

ASPHALT COMPOSITION

Technological Field

The present invention relates to an asphalt composition used in road paving, waterproof sheet, soundproof sheet and roofing sheet.

Background Technology

Hitherto, asphalt has been used in a wide range of fields such as road paving and water-proofing. However, since the properties of asphalt generally change greatly with temperature, there are problems in that, for example, in road paving applications, under the climatic conditions in the summer season the temperature of the paved surface reaches about 60°C in direct sunlight, so that the asphalt softens and damage such as rut formation readily occurs, and furthermore in cold regions in the winter season the temperature of the paved surface falls to about -20°C, so that the elasticity of the asphalt decreases, it becomes hard and brittle, and loss of aggregate and cracking readily occur. Further, if the viscosity of the asphalt is increased in order to improve its durability in service, the viscosity during application also becomes high, and mixing with the aggregate becomes unsatisfactory, or the flatness of the paved surface is decreased. Moreover, if the viscosity of the asphalt is decreased in order to improve its application properties, its strength in service becomes inadequate.

Accordingly, hitherto, in order to decrease the temperature-dependence of the properties of asphalt, asphalt compositions were proposed wherein a thermoplastic elastomer was mixed with straight asphalt or a modified asphalt or the like (for example, see Patent Documents JP 2004-307612, JP 2003-238813 and JP 2003-003070.) For example, in the asphalt compositions described in JP 2004-307612, in order to obtain high mechanical strength, high viscosity at 60°C, low viscosity at 200°C and stability as a composition, a 2-chloro-1,3-butadiene polymer and/or a 2,3-dichloro-1,3-butadiene polymer is mixed with an asphalt composition made by thermally melt-mixing a copolymer obtained by copolymerising a vinylaromatic compound and a conjugated diene compound and/or a block copolymer made up of polymer blocks having a vinylaromatic compound as the main component and polymer blocks having a conjugated diene compound as the main component with asphalt.

Further, in JP 2003-238813, an asphalt composition is disclosed wherein 0.5 to 50 parts by weight of a hydrogenated copolymer prepared by addition of hydrogen to a copolymer made up of a conjugated diene and a vinylaromatic compound, wherein the content of the vinylaromatic compound in the hydrogenated copolymer is 50 weight % to 90 weight %, the weight of vinylaromatic compound polymer blocks in the vinylaromatic compound in the hydrogenated copolymer is 40 weight % or less, the weight average molecular weight of the hydrogenated copolymer is 50 000 to 1 000 000, and 75% or more of the double bonds based on the conjugated diene in the hydrogenated polymer have been hydrogenated is added per 100 parts by weight of asphalt.

Furthermore, in JP 2003-003070, an improved asphalt composition for road paving is disclosed wherein, in order to improve the mixing homogeneity of asphalt and a thermo-plastic elastomer and prevent phase separation, in addition to asphalt, rubber or a thermo-plastic elastomer and mineral oil, a phenyl ether compound having specific substituent(s) is also added.

Disclosure of Invention

Problem to be Solved by Invention

However, as described below, there are problems with the aforesaid previous technology. In general, when asphalt is used in road paving applications, after the asphalt has been heated to 180 to ca. 200°C at the application site, thus decreasing its viscosity and making it satisfactorily workable, it is mixed with aggregate, etc., and then applied. At this time, the asphalt is sometimes stored for several days at a temperature of ca. 180°C, and there is a problem in that if the asphalt composition with added thermoplastic elastomer as in the technology of JP 2004-307612, is stored at high temperature, the thermoplastic elastomer forms a gel, rendering subsequent handling difficult. Further, if the asphalt composition forms a gel inside a tank, it becomes impossible to transfer or discharge from the tank, rendering major repair costs necessary. Furthermore, depending on the high temperature storage time and conditions, there may be problems with the thermoplastic elastomer decomposing and the effects due to its addition not being obtained.

On the other hand, with the asphalt compositions described in JP 2003-238813, the aim is to improve high temperature storage stability through the use of a

hydrogenated copolymer as the thermoplastic elastomer, however the hydrogenated copolymer added to this asphalt composition mixes poorly with the asphalt, and phase separation readily occurs. Because of this, the asphalt compositions described in JP 2003-238813 are difficult to obtain with uniform composition, and there are also problems in that, because a long time is necessary for mixing, the workability is poor.

Also, in JP 2003-003070, a technology is disclosed whereby the mixing uniformity of the asphalt and thermoplastic elastomer are improved by adding a phenyl ether compound having specified substituent(s), however, since prolonged high temperature storage is not envisaged in the technology described in this JP 2003-003070, and no studies were made of the changes in properties in that event, there is a problem in that the thermal stability and high temperature storage stability are insufficient.

The present invention was devised in the light of the aforesaid problems, and its purpose is to provide an asphalt composition of excellent thermal stability and high temperature storage stability, which also has excellent durability in the temperature range from -20°C to 60°C.

Means of Solving Problem

The asphalt composition of the present invention contains 4-18 weight % of hydrogenated thermoplastic elastomer, the remainder being made up of asphalt, and is characterised in that the aforesaid hydrogenated thermoplastic elastomer has a bromine number of 10 g Br₂/100 g or less, a molecular weight of 50 000 to 200 000, and a styrene content of 15 to 40 weight %, based on the total weight of the aforesaid hydrogenated

thermoplastic elastomer, and in that 5 to 35 weight % of styrene block copolymer per molecule of elastomer is present at each end region of the aforesaid hydrogenated thermoplastic elastomer molecules.

5 In the present invention, since 4-18 weight % of a hydrogenated thermoplastic elastomer having a bromine number of 10 g Br₂/100 g or less, a molecular weight of 50 000 to 200 000, and a styrene content of 15 to 40 weight %, based on the total weight of the aforesaid
10 hydrogenated thermoplastic elastomer, and of a structure wherein 5 to 35 weight % of styrene block copolymer per molecule of elastomer is present in each end region of the elastomer molecules is incorporated in the asphalt, it is capable of retaining its flexibility at
15 temperatures of about -20°C, and softening at temperatures of about 60°C can also be prevented; moreover, even if stored for a long period at about 180°C, gelling of the thermoplastic elastomer can be prevented. As a result, an asphalt composition of
20 excellent thermal stability and high temperature storage stability, which also has excellent durability in the temperature range from -20°C to 60°C, is obtained.

Further, the aforesaid hydrogenated thermoplastic elastomer is for example a styrene-ethylene-butylene-
25 styrene copolymer or a styrene-ethylene-propylene-styrene copolymer.

Effect of Invention

By means of the present invention, since 4-18 weight % of a hydrogenated thermoplastic elastomer having
30 a bromine number of 10 g Br₂/100 g or less, a molecular weight of 50 000 to 200 000, and a styrene content of 15 to 40 weight %, and of a structure wherein 5 to 35

weight % of styrene block copolymer per molecule of elastomer is present in each end region of the elastomer molecules is incorporated in the asphalt, an asphalt composition of excellent thermal stability and high temperature storage stability, which also has excellent durability in the temperature range from -20°C to 60°C, is obtained.

Optimal Modes for Implementation of Invention

Below, optimal modes for implementation of the present invention are explained in detail. The asphalt composition of the present invention is a mixture of 4-18 weight % of hydrogenated thermoplastic elastomer and 82-96 weight % of asphalt. As the asphalt in the present invention, for example asphalts such as straight asphalt (see JIS K 2207), blown asphalt (see JIS K 2207), semiblow asphalt (see 'Principles of Asphalt Paving', publ. Japanese Roads Association, 13.01.2001, p.51, Table 3.3.4), and deasphalted asphalt (see 'New Petroleum Dictionary', Ed. Petroleum Society, 1982, p.308) or mixtures thereof, or products of the addition of softeners such as petroleum solvent-extracted oils (see 'New Petroleum Dictionary', Ed. Petroleum Society, 1982, p.304) and/or lubricating oils to these various asphalts, and the like, can be used.

On the other hand, the hydrogenated thermoplastic elastomer in the asphalt composition of the present invention has a bromine number of 10 g Br₂/100 g or less, a molecular weight of 50 000 to 200 000, and a styrene content of 15 to 40 weight %, based on the total weight of the hydrogenated thermoplastic elastomer, and 5 to 35 weight % of styrene block copolymer per molecule of elastomer is present in each end region of the molecules

of this hydrogenated thermoplastic elastomer, and, for example styrene-ethylene-butylene-styrene copolymers, styrene-ethylene-propylene-styrene copolymers and the like can be used.

5 Below, the reasons for the numerical limits as to the hydrogenated thermoplastic elastomer in the asphalt compositions of the present invention are explained.

Hydrogenated thermoplastic elastomer content: 4-18 weight %

10 If the hydrogenated thermoplastic elastomer content per total weight of asphalt composition is less than 4 weight %, its strength at temperatures of about 60°C or its flexibility at temperatures of about -20°C decrease and as a result it becomes impossible to obtain adequate
15 durability over the whole temperature range from -20 to 60°C. On the other hand, if the hydrogenated thermoplastic elastomer content per total weight of asphalt composition exceeds 18 weight %, the viscosity of the asphalt composition becomes too high, and as a result
20 transfer becomes difficult, and it must be heated to a high temperature when it is mixed with aggregate, hence its workability and application properties are diminished. Hence, the hydrogenated thermoplastic elastomer content is set at 4-18 weight %.

25 Bromine number: 10 g Br₂/100 g or less

 If the bromine number of the hydrogenated thermoplastic elastomer exceeds 10 g Br₂/100 g, changes in properties such as viscosity increases and gelling occur if it is stored at a temperature of about 180°C.
30 Hence, the bromine number of the hydrogenated thermoplastic elastomer is set at 10 g Br₂/100 g or less. Further, through the use of a hydrogenated thermoplastic

elastomer of bromine number 10 g Br₂/100 g or less, deterioration due to ultraviolet light during use can be prevented, and durability markedly improved.

Molecular weight: 50 000 to 200 000

5 If a hydrogenated thermoplastic elastomer of molecular weight less than 50 000 is used, sufficient strength as an asphalt composition is not obtained. On the other hand, if a hydrogenated thermoplastic elastomer of molecular weight exceeding 200 000 is used, the
10 viscosity of the whole asphalt composition increases, and the application properties deteriorate. Further, to create a viscosity at which workability is maintained, heating and storage at higher temperatures become necessary. Hence, the molecular weight of the
15 hydrogenated thermoplastic elastomer is set at 50 000 to 200 000.

Styrene content: 15-40 weight % based on total weight of hydrogenated thermoplastic elastomer

20 If the styrene content based on total weight of hydrogenated thermoplastic elastomer is less than 15 weight %, sufficient strength as an asphalt composition is not obtained. On the other hand, if the styrene content based on total weight of hydrogenated thermoplastic elastomer exceeds 40 weight %, its
25 solubility in asphalt decreases and as a result a long time is needed for mixing, and workability deteriorates. Hence, the styrene content based on total weight of hydrogenated thermoplastic elastomer is set at 15-40 weight %.

30 Styrene block copolymer weight: 5-35 weight % (per elastomer molecule) at both ends of the elastomer molecule

The hydrogenated thermoplastic elastomer in the asphalt composition of the present invention has a structure wherein styrene block copolymer is present at both ends of the straight-chain elastomer molecule. If a hydrogenated thermoplastic elastomer with a structure wherein styrene block copolymer is present only at one end of the elastomer molecules is used, the strength of the asphalt composition decreases, and as a result adequate strength cannot be obtained. Further, if a hydrogenated thermoplastic elastomer with a structure wherein styrene block copolymer is present in the side-chains is used, the viscosity of the asphalt composition becomes too high, and its application properties deteriorate. Furthermore, if the quantity of one of the block copolymers present at the two ends of the elastomer molecule is less than 5 weight % per elastomer molecule, adequate strength is not obtained. On the other hand, if the quantity of at least one of the block copolymers present at the two ends of the elastomer molecule exceeds 35 weight % per elastomer molecule, its solubility in asphalt decreases and as a result a long time is needed for mixing, and workability deteriorates. Hence, the quantity of styrene block copolymer present at each of the two ends of the elastomer molecule is set at 5-35 weight % per elastomer molecule. It should be noted that this styrene block copolymer quantity need not have the same value at both ends of the hydrogenated thermoplastic elastomer, and the styrene block copolymer quantity at one end can differ from that at the other end, provided that each is in the range 5-35 weight %.

As described above, since the hydrogenated thermoplastic elastomer is added to the asphalt composition of the present invention at the large content of 4-18 weight %, flexibility can be maintained at low temperatures of about -20°C. Further, since hydrogenated thermoplastic elastomer with a styrene content of 15-40 weight % based on the total weight is used, softening at temperatures of about 60°C can be prevented. Moreover, since hydrogenated thermoplastic elastomer with a bromine number of 10 g Br₂/100 g or less is used, property changes such as gel formation and viscosity increase in the asphalt composition are unlikely to occur even on prolonged storage at about 180°C. As a result, an asphalt composition of excellent thermal stability and high temperature storage stability, which also has excellent durability in the temperature range from -20°C to 60°C, is obtained.

Now, the asphalt compositions of the present invention are not limited to paving applications and, apart from paving material, they can also be used as roofing sheet, sealing material, pipe coating material and the like.

Practical Examples

Below, the effects of the present invention are specifically explained by the presentation of practical examples and comparison examples. In the present practical examples, the various asphalt compositions of the practical examples and comparison examples were made by adding hydrogenated thermoplastic elastomer or nonhydrogenated thermoplastic elastomer to asphalt (mixture of solvent-deasphalted asphalt and petroleum solvent-extracted oil) maintained at a temperature of

180°C, and mixing for 2 hrs in a homogeniser at a temperature of 180°C and a revolution rate of 3000 rpm. At this time, it was confirmed by sieve passage tests (mesh 1.18 mm) that the asphalt and thermoplastic elastomers had formed uniform asphalt compositions. Also, the mixing ratio of the solvent-deasphalted asphalt and petroleum solvent-extracted oil was set such that, in each asphalt composition of these practical examples and comparison examples, the penetration was 45-75.

Further, in these practical examples, a solvent-deasphalted asphalt with a penetration of 8 (1/10 mm) at 25°C, softening point 66.5°C, density 1028 kg/m³ at 15°C and flash point of 352°C was used. Further, a petroleum solvent-extracted oil of viscosity 0.07 Pa.sec at 100°C, aromatics content 33 weight %, naphthenes content 26 weight %, paraffins content 41 weight % and flash point 254°C was used. Moreover, a styrene-ethylene-butylene-styrene copolymer (SEBS) (referred to below as copolymer A) with a bromine number of 5 g Br₂/100 g, molecular weight 150 000 and styrene content 30 weight % was used as the hydrogenated thermoplastic elastomer, and a styrene-butadiene-styrene copolymer (SBS) (referred to below as copolymer B) with a bromine number of 220 g Br₂/100 g, molecular weight 150 000 and styrene content 32 weight % was used as the non-hydrogenated thermoplastic elastomer. The compounding proportions for each asphalt composition are shown in Table 1 below.

TABLE 1

	Composition (weight %)			
	Asphalt		Thermoplastic elastomer	
	Solvent-deasphalted asphalt	Petroleum solvent-extracted oil	Polymer A	Polymer B
Pract. Ex. 1	34	49	17	—
Pract. Ex. 2	40	45	15	—
Pract. Ex. 3	52	36	12	—
Pract. Ex. 4	58	32	10	—
Pract. Ex. 5	70	24	6	—
Comp. Ex. 1	43	41	—	16
Comp. Ex. 2	67	25	—	8
Comp. Ex. 3	74	24	2	—
Comp. Ex. 4	74	24	—	2
Comp. Ex. 5	19	60	21	—
Comp. Ex. 6	30	49	—	21

Next, the penetration at 25°C and softening point of the asphalt compositions of the practical examples and comparison examples prepared by the aforesaid method were measured, and also, measurements of the viscosity at 200°C for assessment of the application properties, measurements of the complex modulus of elasticity G^* at 60°C for assessment of the service durability (summer season), measurements of the stiffness at -20°C for assessment of the service durability (winter season) and measurements of the percentage increase in viscosity

after 7 days' curing for assessment of the high temperature storage stability were each performed. The specific measurement methods for each assessment heading are explained below.

5 The penetration and softening point were measured by the methods specified in JIS K2207. Further, the viscosity at 200°C was measured using a Brookfield Co. rotary viscometer with a No.SC4-21 spindle and a measurement revolution rate of 20 rpm. In these practical
10 examples, since the on-site working properties are improved by lowering the viscosity of the asphalt composition, a viscosity value at 200°C of 1200 mPa.sec or less was regarded as a pass.

 Further, the complex modulus of elasticity G^* at
15 60°C was measured by DSR (Dynamic Shear Viscometer) testing (see 'Paving Test Methods Handbook Supplement (Provisional Test Methods)', Japanese Roads Association, October 1996, pp.91-99). In these practical examples, since the anti-flow performance of the asphalt was
20 improved, the asphalt flow was assessed with a low shear velocity at which asphalt flow readily occurred, and a complex modulus of elasticity G^* of 5 kPa or greater at a measurement temperature of 60°C and measurement frequency of 0.1 rad/sec was regarded as a pass.

25 Furthermore, the stiffness at -20°C was assessed by BBR (Bending Beam Rheometer) testing (see 'Paving Test Methods Handbook Supplement (Provisional Test Methods)', Japanese Roads Association, October 1996, pp.81-87). For this, a value at which adequate flexibility is obtained
30 even in cold regions in the winter season, in other words a stiffness after 60 secs under temperature conditions of

-20°C using the SHRP (Strategic Highway Research Program) SUPERPAVE (US general system for assessment and analysis of asphalt paving) of 600 MPa or less was regarded as a pass.

5 As regards the high temperature stability, the asphalt compositions were filled into 2.5 kg cans and sealed, cured in this state for 7 days in an oven maintained at 190°C, and the viscosity before and after curing compared. The viscosity measurements were
10 performed using a Brookfield Co. rotary viscometer, at a measurement temperature of 180°C, with a No.SC4-21 spindle and a measurement revolution rate of 20 rpm. Then the ratio of the viscosity after curing to the viscosity directly after production (viscosity after
15 curing/viscosity directly after production) was expressed as an index, and taken as the percentage viscosity increase (%). Since the application properties deteriorate greatly if the viscosity increases when, after production of the asphalt, the asphalt is used for
20 application on site, results where this percentage viscosity increase was in the range 90-120% were taken as passes. The above results are shown in summary in Table 2 below.

TABLE 2

	Penetration at 25°C (1/10 mm)	Softening point (°C)	Viscosity at 200°C (mPa.sec)	Complex modulus of elasticity G* (kPa)	Stiffness (MPa)	Percentage viscosity increase (%)
Pract. Ex. 1	62	90.5	1150	29	11	97
Pract. Ex. 2	60	89.0	842	21	18	101
Pract. Ex. 3	63	93.0	505	12	62	98
Pract. Ex. 4	59	94.0	348	10	112	97
Pract. Ex. 5	49	89.0	155	5	549	101
Comp. Ex. 1	80	86.0	815	14	3	500 or more
Comp. Ex. 2	57	87.0	208	6	94	180
Comp. Ex. 3	61	48.5	60	0.03	837	100
Comp. Ex. 4	65	48.5	58	0.03	691	117
Comp. Ex. 5	57	86.5	1980	10	3	102
Comp. Ex. 6	72	87.5	1645	10	2	500 or more

As shown in the aforesaid Table 2, although the thermoplastic elastomer contents of the asphalt compositions of Comparison Example 1 and Comparison Example 2 were within the range of the present invention (4-18 weight %), the percentage viscosity increase due to curing for 7 days was high, and their high temperature storage stability was inferior. Further, with the asphalt compositions of both Comparison Example 3 and Comparison Example 4, the complex modulus of elasticity G^* at 60°C was low, and the durability at 60°C was inferior. Further, with these asphalt compositions, the stiffness values at -20°C were high, and the durability at -20°C was also inferior. Furthermore, with the asphalt composition of Comparison Example 5, the viscosity at 200°C was high, and the application properties were inferior. Furthermore, with the asphalt composition of Comparison Example 6, the viscosity at 200°C was high, and the application properties were inferior; also, the percentage viscosity increase due to curing for 7 days was high, and its high temperature storage stability was also inferior.

In contrast to this, the asphalt compositions of Practical Examples 1-5 within the scope of the present invention had viscosities of 1200 mPa.sec or less at 200°C, and their application properties were excellent. Further, since the complex modulus of elasticity G^* at 60°C of the asphalt compositions of Practical Examples 1-5 was 5 kPa or more, and the stiffness values at -20°C were 600 MPa or less, it was confirmed that in the temperature range from -20 to 60°C superior durability compared to the asphalt compositions of the aforesaid

Comparison Examples is obtained. Further, even after curing for 7 days at temperature 190°C, the percentage viscosity increase of the asphalt compositions of Practical Examples 1-5 was in the range 90 to 120%, and
5 their high temperature stability was excellent.

Further, as a comparison example of the present invention, an asphalt composition was made with the same compounding ratios and production method as for the asphalt composition of Practical Example 1 shown in the
10 aforesaid Table 1, using a styrene-ethylene-butylene-styrene copolymer of bromine number 5 g Br₂/100 g, molecular weight 50 000 and styrene content 72 weight %, wherein the quantity of styrene block copolymer present at both ends of the elastomer molecule was 36 weight %, as the hydrogenated thermoplastic elastomer. As a result,
15 when, as confirmation of completeness of mixing, its solubility was checked in terms of whether the asphalt and thermoplastic elastomer formed a uniform asphalt solution, by the passage test using a sieve (mesh
20 1.18 mm), a residual fraction of hydrogenated thermoplastic elastomer was identified, hence its dissolution was judged to be unsatisfactory, and further processing and assessments were not performed.

C L A I M S

1. Asphalt composition containing 4-18 weight % of hydrogenated thermoplastic elastomer, the remainder whereof is made up of asphalt, characterised in that the aforesaid hydrogenated thermoplastic elastomer has

5 a bromine number of 10 g Br₂/100 g or less,
a molecular weight of 50 000 to 200 000, and
a styrene content of 15 to 40 weight %, based on the total weight of the aforesaid hydrogenated thermoplastic elastomer, and

10 5 to 35 weight % of styrene block copolymer per molecule of elastomer is present in each end region of the aforesaid hydrogenated thermoplastic elastomer molecules.

2. Asphalt composition according to Claim 1, characterised in that the aforesaid hydrogenated thermoplastic elastomer is
15 a styrene-ethylene-butylene-styrene copolymer or a styrene-ethylene-propylene-styrene copolymer.

3. Asphalt composition according to claim 1 or claim 2, characterised in that the asphalt is selected from the group consisting of straight asphalt, blown asphalt, semiblow
20 asphalt and deasphalted asphalt or mixtures thereof, or is a product of the addition of softeners such as petroleum solvent-extracted oils and/or lubricating oils to an asphalt selected from the group consisting of straight asphalt, blown asphalt, semiblow asphalt and deasphalted asphalt or
25 mixtures thereof.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2007/053055

A. CLASSIFICATION OF SUBJECT MATTER

INV. C08L95/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

C08L C08F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	GB 2 257 708 A (SHELL INT RESEARCH [NL]) 20 January 1993 (1993-01-20) page 5, line 4 - page 6, line 14; claims 1-16; table 2	1-3
X	US 5 246 987 A (MASSE MICHAEL A [US] ET AL) 21 September 1993 (1993-09-21) claims 1-20; example 1	1-3
X	EP 0 731 111 A2 (KURARAY CO [JP]) 11 September 1996 (1996-09-11) page 6, line 13; examples 1-3	1-3

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents:

A document defining the general state of the art which is not considered to be of particular relevance

E earlier document but published on or after the international filing date

L document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

O document referring to an oral disclosure, use, exhibition or other means

P document published prior to the international filing date but later than the priority date claimed

T later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

X document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

Y document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

G document member of the same patent family

Date of the actual completion of the international search

6 August 2007

Date of mailing of the international search report

16/08/2007

Name and mailing address of the ISA/

European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax: (+31-70) 340-3016

Authorized officer

oide Scheper, Bernd

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2007/053055

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
GB 2257708	A	20-01-1993	NONE	
US 5246987	A	21-09-1993	GB 2267093 A	24-11-1993
EP 0731111	A2	11-09-1996	CA 2171045 A1	07-09-1996
			DE 69602647 D1	08-07-1999
			DE 69602647 T2	10-02-2000
			US 5616652 A	01-04-1997