

19



Octrooi centrum  
Nederland

11

2019501

12 B1 OCTROOI

21 Aanvraagnummer: **2019501**

22 Aanvraag ingediend: **7 september 2017**

51 Int. Cl.:

**C08L 53/00** (2018.01) **C08L 23/04** (2018.01) **C08L 23/08** (2018.01) **C08K 5/103** (2018.01) **C08K 5/20** (2018.01) **C08J 9/00** (2018.01)

41 Aanvraag ingeschreven:  
**14 maart 2019**

43 Aanvraag gepubliceerd:  
-

47 Octrooi verleend:  
**14 maart 2019**

45 Octrooischrift uitgegeven:  
**21 mei 2019**

73 Octrooihouder(s):  
**Thermafex International Holding B.V.**  
te **WAALWIJK**.

72 Uitvinder(s):  
**Maikel Josef Paulus Johannes Renders**  
te **VALKENSWAARD**.  
**Tomasz Duzak** te **WAALWIJK**.  
**Musa Aksoy** te **WAALWIJK**.

74 Gemachtigde:  
**ir. F.A. Geurts c.s. te Den Haag**.

54 **Flexible polyolefin thermal insulation foam and use thereof, and a method for producing a flexible polyolefin thermal insulation foam.**

57 The present invention relates to a polyolefin thermal insulation foam and use thereof, and to a method for preparing a physically foamed polyolefin thermal insulation foam, which can be recycled well and which has excellent flexibility characteristics.

NLP201747

**Flexible polyolefin thermal insulation foam and use thereof, and a method for producing a flexible polyolefin thermal insulation foam.**

The present invention relates to a polyolefin thermal insulation foam and use thereof, and to a method for preparing a physically foamed polyolefin thermal insulation foam.

For the thermal insulation of pipes such as hot water conduits, high and low pressure steam pipes, and pipes for split-air conditioning, district heating, solar energy exploitation and the process industry, hollow profiles having a wall of synthetic foam are used on a large scale.

The most common types of pipe insulation that are currently commercially available are polyethylene (PE) pipe insulation and rubber pipe insulation.

PE pipe insulation consists of foam that is composed of thermoplasts and has a density of approximately 35 kg/m<sup>3</sup>. This type of foam is usually produced using physical blowing agents (for instance iso-butane) and is not crosslinked. This type of foam has good product properties such as insulation value, fire behaviour, water vapor transmission and water absorption capacity. The foam can be recycled excellently due to its non-crosslinked nature. The foam is prepared in a single process, i.e. the production takes place in one step. A drawback of PE pipe insulation material is however that it is less flexible than rubber pipe insulation as a result of which it is difficult to apply around thin and twisting pipes. As a result the material is not suitable for some uses such as for instance air conditioning and cooling.

Rubber pipe insulation consists of foam that is composed of elastomers and has a density of about 60 kg/m<sup>3</sup>. This type of foam is often produced using chemical blowing agents (for instance azo-compounds) and usually is cross-linked. Rubber pipe insulation is a very flexible material that is easy to apply. This type of foam has good product properties such as insulation value and fire behaviour. A drawback of rubber pipe insulation is that it cannot be recycled and that it is relatively heavy (that means that a lot of material is necessary for insulation). The water absorption capacity is good as such, but in case of damage to its skin the material behaves like a sponge and good properties are lost. Another drawback of rubber pipe insulation is that this material is produced using a method comprising three steps: kneading, extruding and foaming. In combination with the high density this makes the cost price of rubber pipe insulation higher than that of PE pipe insulation.

A pipe insulation that is flexible, has excellent thermal insulation capacity, and that can be recycled well was described in W002/42679 of the present applicant. The polyolefin foam disclosed therein is made from a basis of metallocene polyethylene. W002/42679 discloses in particular a thermal insulation foam which is made by extruding, using a physical blowing agent, a foam composition comprising a metallocene polyethylene, a flame extinguisher and a cell stabilizer, characterised in that said composition comprises 77-92 % by weight of metallocene polyethylene, 5-10 % by weight of flame extinguisher, and 3-8 % by weight of cell stabilizer.

The present inventors have however found that there is still a need for improvement with regard to the flexibility of the foam. The present invention therefore aims to provide a foam which has excellent thermal insulation capacity, which can be recycled well and which has excellent flexibility characteristics.

### Summary of the invention

In a first aspect the invention therefore provides a non-crosslinked polyolefin thermal insulation foam, which is obtainable by extruding a foam composition comprising:

5 i) an ethylene/  $\alpha$ -olefin block copolymer, which comprises alternating blocks of: crystalline or semi-crystalline blocks characterized by comprising ethylene in an amount of higher than 95 weight %; and elastomeric blocks characterized by comprising ethylene in an amount of 95 weight %

10 or less and a comonomer content of 5 weight % or higher. The foam composition also comprises ii) one or more random polymers with a density of between 0,880 g/cm<sup>3</sup> and 960 g/cm<sup>3</sup>, selected from the group of a metallocene polyethylene and/or a polyethylene. The foam composition also

15 comprises iii) a cell stabilizer. In a second aspect, the invention relates to a method for producing a non-crosslinked polyolefin thermal insulation foam, comprising the steps of

a) extruding, using a physical blowing agent, a

20 foam composition as defined under the first aspect of the invention, in an extruder,

b) melting said mixture in the melting zones of the extruder adjusted to temperatures of 160 to 220°C, at a pressure increasing from 1 bar up to 400 bar,

25 c) injecting said physical blowing agent at an injection temperature of 140 to 180°C and an injection pressure of 30 to 300 bar,

d) cooling the molten mixture in one or more cooling zones of the extruder adjusted to temperatures of 85

30 to 115°C, and

e) extruding the mixture through an extrusion nozzle adjusted to a temperature of 85 to 115°C, so that the mixture expands to a foam at a pressure of 1 atm.

In a third aspect the invention relates to the use of

35 the non-crosslinked polyolefin thermal insulation foam for thermal isolation.

The inventors have surprisingly found that while an ethylene/  $\alpha$ -olefin block copolymer as defined above alone cannot be foamed to a suitable thermal insulation foam, when such a block copolymer is combined into a composition with one or more olefin based random polymers with a density of between 0,880 g/cm<sup>3</sup> and 960 g/cm<sup>3</sup>, this composition can be foamed easily and results in an excellent thermal insulation foam. The resulting composition behaves well in an extruder and the polyolefin thermal insulation foam according to the invention and made with this composition has excellent thermal insulation capacity. Due to the high flexibility of the foam, pipe insulation made from the foam according to the invention is easier applied around twisting pipes than pipe insulations from existing polyolefin based foams such as the one described in W002/42679. Analogous, when the foam is made in the form of a sheet, such sheets can be easier applied over irregular surfaces. Because the foam is prepared using physical blowing agents and is not chemically crosslinked, it can be recycled easily.

#### **Description of the figures**

Fig.1: Flexibility of a pipe insulation of a prior art foam.

Fig.2: Flexibility of a pipe insulation of an exemplary foam according to the invention.

#### **Detailed description**

The foam composition used for obtaining the foam according to the invention comprises: i) an ethylene/  $\alpha$ -olefin block copolymer, ii) one or more olefin based random polymers; and iii) one or more cell stabilizer.

The ethylene/  $\alpha$ -olefin block copolymer comprises blocks of: crystalline or semi-crystalline blocks characterized by comprising ethylene in an amount of higher than 95 weight % based on the weight of said crystalline or semi-crystalline blocks; and elastomeric blocks character-

ized by comprising ethylene in an amount of 95 weight % or less and a comonomer content of 5 weight % or higher based on the weight of said elastomeric blocks. The terms "soft blocks" and "elastomeric blocks" can be used interchangeably. The terms "hard blocks" and "crystalline or semi-crystalline blocks" can also be used interchangeably.

The comonomer content in the hard blocks is less than 5 weight percent based on the weight of said hard blocks, and preferably less than 2 weight percent. The hard blocks may be comprised of only ethylene or substantially only ethylene. Elastomeric blocks (soft blocks), on the other hand, are blocks of polymerized units in which the comonomer content is higher than 5 weight percent of the weight of the soft blocks, preferably higher than 8 weight percent, higher than 10 weight percent, or higher than 15 weight percent. The comonomer content in the soft blocks may even be higher than 20 weight percent, higher than 25 weight percent, higher than 30 weight percent, higher than 35 weight percent, higher than 40 weight percent, higher than 45 weight percent, higher than 50 weight percent, or higher than 60 weight percent.

The term "crystalline" refers to a block that possesses a first order transition or crystalline melting point ( $T_m$ ) as determined by differential scanning calorimetry (DSC) or equivalent technique. The term may be used interchangeably with the term "semicrystalline".

The hard blocks may suitably be present in an amount from 5% to 85% by weight of the block copolymer. The hard blocks and soft blocks may be connected in a linear fashion to form a linear chain and be randomly distributed along this chain. The block copolymer suitably comprises said hard and soft blocks in an alternating fashion.

Preferably, the hard blocks comprise at least 98% of ethylene by weight, and the soft blocks comprise less than 95%, preferably less than 50%, of ethylene by weight.

It is preferred that in the block copolymer the  $\alpha$ -olefin comonomer is a  $C_3$ - $C_{10}$   $\alpha$ -olefin. Suitable  $C_3$ - $C_{10}$   $\alpha$ -

olefins include styrene, propylene, 1-butene, 1-hexene, 1-octene, 4-methyl-1-pentene, norbornene, 1-decene, 1,5-hexadiene or a mixture thereof. In a particularly preferred embodiment said  $\alpha$ -olefin is 1-octene.

5 Preferably, ethylene comprises the majority molar fraction of the block copolymer. In this respect it is preferred that ethylene comprises at least 50 mole percent of the whole block copolymer. More preferably ethylene comprises at least 60 mole percent, at least 70 mole per-  
10 cent, or at least 80 mole percent, with the substantial remainder of the whole polymer comprising at least one other comonomer that is C<sub>3</sub>-C<sub>10</sub>  $\alpha$ -olefin. In case the block copolymer is an ethylene/1-octene block copolymer, suitable ethylene/1-octene block copolymers may comprise an  
15 ethylene content greater than 80 mole percent of the whole polymer and a 1-octene content of from 10 to 15 mole percent, preferably from 15 to 20 mole percent of the whole polymer.

In a particularly preferred embodiment said ethylene/  
20  $\alpha$ -olefin block copolymer comprises alternating blocks of hard blocks of linear medium-density polyethylene and soft blocks of ethylene/1-octene. Medium-density polyethylene (MDPE) is a type of polyethylene defined by a density range of 0,926-0,940 g/cm<sup>3</sup> (as measured according to ASTM  
25 D792).

Suitable ethylene/  $\alpha$ -olefin block copolymers have a density between 800 and 880 g/cm<sup>3</sup> as measured according to ASTM D792. Other physical properties of suitable ethylene/  
30  $\alpha$ -olefin block copolymers include a melt index (g/10 min (2,16 kg at 190°) as determined in accordance with ASTM D1238 between 0,5 and 30, preferably between 0,5 and 5 or a DSC melting point between 115 and 125°C, such as between 118 and 122°C, preferably both of these physical properties. Suitable ethylene/  $\alpha$ -olefin block copolymers with  
35 such properties are commercially available and may be provided in the form of granules.

The inventors have observed that said ethylene/  $\alpha$ -olefin block copolymer as defined above on it itself cannot be foamed to a suitable thermal insulation foam, because this block copolymer does not retain gas. The present invention overcomes this problem by the addition of one or more random polymers with a density of between 0,880 g/cm<sup>3</sup> and 960 g/cm<sup>3</sup> (densities are as measured according to ASTM D792), selected from the group of a metallocene polyethylene and/or a polyethylene.

10 It is to be understood that the term "polyethylene" in this application encompasses both ethylene copolymers and ethylene homopolymers. In case of a random copolymer, it is preferred to use an ethylene/ C<sub>3</sub>-C<sub>10</sub>  $\alpha$ -olefin random copolymer. Suitable C<sub>3</sub>-C<sub>10</sub>  $\alpha$ -olefins in this respect include styrene, propylene, 1-butene, 1-hexene, 1-octene, 15 4-methyl-1-pentene, norbornene, 1-decene, 1,5-hexadiene or a mixture thereof. In a particularly preferred embodiment in the random polymer said  $\alpha$ -olefin is 1-octene.

The random polyethylene polymer component contributes to enhanced melt strength of the produced foam and contributes to foamability. It is preferred that said polyethylene is LDPE. LDPE is defined by a density range of 0,910-0,940 g/cm<sup>3</sup>. A suitable LDPE may have a melt flow index (MFI) as determined in accordance with ASTM D1238 of 20 between 0,5 and 5, such as 0,65 and a melting temperature between 108 and 118 °C, such as between 110 and 115 °C. The inventors have observed that a foaming composition of the ethylene/  $\alpha$ -olefin block copolymers in combination with LDPE retains gas well, so that a foam can be produced with 25 sufficient thermal insulation capacity. The inventors have found that metallocene polyethylene also contributes particularly to achieving high foamability of materials with low material density such as the  $\alpha$ -olefin block copolymer component. The term "metallocene polyethylene" refers to 30 polyethylenes that are prepared by polymerising ethylene in the presence of a metallocene catalyst. For preparing and processing metallocene polyethylene reference is made

to for instance Kurt W. Schwogger, An outlook for metallocene and single site catalyst technology into the 21<sup>st</sup> century, Antec 98, Processing Metallocene Polyolefines, Conference Proceedings, October 1999, Rapra Technology, and  
5 Proceedings of 2<sup>nd</sup> International Congress on Metallocene Polymers, Scotland Conference Proceedings, March 1998. A preferred metallocene polyethylene for use in this invention is an ethylene based octane metallocene plastomer.

It is preferred that in addition to the block copolymer  
10 both a metallocene polyethylene and a polyethylene are included in the foaming composition. This way optimal melt strength and foamability are obtained.

While preparing the foaming composition the balance  
15 between the amount of block copolymer component and amount of the metallocene polyethylene and/or polyethylene component depends on the precise properties of the components that are selected to prepare the foam. It has been observed that a high amount of block copolymer component provides high flexibility, but may be detrimental to  
20 foamability if too high, while a high amount of the metallocene polyethylene and/or polyethylene component promotes foamability, but is detrimental to flexibility of the foam if too high. When a block copolymer comprising alternating blocks of crystalline blocks of linear MDPE and elastomeric blocks of ethylene/1-octene is used, a suitable foaming  
25 composition preferably comprises between 25 and 80 weight % of said one or more random polymers and 20 and 75 weight % of said block copolymer, based on the weight of the composition. Preferred compositions comprise between 30 and  
30 70 weight % of said metallocene polyethylene (e.g. ethylene based octane metallocene) and/or polyethylene component (e.g. LDPE), such as between 30 and 60 weight % or between 40 and 50 weight % based on the weight of the composition. A suitable foam composition may comprise for  
35 instance 60 weight % of said ethylene/  $\alpha$ -olefin block copolymer, 10 weight % LDPE, and 15 weight % ethylene based octane metallocene based on the weight of the composition.

To produce the foam according to the invention, a cell stabilizer is required. Such a cell stabilizer prevents the blowing agent from escaping from the polymer melt immediately after injection, as a result of which no foam is formed. Any cell stabilizer normally used in the art can be used as cell stabilizer, provided that it does not affect the properties of the foam. Examples of suitable cell stabilizers include cell stabilizers of the type stearic acid amide, glycol monostearate and fatty acids of glycine. It is also possible to use more than one cell stabilizer. The total quantity of cell stabilizer in the foam composition may suitably be about 1-8 % by weight, such as between 2-5 weight %, based on the total quantity of polymers and additives. In the production process, the cell stabilizer is suitably added by means of for instance a side feeder, to the polymer mixture before the mixture is melted.

Said foam composition preferably comprises one or more additives selected from the group consisting of a flame retardant, a cell nucleator, a colourant, an isolation improver, an uv stabilizer, a processing aid, a processing stabilizer, and an anti-oxidant.

A suitable foam has a density of 10-45 kg/m<sup>3</sup>, volumetrically determined, for instance 25-30 kg/m<sup>3</sup>, a cross-section of cells between 0,30-0,80 mm and an insulation value of 0,035-0,040 λ40, W/m.K.

The non-crosslinked polyolefin thermal insulation foam according to the invention may suitably be made in accordance with the method of the invention, which comprises the steps of

a) extruding, using a physical blowing agent, a foam composition as defined under the first aspect of the invention, in an extruder,

b) melting said mixture in the melting zone(s) of the extruder adjusted to temperatures of 160 to 220°C, at a pressure increasing from 1 bar up to 400 bar,

c) injecting said physical blowing agent at an injection temperature of 140 to 180°C and an injection pressure of 30 to 300 bar,

5 d) cooling the molten mixture in one or more cooling zones of the extruder adjusted to temperatures of 85 to 115°C, and

e) extruding the mixture through an extrusion nozzle adjusted to a temperature of 85 to 115°C, so that the mixture expands to a foam at a pressure of 1 atm.

10 The foam composition preferably is a water-free mixture which is made by mixing the solid components contained therein. Polymers may suitably be provided in the form of granules.

15 As blowing agent any substance can be used that is liquid at high pressure, particularly the pressure prevailing in the extruder used for carrying out the method, but which substance evaporates at lower pressure. Non-limiting examples of the blowing agent comprise alkanes having 3 to 8 carbon atoms, such as for instance propane,  
20 butane, isobutane and hexane. The blowing agent is brought to a temperature of 140 to 180°C and a pressure of 30 to 300 bar and continuously injected into the melted mixture in the extruder.

25 It is important that the foaming composition is melted well in the extruder, i.e. the polymer is brought in the liquid phase resulting in such a viscoelastic behaviour that polymers and additives are mixed well into each other and that in a later stage also the physical blowing agent is incorporated well into the polymer mixture. When  
30 for the preparation of the polyolefin foam according to the invention a mixer is used in which only low shearing forces are exerted on the mixture, it is advantageous to choose such a temperature that the viscoelastic behaviour of the polymer and additives is almost equal. A parameter  
35 to express viscous behaviour is the melt flow index (MFI) (the throughput of material at a certain temperature and pressure). With the present method the MFI-values of poly-

mers and additives are suitably between 0.5 and 5 g/10 minutes at 2.16 kg @ 190°C as determined in accordance with ASTM D1238.

5 The method of the invention can suitably be carried out on a single or double screw extruder having an L/D between 30 and 60, provided with mixing parts and a static mixer having for instance a throughput of 50 to 600 kg/h.

10 The insulation material according to the invention suitably has a wall thickness of 3 to 50 mm at an inner diameter of 4-130 mm.

### Examples

The following examples are meant to illustrate and not to limit the invention.

15

#### Example 1

To produce a foam composition a mixture was prepared by mixing the compounds listed below to obtain an exemplary foam composition for making an exemplary foam according to the invention (percentages are weight percentages based on weight of the composition:

- 23,2% of LDPE;
- 50% of an ethylene/  $\alpha$ -olefin block copolymer comprises alternating blocks of crystalline blocks of linear medium-density polyethylene and elastomeric blocks of ethylene/1-octene;
- 15% of an ethylene based octene plastomer produced with metallocene catalyst;
- 2,5% of a glycerol monostearate / stearamide cell stabilizer;
- the remainder being additives (flame retardant, cell nucleator, isolation improver, processing aid/stabiliser, anti-oxidant).

35

#### Example 2

A single screw extruder of the type described above was provided with an open space of 5-100 mm<sup>2</sup>, after which the number of revolutions was adjusted at 15-40 rpm. The foaming composition of example 1 was added. The melting

zones of the extruder were adjusted at 160-220°C, the cooling zones were adjusted at 85-115°C. Blowing agent (propellant) was injected at an injection pressure of 67 bar and an injection temperature of approximately 170° in a quantity of 20-25 l/h (as liquid). The pressure in the extruder decreased to approximately 10-30 bar at the nozzle of the extruder, after which the mixture expanded to a foam having a density of 10 to 15 kg/m<sup>3</sup> in the form of pipe insulation having an internal diameter of 18-28 mm and a wall thickness of 20-30 mm. The foam has a density of 10-30 kg/m<sup>3</sup>, volumetrically determined, for instance 10-15 kg/m<sup>3</sup>, a cross-section of cells between 0,30-0,50 mm and an insulation value of 0,035-0,040 λ40, W/m.K.

### Example 3

The properties of the pipe insulation prepared as described in example 2 were compared to a pipe insulation as described in the example of W002/42679 which is made of a composition comprising predominantly metallocene polyethylene, but no block copolymer (Comparative example). The pipe insulation prepared as described in example 2 had similar thermal insulation characteristics compared to the pipe insulation of the comparative example. Elongation at break, also known as fracture strain, was also tested. Elongation at break is the ratio between changed length and initial length after breakage of the test specimen. It expresses the capability of a material to resist changes of shape without crack formation. The elongation at break was by tensile testing in accordance with EN ISO 527. From table 2 below it appears that the pipe insulation made with the foam of example 2 has improved elongation at break; it breaks at an elongation of 175%, while the pipe insulation of the comparative example breaks at an elongation of 104%.

	Elongation at break (%)
Pipe insulation	175

(example 2)	
Pipe insulation (Comparative example)	104

**Table 2: Elongation at break**

The improved flexibility of the polyolefin thermal insulation foam of the invention is visualised in the photos shown in Figures 1 and 2. The pipe insulations shown in Figs. 1 and 2 had the same dimensions. In figure 1 the pipe insulation of the comparative example was held in the air horizontal along the dotted line, manually holding one end and leaving the other end unsupported. In figure 2 the pipe insulation of example 2 was held in the air horizontal along the dotted line holding one end and leaving the other end unsupported. It is clear from the photos that the pipe insulation made of a foam according to the invention (Fig. 2) bends significantly more than the pipe insulation of the comparative example (Fig. 1), which shows that it has improved flexibility compared to the pipe insulation of the comparative example. This proves the enhanced flexibility of the polyolefin thermal insulation foam of the invention.

## CONCLUSIES

1. Niet-verknoopt thermisch isolatieschuim op polyolefinebasis, verkrijgbaar door het extruderen van een schuimsamenstelling omvattende:

5 i) een ethyleen/ $\alpha$ -olefineblokcopolymeer, dat blokken omvat van:

A) kristallijne of semikristallijne blokken gekenmerkt door het omvatten van ethyleen in een hoeveelheid van hoger dan 95 gew.%; en

10 B) elastomere blokken gekenmerkt door het omvatten van ethyleen in een hoeveelheid van 95 gew.% of minder en een comonomeergehalte van 5 gew.% of hoger;

15 ii) één of meer willekeurige polymeren met een dichtheid van tussen 0,880 g/cm<sup>3</sup> en 960 g/cm<sup>3</sup>, gekozen uit de groep bestaande uit een metalloceenpolyethyleen en/of een polyethyleen; en

iii) een celstabilisator.

2. Niet-verknoopt thermisch isolatieschuim op polyolefinebasis volgens conclusie 1, waarbij het  $\alpha$ -olefine een C3-C10  $\alpha$ -olefine is.

3. Niet-verknoopt thermisch isolatieschuim op polyolefinebasis volgens conclusie 1 of 2, waarbij het ethyleen/ $\alpha$ -olefineblokcopolymeer afwisselende blokken van 25 kristallijne blokken van lineair polyethyleen van medium dichtheid en elastomere blokken van ethyleen/1-octeen omvat.

4. Niet-verknoopt thermisch isolatieschuim op polyolefinebasis volgens één van de conclusies 1 tot 3, waar- 30

bij het ethyleen/ $\alpha$ -olefineblokcopolymeer een dichtheid tussen 800 en 880 g/cm<sup>3</sup> heeft.

5 5. Niet-verknoopt thermisch isolatieschuim op poly-  
olefinebasis volgens één van de conclusies 1 tot 4, waar-  
bij het polyethyleen onder ii) LDPE is.

10 6. Niet-verknoopt thermisch isolatieschuim op poly-  
olefinebasis volgens één van de conclusies 1 tot 5, waar-  
bij het metalloceenpolyethyleen onder ii) een op ethyleen  
gebaseerd octaanmetalloceen is.

15 7. Niet-verknoopt thermisch isolatieschuim op poly-  
olefinebasis volgens één van de conclusies 1 tot 5, waar-  
bij de één of meer willekeurige polymeren zowel een ge-  
noemd metalloceenpolyethyleen als een genoemd polyethyleen  
omvatten.

20 8. Niet-verknoopt thermisch isolatieschuim op poly-  
olefinebasis volgens één van de conclusies 1 tot 7, waar-  
bij de schuimsamenstelling één of meer additieven omvat  
gekozen uit de groep bestaande uit een vlamvertrager, een  
celnucleator, een kleurmiddel, een isolatieverbeteraar,  
een UV-stabilisator, een verwerkingshulpmiddel, een ver-  
werkingstabilisator, en een antioxidant.

30 9. Niet-verknoopt thermisch isolatieschuim op poly-  
olefinebasis volgens één van de conclusies 1 tot 8, waar-  
bij de schuimsamenstelling tussen 25 en 80 gew.% van de  
een of meer op olefine gebaseerde polymeren onder ii) om-  
vat.

35 10. Niet-verknoopt thermisch isolatieschuim op poly-  
olefinebasis volgens één van de conclusies 1 tot 9, waar-  
bij de schuimsamenstelling tussen 20 en 75 gew.% van de  
ethyleen/ $\alpha$ -olefineblokcopolymeer onder i) omvat.

11. Niet-verknoopt thermisch isolatieschuim op polyolefinebasis volgens één van de conclusies 1 tot 10, dat in de vorm van een cilinder is.

5 12. Niet-verknoopt thermisch isolatieschuim op polyolefinebasis volgens één van de conclusies 1 tot 10, dat in de vorm van een vel is.

10 13. Werkwijze voor het maken van niet-verknoopt thermisch isolatieschuim op polyolefinebasis, omvattende de stappen van:

a) het extruderen, bij gebruik van een fysisch blaasmiddel, van een schuimsamenstelling zoals gedefinieerd in één van de conclusies 1 tot 10, in een extruder;

15 b) het smelten van het mengsel in één of meer smeltgebieden van de extruder die ingesteld zijn op temperaturen van 160 tot 220 °C, bij een druk die verhoogd wordt van 1 bar tot 400 bar;

20 c) het injecteren van het fysische blaasmiddel bij een injectietemperatuur van 140 tot 180 °C en een injectiedruk van 30 tot 300 bar;

d) het afkoelen van het gesmolten mengsel in één of meer koelgebieden van de extruder die ingesteld zijn op temperaturen van 85 tot 115 °C; en

25 e) het extruderen van het mengsel door een extrusiemond die ingesteld is op een temperatuur van 85 tot 115 °C, zodat het mengsel expandeert tot een schuim bij een druk van 1 atm.

30 14. Werkwijze volgens conclusie 13, waarbij de schuimsamenstelling een watervrij mengsel is.

35 15. Gebruik van een niet-verknoopt thermisch isolatieschuim op polyolefinebasis volgens één van de conclusies 1 tot 12, voor thermische isolatie.

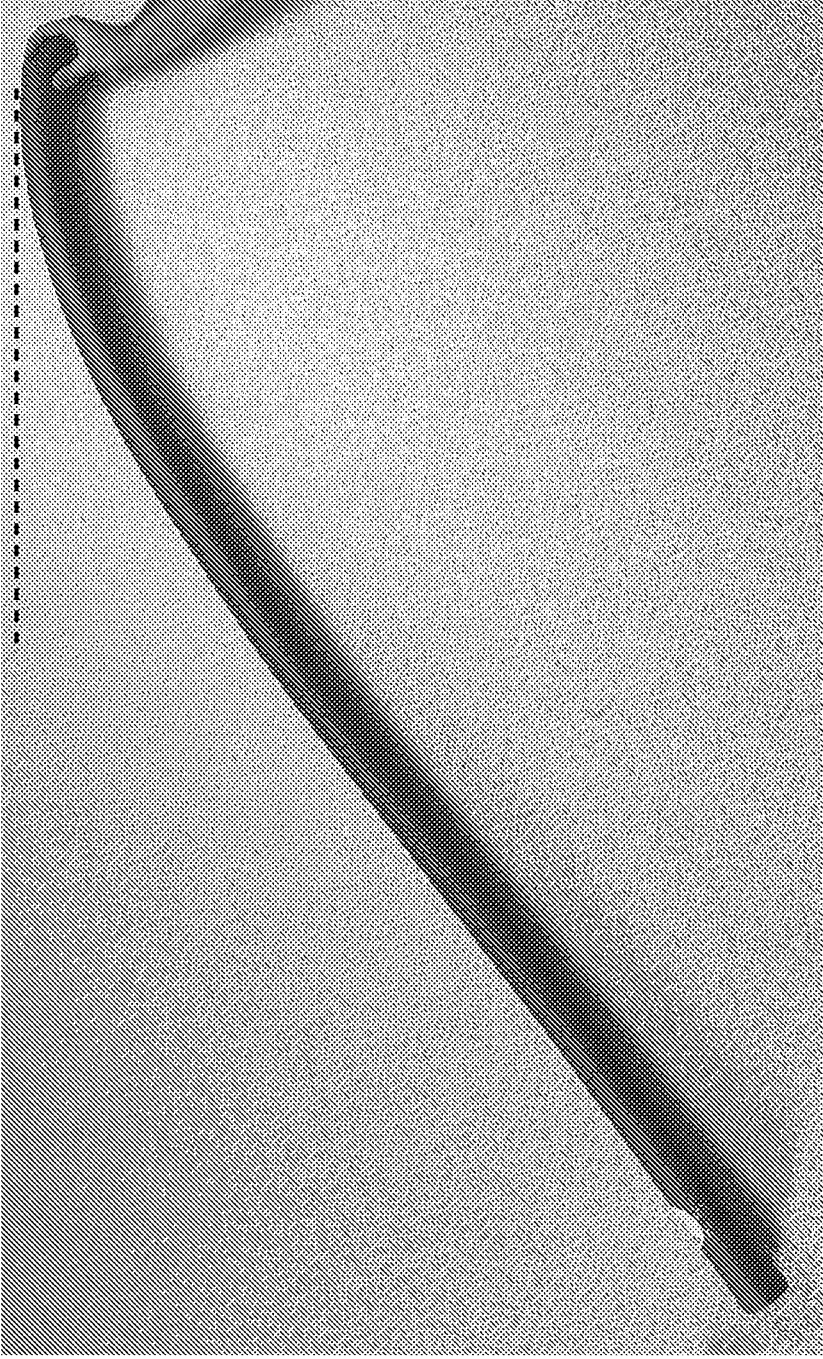


FIG. 1



FIG. 2

**ABSTRACT**

The present invention relates to a polyolefin thermal insulation foam and use thereof, and to a method for preparing a physically foamed polyolefin thermal insulation foam, which can be recycled well and which has excellent flexibility characteristics.

5

# SAMENWERKINGSVERDRAG (PCT)

## RAPPORT BETREFFENDE NIEUWHEIDSONDERZOEK VAN INTERNATIONAAL TYPE

IDENTIFICATIE VAN DE NATIONALE AANVRAGE	KENMERK VAN DE AANVRAGER OF VAN DE GEMACHTIGDE
	<b>NLP201747</b>
Nederlands aanvraag nr.	Indieningsdatum
<b>2019501</b>	<b>07-09-2017</b>
	Ingeroepen voorrangsdatum
Aanvrager (Naam)	
<b>Thermaflex International Holding B.V.</b>	
Datum van het verzoek voor een onderzoek van internationaal type	Door de instantie voor Internationaal Onderzoek aan het verzoek voor een onderzoek van internationaal type toegekend nr.
<b>23-09-2017</b>	<b>SN69696</b>
I. CLASSIFICATIE VAN HET ONDERWERP (bij toepassing van verschillende classificaties, alle classificatiesymbolen opgeven)	
Volgens de internationale classificatie (IPC)	
<b>C08L53/00;C08L23/04;C08L23/08;C08K5/103;C08K5/20;C08J9/00</b>	
II. ONDERZOCHETE GEBIEDEN VAN DE TECHNIEK	
Onderzochte minimumdocumentatie	
Classificatiesysteem	Classificatiesymbolen
<b>IPC</b>	<b>C08L;C08J</b>
Onderzochte andere documentatie dan de minimum documentatie, voor zover dergelijke documenten in de onderzochte gebieden zijn opgenomen	
III. <input type="checkbox"/>	GEEN ONDERZOEK MOGELIJK VOOR BEPAALDE CONCLUSIES (opmerkingen op aanvullingsblad)
IV. <input type="checkbox"/>	GEBREK AAN EENHEID VAN UITVINDING (opmerkingen op aanvullingsblad)

**ONDERZOEKSRAPPORT BETREFFENDE HET  
RESULTAAT VAN HET ONDERZOEK NAAR DE STAND  
VAN DE TECHNIEK VAN HET INTERNATIONALE TYPE**

Nummer van het verzoek om een onderzoek naar  
de stand van de techniek

NL 2019501

<p>A. CLASSIFICATIE VAN HET ONDERWERP INV. C08L53/00 C08L23/04 C08L23/08 C08K5/103 C08K5/20 C08J9/00</p> <p>ADD.</p> <p>Volgens de Internationale Classificatie van octrooien (IPC) of zowel volgens de nationale classificatie als volgens de IPC.</p>										
<p>B. ONDERZOCHETE GEBIEDEN VAN DE TECHNIEK</p> <p>Onderzochte minimum documentatie (classificatie gevolgd door classificatiesymbolen) C08L C08J</p> <p>Onderzochte andere documentatie dan de minimum documentatie, voor dergelijke documenten, voor zover dergelijke documenten in de onderzochte gebieden zijn opgenomen</p> <p>Tijdens het onderzoek geraadpleegde elektronische gegevensbestanden (naam van de gegevensbestanden en, waar uitvoerbaar, gebruikte trefwoorden) EPO-Internal, WPI Data</p>										
<p>C. VAN BELANG GEACHTE DOCUMENTEN</p> <table border="1"> <thead> <tr> <th>Categorie *</th> <th>Geciteerde documenten, eventueel met aanduiding van speciaal van belang zijnde passages</th> <th>Van belang voor conclusie nr.</th> </tr> </thead> <tbody> <tr> <td>A, D</td> <td> <p>WO 02/42679 A1 (THERMAFLEX INTERNAT HOLDING B [NL]; VEN EMANUEL JOZEPH HERMAN MARI [NL] 30 mei 2002 (2002-05-30) in de aanvraag genoemd * conclusie 1 * * bladzijde 1, regel 1 - regel 4 *</p> </td> <td>1-15</td> </tr> <tr> <td>A</td> <td> <p>EP 2 070 976 A1 (NITTO DENKO CORP [JP]) 17 juni 2009 (2009-06-17) * conclusie 1 * * voorbeelden 1, vergelijkend voorbeeld 1 * * alinea's [0001], [0002], [0011] - [0013], [0017] - [0019] * ----- -/--</p> </td> <td>1-15</td> </tr> </tbody> </table>		Categorie *	Geciteerde documenten, eventueel met aanduiding van speciaal van belang zijnde passages	Van belang voor conclusie nr.	A, D	<p>WO 02/42679 A1 (THERMAFLEX INTERNAT HOLDING B [NL]; VEN EMANUEL JOZEPH HERMAN MARI [NL] 30 mei 2002 (2002-05-30) in de aanvraag genoemd * conclusie 1 * * bladzijde 1, regel 1 - regel 4 *</p>	1-15	A	<p>EP 2 070 976 A1 (NITTO DENKO CORP [JP]) 17 juni 2009 (2009-06-17) * conclusie 1 * * voorbeelden 1, vergelijkend voorbeeld 1 * * alinea's [0001], [0002], [0011] - [0013], [0017] - [0019] * ----- -/--</p>	1-15
Categorie *	Geciteerde documenten, eventueel met aanduiding van speciaal van belang zijnde passages	Van belang voor conclusie nr.								
A, D	<p>WO 02/42679 A1 (THERMAFLEX INTERNAT HOLDING B [NL]; VEN EMANUEL JOZEPH HERMAN MARI [NL] 30 mei 2002 (2002-05-30) in de aanvraag genoemd * conclusie 1 * * bladzijde 1, regel 1 - regel 4 *</p>	1-15								
A	<p>EP 2 070 976 A1 (NITTO DENKO CORP [JP]) 17 juni 2009 (2009-06-17) * conclusie 1 * * voorbeelden 1, vergelijkend voorbeeld 1 * * alinea's [0001], [0002], [0011] - [0013], [0017] - [0019] * ----- -/--</p>	1-15								
<p><input checked="" type="checkbox"/> Verdere documenten worden vermeld in het vervolg van vak C. <input checked="" type="checkbox"/> Leden van dezelfde octroofamilie zijn vermeld in een bijlage</p>										
<p>* Speciale categorieën van aangehaalde documenten</p> <p>*A* niet tot de categorie X of Y behorende literatuur die de stand van de techniek beschrijft</p> <p>*D* in de octrooiaanvraag vermeld</p> <p>*E* eerdere octrooiaanvraag, gepubliceerd op of na de indieningsdatum, waarin dezelfde uitvinding wordt beschreven</p> <p>*L* om andere redenen vermeldde literatuur</p> <p>*O* niet-schriftelijke stand van de techniek</p> <p>*P* tussen de voorrangdatum en de indieningsdatum gepubliceerde literatuur</p> <p>*T* na de indieningsdatum of de voorrangdatum gepubliceerde literatuur die niet bezwarend is voor de octrooiaanvraag, maar wordt vermeld ter verheldering van de theorie of het principe dat ten grondslag ligt aan de uitvinding</p> <p>*X* de conclusie wordt als niet nieuw of niet inventief beschouwd ten opzichte van deze literatuur</p> <p>*Y* de conclusie wordt als niet inventief beschouwd ten opzichte van de combinatie van deze literatuur met andere geciteerde literatuur van dezelfde categorie, waarbij de combinatie voor de vakman voor de hand liggend wordt geacht</p> <p>*Z* lid van dezelfde octroofamilie of overeenkomstige octrooipublicatie</p>										
<p>Datum waarop het onderzoek naar de stand van de techniek van internationaal type werd voltooid</p> <p>10 april 2018</p>	<p>Verzenddatum van het rapport van het onderzoek naar de stand van de techniek van internationaal type</p>									
<p>Naam en adres van de instantie</p> <p>European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016</p>	<p>De bevoegde ambtenaar</p> <p>van Bergen, Marc</p>									

**ONDERZOEKSRAPPORT BETREFFENDE HET  
 RESULTAAT VAN HET ONDERZOEK NAAR DE STAND  
 VAN DE TECHNIEK VAN HET INTERNATIONALE TYPE**

Nummer van het verzoek om een onderzoek naar  
 de stand van de techniek  
**NL 2019501**

**C (Vervolg). VAN BELANG GEACHTE DOCUMENTEN**

Categorie	Geciteerde documenten, eventueel met aanduiding van speciaal van belang zijnde passages	Van belang voor conclusie nr.
X	WO 2006/102151 A1 (DOW GLOBAL TECHNOLOGIES INC [US]; MARTINEZ FELIPE [US]; CHEUNG YUNWA W) 28 september 2006 (2006-09-28) * voorbeelden 23,23P,24-29 * * tabellen 8,9A * * alinea's [0001], [0020], [0045], [0046], [0052], [0269], [0272] - [0273] * -----	1-15

**ONDERZOEKSRAPPORT BETREFFENDE HET  
RESULTAAT VAN HET ONDERZOEK NAAR DE STAND  
VAN DE TECHNIEK VAN HET INTERNATIONALE TYPE**

Informatie over leden van dezelfde octrooifamilie

Nummer van het verzoek om een onderzoek naar  
de stand van de techniek

NL 2019501

In het rapport gencenrid octrooigeschrift	Datum van publicatie	Overeenkomend(e) geschrift(en)	Datum van publicatie
WO 0242679	A1	30-05-2002	AT 288049 T 15-02-2005
			AU 2557001 A 03-06-2002
			CN 1461393 A 10-12-2003
			DE 60017807 D1 03-03-2005
			DE 60017807 T2 05-01-2006
			DK 1336064 T3 30-05-2005
			EP 1336064 A1 20-08-2003
			ES 2236030 T3 16-07-2005
			HK 1058816 A1 09-09-2005
			JP 2004514747 A 20-05-2004
			PL 360918 A1 20-09-2004
			PT 1336064 E 30-06-2005
			TR 200300730 T2 23-08-2004
			UA 75618 C2 15-08-2003
			US 2003232898 A1 18-12-2003
WO 0242679 A1 30-05-2002			
EP 2070976	A1	17-06-2009	AT 556109 T 15-05-2012
			CN 101522769 A 02-09-2009
			EP 2070976 A1 17-06-2009
			JP 5153110 B2 27-02-2013
			JP 2008088283 A 17-04-2008
			KR 20090073127 A 02-07-2009
			TW 200838912 A 01-10-2008
			US 2010016458 A1 21-01-2010
WO 2008041617 A1 10-04-2008			
WO 2006102151	A1	28-09-2006	AR 053830 A1 23-05-2007
			AT 507261 T 15-05-2011
			AU 2006227349 A1 28-09-2006
			BR PI0609833 A2 27-04-2010
			CA 2601286 A1 28-09-2006
			CN 101313015 A 26-11-2008
			EP 1858960 A1 28-11-2007
			JP 4881370 B2 22-02-2012
			JP 2008538377 A 23-10-2008
			KR 20070119642 A 20-12-2007
			TW 1375684 B 01-11-2012
			WO 2006102151 A1 28-09-2006

WRITTEN OPINION

File No. SN69696	Filing date (day/month/year) 07.09.2017	Priority date (day/month/year)	Application No. NL2019501
International Patent Classification (IPC) INV. C08L53/00 C08L23/04 C08L23/08 C08K5/103 C08K5/20 C08J9/00			
Applicant Thermaflex International Holding B.V.			

This opinion contains indications relating to the following items:

- Box No. I Basis of the opinion
- Box No. II Priority
- Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- Box No. IV Lack of unity of invention
- Box No. V Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- Box No. VI Certain documents cited
- Box No. VII Certain defects in the application
- Box No. VIII Certain observations on the application

	Examiner van Bergen, Marc
--	------------------------------

**WRITTEN OPINION****Box No. I Basis of this opinion**

1. This opinion has been established on the basis of the latest set of claims filed before the start of the search.
2. With regard to any **nucleotide and/or amino acid sequence** disclosed in the application and necessary to the claimed invention, this opinion has been established on the basis of:
  - a. type of material:
    - a sequence listing
    - table(s) related to the sequence listing
  - b. format of material:
    - on paper
    - in electronic form
  - c. time of filing/furnishing:
    - contained in the application as filed.
    - filed together with the application in electronic form.
    - furnished subsequently for the purposes of search.
3.  In addition, in the case that more than one version or copy of a sequence listing and/or table relating thereto has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.
4. Additional comments:

**Box No. V Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

## 1. Statement

Novelty	Yes: Claims	3, 4, 6, 7, 11-14
	No: Claims	1, 2, 5, 8-10, 15
Inventive step	Yes: Claims	
	No: Claims	1-15
Industrial applicability	Yes: Claims	1-15
	No: Claims	

## 2. Citations and explanations

see separate sheet

**WRITTEN OPINION**

Application number

NL2019501

---

**Box No. VII Certain defects in the application**

see separate sheet

---

**Box No. VIII Certain observations on the application**

see separate sheet

Re Item V

Reference is made to the following documents:

D1: WO 02/42679 A1 (THERMAFLEX INTERNAT HOLDING B [NL]; VEN EMANUEL JOZEPH HERMAN MARI [NL]) 30 mei 2002 (2002-05-30) in de aanvraag genoemd

D2: EP 2 070 976 A1 (NITTO DENKO CORP [JP]) 17 juni 2009 (2009-06-17)

D3: WO 2006/102151 A1 (DOW GLOBAL TECHNOLOGIES INC [US]; MARTINEZ FELIPE [US]; CHEUNG YUNWA W) 28 september 2006 (2006-09-28)

1. Novelty

1.1 D1 relates to pipe insulation foams based on polyolefins (page 1, lines 1-4). The foam composition comprises 77-92 % by weight of a metallocene polyethylene, 5-18 % by weight of a flame extinguisher, 3-8 % by weight of a cell stabilizer and 0-5 % by weight of other usual foam additives (claim 1).

The subject matter of claim 1 differs from D1 in that the foam composition comprises an ethylene alpha-olefin block copolymer as defined under point i) of claim 1. D1 is silent about block copolymers.

The subject matter of claims 1-15 is therefore novel over D1.

1.2 D2 relates to polyolefin resin foams suitable for use as heat insulating materials (paragraphs 1-2). The polyolefin resin of D2 comprises (A) a rubber and/or a thermoplastic elastomer, (B) a polyolefin resin, and (C) at least one aliphatic compound selected from an aliphatic acid, an aliphatic acid amide and an aliphatic acid metallic soap, said compound having a polar functional group and having a melting point of 50 to 150°C, wherein a content of the aliphatic compound is 1 to 5 parts by weight based on 100 parts by weight of the total amount of the rubber and/or thermoplastic elastomer and the polyolefin resin (claim 1).

Component (A) may be an ethylene polymer (paragraphs 11-13).

Component (B) may be a block copolymer and said block copolymer may comprise ethylene derived units and units derived from higher alpha-olefins (paragraphs 17-19).

D2 does not disclose a foam composition comprising an ethylene alpha-olefin block copolymer as defined under point i) of claim 1, let alone a composition comprising such a block copolymer in combination with a polyethylene as defined under point ii) of D2.

Example 1 and comparative example 1 of D2 do not contain an ethylene alpha-olefin block copolymer either.

The subject matter of claims 1-15 is therefore novel over D2.

1.3 D3 relates to soft foams comprising at least one ethylene alpha-olefin copolymer suitable for use as thermal insulation material (paragraph 1). Said ethylene alpha-olefin copolymer may be a block copolymer (paragraphs 20, 45, 46, 52). Said block copolymers typically comprise "hard" segments which are blocks of polymerized units in which ethylene is present in an amount greater than 95 wt% and "soft" segments which are blocks of polymerized units in which the comonomer content (monomers other than ethylene) is greater than 5 wt% (paragraph 46).

Examples 23, 23P and 24-29 (paragraphs 272-273) disclose foam compositions comprising an ethylene alpha-olefin block copolymer (polymers 19a and 19b), a low density polyethylene (LDPE 620i) and glycerol monostearate (paragraph 269) which is also used in the present application as cell stabilizer. The compositions also comprise stabilizers and nucleating agents (paragraph 269).

Since block polymers 19a and 19b are inventive examples of D3 (table 9A), it can be assumed that they have the properties as described in paragraph 46.

It can be assumed that LDPE 620i has a density between 0.880 and 0.960 g/cm<sup>3</sup>.

The alpha-olefin comonomer in polymers 19a and 19b is octene (table 8).

Ethylene alpha-olefin block copolymer and LDPE are used in different weight ration in the above mentioned examples.

D3 discloses the use of the foams disclosed therein as thermal insulating material (paragraph 1).

Hence, D3 discloses all the technical features of claims 1, 2, 5, 8-10 and 15. The subject matter of these claims is therefore not novel over D3.

## 2. Inventive step

2.1 D3 may be considered as closest prior art to the subject matter of claim 1 (see 1.3).

D3 already anticipates the subject matter of claims 1, 2, 5, 8-10 and 15. The subject matter of these claims is therefore inventive either.

Dependent claims 3, 4, 6, 7, 11 and 12 do not appear to contain any additional features which, in combination with the features of any claim to which they refer, meet the requirements of novelty and/or inventive step, unless any surprising advantageous technical effect associated with any of the additional features can be shown. Most probably, the ethylene alpha-olefin block copolymers of D3 fulfil the requirements of dependent claims 3 and 4. The use of more than one polyethylene polymer does not appear inventive either. The same applies to dependent claims 11 and 12.

Even if it could be shown that block copolymers 19a and 19b of D3 do not fulfil all the requirements as defined in point i) of claim 1, an insulation foam according to claim 1 would not be considered inventive in view of D3 (see paragraph 46).

The process for producing the claimed insulation foam according to claims 13 and 14 does not appear inventive either in the absence of a surprising advantageous technical effect associated with any of the process features defined in these two claims. D3 also discloses the use of a blowing agent and the process is carried out in the absence of water (paragraph 269).

#### **Re Item VII**

The relevant background art disclosed in D2 and D3 is not mentioned in the description, nor are these documents identified therein.

#### **Re Item VIII**

The unit "atm" employed in claim 13 is not additionally expressed in terms of the metric system.