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(54) Title: EXTRUDED POLYOLEFIN FOR THE MANUFACTURE OF CELLULAR MATERIAL

(57) Abstract: Extruded polyolefin characterized by a high foamability number compared to known polyolefins suitable for foam applications.

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Extruded Polyolefin for the Manufacture of Cellular Material

The present invention relates to extruded polyolefin, in particular extruded polyolefin beads suitable for technical foam applications and protective
5 packaging. Furthermore, the present invention is related to the use of the extruded polyolefin for foamed articles.

In the recent years, there have been efforts undertaken to replace foamed polystyrene by foamed polypropylene products. It is known that foamed
10 polypropylene products possess chemical inertia, heat resistance, stiffness and impact resistance characteristics far superior to those of foamed polystyrene. Therefore, there is an increasing demand using foamed polypropylene in application fields where the foamed polystyrene cannot be used or in fields where the foamed polystyrene is widely used, such as for exam-
15 ple in insulation and in packaging, but does not completely satisfy the demands of the uses in terms of performance. For the technical foam application and the protecting packaging, the natural requirements for the used polymers are their foamability, high stiffness and high impact strength. One essential requirement that an extruded polyolefin achieves these desired
20 requirements is a relatively low density of the foamed products. Especially foamed articles with a high impact strength and being simultaneously rather light, are achieved in cases where these products have only a closed cell structure. There is a prejudice in literature that these products can only be achieved when the used extruded polyolefin has a high melt strength.

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Therefore, it is the object of the present invention to provide an extruded polyolefin suitable for technical foam applications and protective packag-
ing which are an alternative to the known products characterized by a high melt strength.

30

The finding of the present invention is to provide an extruded polyolefin with a high foamability number.

Hence, the present invention is related to an extruded polyolefin having

5

- a) MFR₂ of 0.01 to 1.00 g/10 min and
- b) a foamability number (FN) of higher than 0.25 N, whereby the foamability number is defined by the integral $\int_{\lambda=0}^{\lambda_{\max}} F \cdot d\lambda$, wherein λ is the stretch ratio and F is the force given in cN recorded as a function of the stretch ratio λ .

10

Surprisingly, it has been found that such an extruded polyolefin possesses a good foamability, high stiffness and high impact strength.

- 15 It has been in particular found that there is a correlation of minimum foam density and foamability number FN, i.e. with a high foamability number, very low foam density articles can be obtained (see Fig. 2).

The foamability number is determined from the area under the Rheotens-curve, i.e. the integral defined by the force given in cN as a function of the stretch ratio λ . The Rheotens test, as used for this invention, to obtain the Rheotens-curve as inter alia given in Figure 1 is explained in detail in the experimental section. Preferably, the foamability number (FN) is higher than 0.3 N, more preferably higher than 0.4 N and most preferably higher than 0.45 N. In a preferred embodiment, the foamability number is 0.5 N or higher.

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However, not only the foamability number of the extruded polyolefin must be rather high but also the molecular weight of the extruded polyolefin must be in the range as given above. More preferably, the MFR₂ is in the range of 0.1 to 0.6

g/10 min, still more preferably in the range of 0.2 to 0.4. In a preferred embodiment, the MFR_2 is of about 0.25 g/10 min. The melt flow rate (MFR) is an indicator of the molecular weight of the polymer used. The melt flow rate mainly depends on the average molecular weight. This is due to the fact that long molecules render the material a lower flow tendency than short molecules. An increase in molecular weight means a decrease in the MFR-value. The melt flow rate (MFR) is measured in g/10 min of the polymer discharged through a defined dye under specified temperature and pressure conditions and the measure of the viscosity of the polymer, which, in turn, for each type of polymer is mainly influenced by its molecular weight but also by its degree of branching. The melt flow rate measured under a load of 2.16 kg at 230°C (ISO 1133) is denoted as MFR_2 .

Preferably, the extruded polyolefin has a melt strength lower than 3 cN. More preferably, the melt strength is lower than 2 cN and most preferred, the melt strength is lower than 1.6. In a preferred embodiment, the melt strength is about 1.5 cN.

The advantage of having low melt strength is (a) such polymers are easy to make in reactor processes without modification and (b) the cell nucleation is more pronounced, i.e. the foam is more fine-cellular (see P. Spitael and Ch. W. Macosko, Polymer Engineering and Science, November 2004, Vol. 44, No. 11).

The melt strength is measured by the Rheotens experiment. The Rheotens experiment is a quasi-isothermal fiber spinning experiment. A polymer melt pressured in a capillary dye is stretched under the action of a constant draw-down force until rupture of the filament. With the data obtained from such an experiment an extension diagram can be generated which describes the elongational behavior of the polymer melt. A good elongational behavior is needed for many polymer processes like blow molding or film blowing. Also the rupture stress of a polymer melt can be calculated with the data obtained from the Rheotens ex-

periment. A high rupture stress is of importance for many industrial applications. In general, the melt strength and the drawability depend on the material properties of the melt and on the processing conditions of the experiment. The exact conditions under which the extruded polyolefin according to this invention have
5 been measured can be deducted from the experimental section.

Moreover, it is preferred that the extruded polyolefin has a crystallization temperature T_c in the range of 110 to 135°C, more preferably, in the range of 115 to 130°C. The crystallization temperature T_c has been measured with DSC accord-
10 ing to ISO 3146 with a heating/cooling rate of 10°C. The advantage of a polymer with a crystallization temperature in the range of 110 to 135°C, more preferably in the range of 115 to 130°C is that it has been found that these polymers are particularly stiff.

15 In addition, it is preferred that the foamed articles of the extruded polyolefin shall have a rather high crystallinity. A highly structured (crystal) polymer has improved optical and mechanical properties.

Therefore, the extruded polyolefin shall preferably comprise an α -crystallization
20 nucleating agent and/ or a β -crystallization nucleating agent, whereby the crystallization nucleating agent is not from a metal salt. Preferably, the nucleating agent is present in an amount of 0.0001 to 1.00 wt%, more preferably in the amount of 0.001 to 0.50 wt%. Nucleating agents of polyolefin are substances with the ability to act as substrates for crystal formation by epitaxy thus increasing the num-
25 ber of nuclei of the system. Thereby, the nucleating agents improve the crystallization behavior in processing, i.e. decreasing the cycle time or increasing the line speed, increase the crystallinity in the final product and, additionally, influence the optical and mechanical properties in terms of improved stiffness (with α -crystallization nucleating agents) or toughness (with β -crystallization nucleating
30 agents).

Any suitable α -nucleating agents known in the art can be used. However, it is preferred to use 1,3:2,4-bis-(3,3-dimethyl-benzylidene)-sorbitol. A further α -nucleation method, herein referred to as "BNT" is a special reactor technique, where the catalyst is pre-polymerized with monomers like vinyl-cyclohexan (VCH). This method is described in greater detail in e.g. EP 0 316 187 A2. For the purpose of this invention, BNT is referred to as an α -nucleating agent.

10 In particular suitable β -nucleation agents are quinacridone type compounds, especially quinacridone, dimethylquinacridone and dimethoxyquinacridone; quinacridonequinone type compounds, especially quinacridonequinone, a mixed crystal of 5,12-dihydro(2,3b)acridine-7,14-dione with quino(2,3b)acridine-6,7,13,14-(5H,12H)-tetrone as disclosed in EP 0 177 961 and dimethoxyquinacridonequinone; and dihydroquinacridone type compounds, especially dihydroquinacridone, di-methoxydihydroquinacridone and dibenzodihydroquinacridone.

Still further suitable β -nucleating agents are dicarboxylic acid salts of metals from group IIa of periodic table, e.g. pimelic acid calcium salt and suberic acid calcium salt.

Preferably, the extruded polyolefin is a polypropylene. Even more preferred, the polyolefin is a linear polypropylene, i.e. having a branching index g' of 0.95 to 1.00, more preferable of about 1.00. The branching index g' is defined as $g' = [\text{IV}]_{\text{br}} / [\text{IV}]_{\text{lin}}$ in which g' is the branching index, $[\text{IV}]_{\text{br}}$ is the intrinsic viscosity of the branched polypropylene and $[\text{IV}]_{\text{lin}}$ is the intrinsic viscosity of the linear polypropylene having substantially the same weight average molecular weight as the branched polypropylene. Thereby, a low g' -value is an indicator for a high branched polymer. In other words, if the

g'-value decreases, the branching of the polypropylene increases. Reference is made in this context to B.H. Zimm and W.H. Stockmeyer, J. Chem. Phys. 17,1301 (1949). This document is herewith included by reference.

- 5 The intrinsic viscosity needed for determining the branching index g' is measured according to DIN ISO 1628/1, October 1999 (in Decalin at 135°C)

10 It is in particular preferred that the extruded polyolefin comprises a propylene matrix and an ethylene-propylene rubber (EPR). The type of polypropylene matrix is not restricted to a specific one, as long as the matrix fulfills, together with the ethylene-propylene rubber (EPR), the above-mentioned requirements. Hence, each homopolymer or copolymer can be used. In case a copolymer is used, the comonomer content, however, shall
15 be less than 3 wt%. Preferably, the comonomer is ethylene.

The choice of ethylene-propylene rubber (EPR) must conform with the requirements to be achieved, i.e. an extruded polyolefin with a rather high foamability number. Hence, an ethylene-propylene rubber (EPR) is needed
20 having a high propylene amount, i.e. with a low amount of ethylene. Therefore, it is preferred that the ethylene-propylene rubber (EPR) has an ethylene content of lower than 50.0 wt%, more preferably in the range of 10.0 to 50.0 wt% and most preferably, in the range of 20.0 to 40.0 wt%. In a preferred embodiment, the ethylene content is about 33.0 wt%.

25

Furthermore, the total amount of ethylene used in the polypropylene, more preferably in the polypropylene comprising propylene matrix and the ethylene-propylene rubber (EPR), shall be preferably less than 15.0 wt%, more preferably in the range of 4.0 to 12.0 wt%, still more preferably in the

range of 5.0 to 10.0 wt%. In a preferred embodiment, the total ethylene content is about 9.0 wt%.

Furthermore, to achieve an extruded polypropylene suitable for the manufacture of an article having a low foaming density, the polypropylene matrix and the ethylene-propylene rubber (EPR) must be kept in a specific range. Accordingly, the extruded polypropylene does not have more than 30 wt% of the ethylene-propylene rubber (EPR). Therefore, the amount of polypropylene matrix must range from 70 to 95 wt%, preferably in the range of 70 to 85 wt%. In turn, the ethylene-propylene rubber (EPR) must be in the range of 5 to 30 wt%, preferably in the range of 15 to 30 wt%.

Further, the extruded polyolefin, in particular the extruded polypropylene as described above, is in a form of beads.

15

The extruded polypropylene is preferably produced by combining the polypropylene matrix in the form of powder or granules, the ethylene propylene rubber (EPR) and the crystallization nucleating agent, if present, in a melt mixing device.

20

Melt mixing devices suitable for the preferred process are discontinuous and/or continuous kneaders, twin screw extruders and single screw extruders with special mixing sections and co-kneaders. The residence time must be chosen such that a sufficiently high degree of homogenization is achieved.

25

The polypropylene matrix may be produced by a single or multi-stage process polymerization of propylene or propylene and an α -olefin and/or ethylene such as bulk-polymerization, gas phase polymerization, slurry polymerization, solution polymerization or combinations thereof using con-

30

ventional catalysts. A homo- or copolymer can be either made in a loop reactor or in a combination of loop and gas phase reactor. Those processes are well-known to one skilled in the art.

- 5 A suitable catalyst for the polymerization of the propylene polymer matrix is any stereo-specific catalyst for propylene polymerization which is capable of polymerizing and copolymerizing propylene and comonomers at a temperature of 40 to 100°C and at a pressure from 10 to 100 bar. Ziegler-Natta catalysts as well as metallocene catalysts are suitable catalysts.

10

One skilled in the art is aware of the various possibilities to produce propylene homo- and copolymers and will simply find out a suitable procedure to produce suitable polymers which are used in the present invention.

- 15 An ethylene-propylene rubber (EPR) may be produced by a known polymerization process such as solution, suspension and gas phase polymerization using conventional catalysts. Ziegler-Natta catalysts as well as a metallocene catalyst are suitable catalysts.

- 20 A widely used process is the solution polymerization. Ethylene, propylene and catalyst systems are polymerized in an excess of hydrocarbon solvent. Stabilizers and oils, if used, are added directly after polymerization. The solvent and unreacted monomers are then flashed off with hot water or steam, or with mechanical devolatilization. The polymer, which is in crumb
25 form, is dried with de-watering in screens, mechanical pressures or drying ovens. The crumb is formed into wrapped balls or extruded into pellets.

The suspension polymerization process is a modification of bulk polymerization. The monomers and catalyst system are injected into the reactor

filled with propylene. The polymerization takes place immediately, forming crumbs of polymer that are not soluble in propylene.

5 Flashing off the propylene and comonomer complete the polymerization process.

The gas phase polymerization technology consists of one or more vertical fluidized beds. Monomers and nitrogen in gas form along with catalyst are fed to the reactor and solid product is removed periodically. Heat of reaction is removed through the use of the circulating gas that also serves to fluidize the polymer bed. Solvents are not used thereby eliminating the need for solvent stripping, washing and drying.

15 The production of ethylene-propylene rubber (EPR) is also described in detail in e.g. US 3,300,459, US 5,919,877, EP 0 060 090 A1 and in a company publication by EniChem "DUTRAL, Ethylene-propylene Elastomers", pages 1 to 4 (1991).

20 Alternatively, ethylene-propylene rubber (EPR), which is commercially available and which fulfills the indicated requirements, can be used.

Alternatively, the propylene polymer matrix and the ethylene-propylene rubber may be produced in a series of reactors, i.e. starting with the production of the propylene matrix in a loop reactor and transferring the production into a gas phase reactor where the ethylene-propylene rubber is polymerized.

30 Furthermore, the present invention also comprises a foam produced from the extruded polyolefin as described above or from extruded polyolefin bead as mentioned above. Thereby, it is preferred that the foam has a den-

sity lower than 400 g/m^3 , more preferably lower than 250 g/m^3 and most preferred lower than 180 kg/m^3 .

Additionally, the present invention is concerned with articles comprising
5 the extruded polyolefin and/or extruded polyolefin beads and/or a foam as defined above. Furthermore, the invention is also concerned with the use of the extruded polyolefin as defined above to produce the foamed articles.

Examples

10 Methods

Foamability Number:

In order to measure the Foamability Number, a high pressure capillary rheometer (Type: Rheograph 2002, Göttfert Werkstoff-Prüfmaschinen
15 GmbH, Buchen, Germany) was used to extrude polymer strands which were stretched by means of a Rheotens melt tension apparatus (Type: 7197, Göttfert Werkstoff-Prüfmaschinen GmbH). The haul-off force F in dependence of draw-down velocity v is recorded. The test procedure is performed in a standard climatized room with controlled room temperature of 23°C .
20 The temperature of the rheometer was set at 200°C . A capillary of 20mm length and 1 mm diameter was used. The acceleration of the wheels of the Rheotens was set to 0.012 cm/s^2 for all the measurements. The diameter of the barrel was 12mm. The polymer was allowed to melt for 4 minutes. The extrusion rate of the strand is 0.0217 mm/s (equivalent to an apparent shear rate
25 of 25s^{-1}). The length of the spinning line was 104mm. The haul-off force F (in N) is measured against the stretch ratio λ . This way a curve is obtained. The maximum force is called "Melt Strength". The area under curve of the force vs. stretch ratio until break of the fiber is taken as Foamability Number (FN).

Density:

The density has been measured according to the Archimedes principle
5 through determining mass (m) and volume (V) of the specimen and calculating its density (d) accordingly ($d=m/V$).

$$d = \frac{m}{V}$$

Tensile modulus:

10

Tensile properties such as tensile modulus, tensile stress at yield, tensile strain at yield, tensile strength, tensile strain at tensile strength, and tensile strength at break have been measured analogue to ISO 527 in extrusion direction.

15

Foaming with Butane:

A tandem foam extrusion setup is used in this study. It consists of a 5-hp extruder drive with a speed control gearbox (Brabender, Prep Center), a
20 first 3/4" extruder (Brabender, 05-25-000) with a mixing screw (Brabender, 05-00144) of 30:1 L/D ratio, a second 1 1/2 " extruder (Killion, KN-150) with a built-in 15-hp variable speed drive unit with a 18:1 L/D ratio. The other systems include a positive displacement pump for butane injection, a diffusion enhancing device containing static mixer (omega, FMX-84441S),
25 a gear pump (Zenith, PEP-11 1.2 cc/rev) where the volumetric displacement is properly controlled by the motor, a heat exchanger for cooling the polymer melt that contains homogenizing static mixers (Labcore Model H-04669-12), a cooling sleeve for the precise control of die temperature. The first extruder is used for the plastication of the polymer resin while the sec-

ond extruder is responsible for mixing and initial cooling of the polymer melt. The gear pump controls the polymer melt flow rate and the heat exchanger further homogenizes and cools the melt. Two filamentary dies ($L/D = 0.3/0.040$ and $0.5/0.040$ inch) were used in this study. The polypropylene material pellets blended with 0.8 wt% talc were first fed into the barrel through the hopper and were completely melted by the screw rotation. Then a metered amount of butane was injected into the extrusion barrel by a positive displacement pump and mixed intensively with the polymer melt stream. When the gas was injected into the extrusion barrel, screw generated shear fields to completely dissolve gas in the polymer melt via convective diffusion. The single-phase polymer/gas solution went through the gear pump and was fed into the heat exchanger where it was cooled to a designated temperature. The cooled polymer/gas solution entered the die and foaming occurred at the die exit through a process of thermodynamic instability induced via rapid pressure drop. The melt and die temperatures were synchronized and adjusted to 170°C.

Foaming with Carbon Dioxide:

A single screw foam extrusion setup was used. Extrusion experiments have been carried out using a single screw extruder (Barmag AG, Remscheid, Germany) with a diameter of 60 mm and a processing length of 40 D. It is equipped with a screw which is designed especially for the use in foam extrusion (Berstorff GmbH, Hannover, Germany). After melting and homogenising the polymer, liquid CO₂ is injected into the extruder barrel using a dual head membrane pump (LEWA GmbH, Leonberg, Germany) at a length of 16 D. The last 11 D of the process length are designed as a water tempered cylinder extension, which ensures an effective cooling of the melt and a precise control of the melt temperature. To improve the homogeneity of the polymer/gas solution, an additional static mixer (SMX-75, Sulzer Chemtech, Winterthur, Switzerland) is mounted between extruder and ex-

trusion die. The melt is thermostated to 175°C and formed out using an annular die with an outlet diameter of 50 mm. The cellular extrudate is calibrated by pulling it over a cooling mandrel with a diameter of 175 mm, followed by slitting and winding. In order to improve the foam structure, a fixed amount of nucleating agent has been added - as is normally done in industrial foam production (Hydrocerol CF20E, Clariant Masterbatch GmbH, Ahrensburg, Germany).

Example 1 (Comparison)

- 10 An essentially linear polypropylene homopolymer with a MFR of 3.1g/10min, a melt strength of 0.020 N (2 cN) and a Foamability Number FN=0.1 N is extruded in contact with 5wt% butane. The final density of the cellular material is 450 kg/m³. Low density foam is not achieved.
- 15 The foamability number of regrind material has been found to be identical to the raw material.

Example 2 (Comparison)

- 20 An essentially linear polyolefin material as used in example 1 with a melt strength of 0.02 N (2 cN) and a Foamability Number FN=0.1 N is extruded in contact with 0.80wt% carbon dioxide. The final density of the cellular material is 420kg/m³. Low density foam is not achieved.
- The foamability number of regrind material has been found to be identical to the raw material.
- 25

Example 3 (butane, without nucleating agent)

- An essentially linear impact polypropylene has an MFR of 0.25g/10min, the total ethylene content is 9wt%, the crystallization temperature is 115°C.
- 30 The ethylene content in the xylene soluble fraction is 33wt%. The intrinsic

viscosity of the xylene soluble fraction is 3.1dl/g. The tensile modulus of the rigid (un-foamed) material is 1150MPa. Used material, which has a melt strength of 0.015 N (1.5 cN) and a Foamability Number FN=0.5 N is extruded in contact with 5 wt% butane. The final density of the cellular material is 40 kg/m³. The tensile modulus of final parts, made from the cellular material is 60 MPa. The foamability number of regrind material has been found to be identical to the raw material.

Example 4 (carbon dioxide, without nucleating agent)

10

The raw material of example 3 is extruded in contact with 0.8 wt% carbon dioxide. The final density of the cellular material is 170 kg/m³. The tensile modulus of final parts, made from the cellular material is 240 MPa.

The foamability number of regrind material has been found to be identical to the raw material.

Example 5 (butane, with alpha nucleant)

A raw material as in example 3, but alpha nucleated using BNT, was used. Such prepared material had a crystallization temperature of 120°C. It was foamed with butane and a cellular material of a density of 40 kg/m³ was obtained. The tensile modulus of final parts, made from the cellular material, is 80MPa. The foamability number of regrind material has been found to be identical to the raw material.

25

Example 6 (carbon dioxide, with alpha nucleant)

The raw material of example 4 is foamed with carbon dioxide, subsequently quenched in a liquid. The cellular material shows a density of 170kg/m³. The tensile modulus of final parts, made from the cellular material is

290MPa. The foamability number of regrind material has been found to be identical to the raw material.

Example 7 (butane, with beta nucleant)

5

The raw material of example 3 is mixed with 2000 ppm Ca-Pimelate acting as beta nucleating agent and foamed with butane, subsequently quenched in a liquid. The cellular material shows a density of 40 kg/m³. The tensile modulus of final parts, made from the cellular material, is 36MPa. The
10 foamability number of regrind material has been found to be identical to the raw material.

Example 8 (carbon dioxide, with beta nucleant)

15 The raw material of example 3 is mixed with 2000 ppm Ca-Pimelate acting as beta nucleating agent and foamed with carbon dioxide, subsequently quenched in a liquid. The cellular material shows a density of 170kg/m³. The tensile modulus of final parts, made from the cellular material is 220MPa. The foamability number of regrind material has been found to be
20 identical to the raw material.

The results of the examples are summarized in the following table.

Table

	Melt Strength [cN]	FN [N]	Nucleation	Density	Tensile Modulus [MPa]
Example 1	2	0,1	No	450	
Example 2	2	0,1	No	410	
Example 3	1,5	0,5	No	40	60
Example 4	1,5	0,5	No	170	240
Example 5	1,5	0,5	Alpha	40	80
Example 6	1,5	0,5	Alpha	170	290
Example 7	1,5	0,5	Beta	40	36
Example 8	1,5	0,5	Beta	170	220

Claims

1. Extruded polyolefin having
 - a) MFR₂ of 0.01 to 1.00 g/10min and
 - 5 b) a foamability number (FN) of higher than 0.25 N whereby the foamability number is defined by the integral $\int_{\lambda=0}^{\lambda_{\max}} F \cdot d\lambda$, wherein λ is the stretch ratio and F is the force given in cN recorded as a function of the stretch ratio λ .
- 10 2. Extruded polyolefin according to claim 1 having a melt strength lower than 3 cN.
3. Extruded polyolefin according to claim 1 or 2 having a crystallization temperature T_c of 110 to 135°C.
- 15 4. Extruded polyolefin according to any one of the claims 1 to 3 comprising a α -crystallization nucleating agent and/or a β -crystallization nucleating agent.
- 20 5. Extruded polyolefin according to any one of the claims 1 to 4, whereby the polyolefin is a polypropylene.
6. Extruded polyolefin according claim 5, whereby the polypropylene is a linear propylene block-copolymer.
- 25 7. Extruded polyolefin according to claim 5 or 6, whereby the polypropylene has an ethylene content of less than 15 wt%.

8. Extruded polyolefin according to any one of the claims 5 to 7, whereby the polypropylene comprises a polypropylene matrix and an ethylene-propylene rubber (EPR).
- 5 9. Extruded polyolefin according to claim 8, whereby the ethylene propylene rubber (EPR) has an ethylene content of less than 50 wt%.
10. Beads comprising an extruded polyolefin according to any one of the claims 1 to 9.
- 10 11. Foam comprising an extruded polyolefin of one of the preceding claims 1 to 9.
12. Foam according to claim 11, wherein the foam has a density lower than
15 400 kg/m³.
13. Article comprising an extruded polyolefin according to any one of the claims 1 to 9 and/or beads according to claim 10 and/or a foam according to any one of claims 11 to 12.
- 20 14. Use of extruded polyolefin according to any one of the claims 1 to 9 or beads according to claim 10 to produce a foamed article.

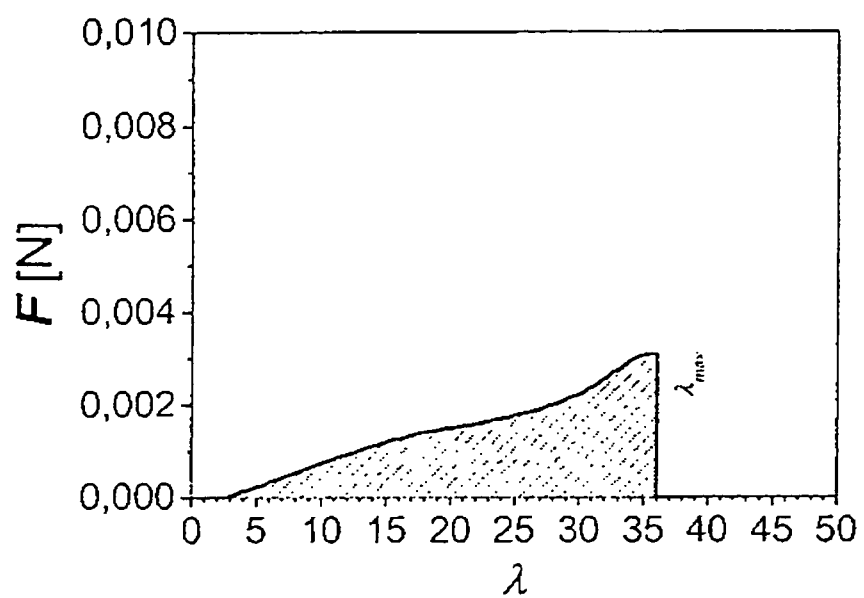
Figures

Figure 1: Determination of the Foamability Number (FN)

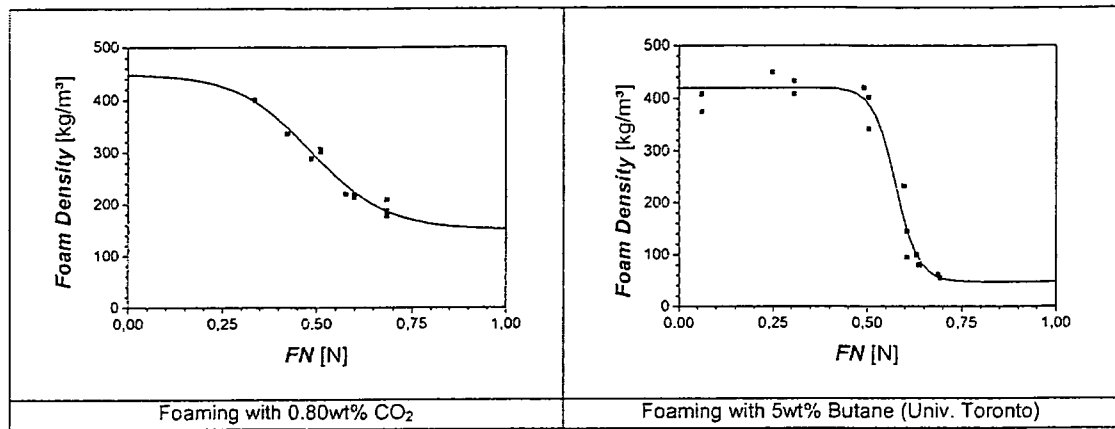


Figure 2: Correlation for minimum foam density and Foamability Number - with FN > 0.30 low density foam is achieved (< 400 kg/m³)

INTERNATIONAL SEARCH REPORT

International application No

PCT/EP2007/000599

A. CLASSIFICATION OF SUBJECT MATTER
 INV. C08L23/10 C08J9/00

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 C08J

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, PAJ, 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	EP 1 323 779 A (BOREALIS GMBH) 2 July 2003 (2003-07-02) the whole document examples	1-14
X	US 5 929 127 A (RAETZSCH ET AL) 27 July 1999 (1999-07-27) the whole document	1-14
X	PATENT ABSTRACTS OF JAPAN vol. 007, no. 235 (C-191), 19 October 1983 (1983-10-19) & JP 58 129025 A (HITACHI KASEI KOGYO KK), 1 August 1983 (1983-08-01) abstract	1-14
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☒ 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.

* & * document member of the same patent family

Date of the actual completion of the international search

26 April 2007

Date of mailing of the international search report

07/05/2007

Name and mailing address of the ISA/

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Droghetti, Anna

INTERNATIONAL SEARCH REPORT

International application No

PCT/EP2007/000599

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	PATENT ABSTRACTS OF JAPAN vol. 015, no. 193 (C-0832), 17 May 1991 (1991-05-17) & JP 03 050245 A (FURUKAWA ELECTRIC CO LTD:THE), 4 March 1991 (1991-03-04) abstract -----	1-14

INTERNATIONAL SEARCH REPORT

International application No.
PCT/EP2007/000599

Box II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☒ Claims Nos.:
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:
see FURTHER INFORMATION sheet PCT/ISA/210
3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; It is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.
- ☐ No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

Continuation of Box II.2

Claims Nos.: -

1. Clarity and disclosure of the invention, Art. 6 and 5 PCT

1.1. Present claim 1 relates to a polyolefin defined (inter alia) by reference to the following unusual parameter: foamability number (FN).

The use of this unusual parameter in the present context is considered to lead to a lack of clarity because the claim does not clearly identify the products encompassed by it as the parameter cannot be clearly and reliably determined either by indications in the description or by objective procedures which are usual in the art (e.g. how the stretch ratio can be determined and defined). This makes it impossible to compare the claim to the prior art. As a result, the application does not comply with the requirement of clarity under Article 6 PCT.

The present claim 1 relates to an extremely large number of possible compounds. Support and disclosure in the sense of Article 6 and 5 PCT is to be found however for only a very small proportion of the polyolefins claimed, see the examples and pages 5-7.

Furthermore the present claim 1 encompasses polyolefins defined only by their desired function (the foamability properties), contrary to the requirements of clarity of Article 6 PCT, because the result-to-be-achieved type of definition does not allow the scope of the claim to be ascertained. The fact that any compound could be screened does not overcome this objection (see pages 6, 1.10-14; page 8, lines 11-13), as the skilled person would not have knowledge beforehand as to whether it would fall within the scope claimed, except for the compounds disclosed in the description, see the examples.

Undue experimentation would be required to screen compounds randomly.

As a consequence, this non-compliance with the substantive provisions is to such an extent, that a meaningful search of the whole claimed subject-matter of the claims 1-14 could not be carried out (Article 17(2) PCT).

The lack of clarity, support and disclosure is to such an extent, that the search was restricted to those claimed polyolefins and products therefrom which appear to be supported and a generalisation of their structural formulae, that is polypropylene (e.g. linear block copolymers) and polypropylene blends comprising a propylene matrix and EPR and having defined Tc, melt strength and MI2, ethylene comonomer content and further nucleating agents and whose foams have specific density as disclosed in claims 1 in part, 2-14 and pages 1-15.

The applicant's attention is drawn to the fact that claims relating to inventions in respect of which no international search report has been established need not be the subject of an international preliminary

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

examination (Rule 66.1(e) PCT). The applicant is advised that the EPO policy when acting as an International Preliminary Examining Authority is normally not to carry out a preliminary examination on matter which has not been searched. This is the case irrespective of whether or not the claims are amended following receipt of the search report or during any Chapter II procedure. If the application proceeds into the regional phase before the EPO, the applicant is reminded that a search may be carried out during examination before the EPO (see EPO Guideline C-VI, 8.5), should the problems which led to the Article 17(2) declaration be overcome.

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2007/000599

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