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(54) **Title:** COMPOSITION BASED ON PROPYLENE POLYMER, PROCESS FOR ITS OBTENTION, USE FOR THE MANUFACTURE OF EXPANDED GRANULES AND CELLULAR ARTICLES

(57) **Abstract:** A composition for expanded granules based on semi-crystalline propylene polymer comprising, per 100 parts by weight, less than 0.025 part by weight of volatile compounds and processes for its preparation.

COMPOSITION BASED ON PROPYLENE POLYMER, PROCESS FOR ITS OBTENTION, USE FOR THE MANUFACTURE OF EXPANDED GRANULES AND CELLULAR ARTICLES

The present invention relates to a composition based on propylene polymer, which can be converted into expanded granules and particularly to a composition having an improved resistance to fogging. It also relates to a process for obtaining such composition as well as to its use for the manufacture of expanded granules. Finally it relates to cellular articles fashioned from said expanded granules.

It is known to use propylene polymers for manufacturing cellular articles from expanded particles. These cellular articles are widely used as interior materials for automobiles and other vehicles since they are light in weight, heat resistant and easy to mold. However, in hot weather, some components of the plastic materials evaporate, attach to the window glasses of the vehicles causing some deterioration of visibility and eventually give offensive odor. Such phenomenon, known as fogging, is a cause of concern for the automotive manufacturers. The following means are proposed for its limitation.

JP-A-06-234898 suggests the use of blends of grafted polyolefin with polyamide and specific inorganic fibers such as glass fiber. However these blends, being different from the conventional propylene polymers used in the other parts of the vehicle, are not easily recycled. EP-A-0905175 or JP-A-11-263863 proposes the addition of fogging inhibitors such as aluminium sulfate or specific carbon black and JP-A-11-028789 and WO 00/06645 the addition of specific additives to the layers laminated on the foamed articles. However the fogging resistance of the cellular articles still needs to be improved.

From above it appears that the fogging problem is not satisfactorily solved and remains worrying particularly for the cellular articles made from expanded beads which are not concerned by the above mentioned disclosures.

Accordingly, the aim of the present invention is to provide a polypropylene based composition having an improved fogging resistance and which can be used for the preparation of expanded granules.

The present invention therefore relates to a composition for expanded granules based on semi-crystalline propylene polymer comprising, per 100 parts by weight, less than 0.025 part by weight of volatile compounds.

According to the present description, the content of volatile compounds contained in a composition is determined as follows :

50 mg of the composition under to form of particle of mean dimensions of from 1 to 2 mm are disposed in a vessel as a strip of 80 to 70 mm under at 120°C during one hour. A helium stream passes on the strip with a flow rate of 15 ml/min before being cooled at -196°C to trap the desorbed compounds.

The content of volatile compounds is the quantity of compounds trapped having an elution time of at most 40 min determined by gas phase chromatography by means of a semi capillary column equipped with a FID detector (column CP-Sil 24 CB, diameter : 0.53 mm, length : 15 m, helium flow : 10 ml/min, temperature : 120 °C during the 2 first minutes, then increase at 8 °C/min till 265 °C).

It is supposed that the volatile compounds contribute to the fogging because of their relatively high volatility. Accordingly their content should be maintained as low as possible. For economic reasons, however, it is generally higher than 0.0005, particularly higher than 0.001 part by weight per 100 parts by weight of composition. Contents of at most 0.02 part by weight gives advantageous results. Contents of at most 0.01 part by weight are particularly preferred.

It is further supposed that the volatile compounds are mainly formed by volatile hydrocarbon compounds contained in the propylene polymer and, eventually, by the polymer additive(s) and/or by the degradation products thereof.

The volatile hydrocarbon compounds are generally oligomers of propylene formed during the polymerization reaction. They usually contain from 9 to 39 carbon atoms.

Additives who contribute to the volatile compounds by themselves or through their degradation product(s) are for instance sterically hindered phenol having a molecular weight lower than 500 daltons (phenol (a)) or organic phosphite of formula $P(OR)_3$ wherein each R represents, independently from each other, alkyl radicals or non sterically hindered aryl radicals (phosphite (a)).

Examples of phenol (a) are 2,4-dimethyl,6-tert-butylphenol, 2,6-ditertbutyl para cresol, and octyl beta- (3,5-ditertbutyl-4-hydroxyphenyl) -

propionate. Examples of phosphites (a) are tris(para-nonylphenyl)phosphite and distearylpentaerythrityldiphosphite.

Accordingly, compositions according to the invention containing, per 100 parts by weight, less than 0.05 part by weight of the sterically hindered phenol (a) and less than 0.05 part by weight of the organic phosphite (a) are particularly preferred.

Compositions according to the invention free of phenol (a) and of phosphite (a) give good anti-fogging properties.

The propylene polymers used in the composition according to the invention are semi-crystalline polymers in the sense of the meaning given in "Plastic Engineering" 2nd edition, R.J. Crawford, Pergamon Press (1987) pages 3 and 4. The propylene polymers used in the present invention are generally chosen from the propylene homopolymers and the copolymers of this latter containing at most 50 weight % of at least one comonomer chosen from ethylene and α-olefins containing from 4 to 8 carbon atoms and their mixtures. The copolymers of propylene are preferably chosen from copolymers of propylene with up to 20 mol % ethylene and/or 1-butene.

Copolymers of propylene containing from 1 to 6 mol % of ethylene are particularly preferred because they provide a good compromise between expansion and molding behavior and rigidity.

Mixtures of two different propylene polymers chosen, on the one hand, from homopolymers of propylene and copolymers of this latter containing less than 2 mol % of ethylene and, on the other hand, from copolymers of propylene containing from 1 to 20 mol % of ethylene and/or 1-butene may also be advantageously used.

The comonomer contents mentioned in the present description are determined by Fourier transform IR spectroscopy on the polymer converted into a 200 µm pressed film. It is the absorption bands at 732 and 720 cm^{-1} that are used to determine the ethylene content and the absorption band at 767 to determine the 1-butene content.

The propylene polymer used in the composition according to the invention has generally a melt flow index (MFI), measured according to ASTM D 1238 (1986) under a load of 2.16 kg at 230 °C in the range from 0.3 to 30 g/10 min. This value is more preferably at least 1 g/10 min. Values of at most 20 g/10 min give satisfactory results. When the propylene polymer is a mixture of different propylene polymers as disclosed hereabove, the polymers may have different MFI.

The compositions according to the invention contain usually additives to improve their stability. Regarding the additives generally used for propylene polymers, it has been found that, some of them are better in terms of fogging resistance. For instance, compositions containing, per 100 parts by weight, from 0.03 to 0.5 part by weight of sterically hindered phenol having a molecular weight of at least 500 daltons (phenol (b)), and up to 0.3 part by weight of organic phosphite of formula $P(OR')_3$ wherein each R' represents independently from each other alkyl or aryl radicals providing that at least one R' represents a sterically hindered alkyl radical (phosphite (b)) are particularly advantageous in terms of fogging resistance.

The content of phenol (b) is usually of at least 0.05 part per 100 parts of the composition. Contents of at least 0.1 part give the best results. Compositions containing at most 0.4 part of phenol (b) are very suitable.

The phenols (b) having a molecular weight of at least 700 daltons are preferred because of they lead to polymers having better compromise stability / fogging resistance.

Examples of such phenols (b) are the pentaerythrityl tetrakis(3,5-ditertbutyl-4-hydroxyphenyl) propionate and the bis(beta-3,5-ditertbutyl-4-hydroxyphenyl-ethyl)suberate.

Compositions containing, per 100 parts by weight, at least 0.02 part per 100 parts of phosphite (b) are preferred. Compositions containing at least 0.04 part of phosphite (b) have a good compromise of properties. The content of phosphite (b) is furthermore usually of at most 0.2, preferably of at most 0.1 part by weight.

With regard to the phosphite (b), the tris(2,4-ditertbutylphenyl)phosphite is particularly suitable.

It is moreover preferable that the amounts of phenol (b) and of phosphite (b) are such that their weight ratio is in the range of 2 to 10.

The compositions according to the invention may comprise, as occasion demands, known other conventional additives such as stabilizers, pigments, colorants, fillers, fire retardants, antistatic agents, antiacid agents, lubricants, slip-promoting agents, etc. Preferably these additives are added in amounts such that the above mentioned conditions are satisfied. Antiacid agents and lubricants are specially preferred. Their contents are usually of from 0.05 to 0.2 part by weight per 100 parts of the composition. Other preferred additives are the thiosynergists such as the 3,3'-thio-bis propanoic acid or the dilauryl thiodipropionate. These additives are usually used in quantities of 0.001 to 0.5 part per 100 parts of the composition.

Preferably the composition according to the invention contains by way of polymers only the propylene polymer(s) defined hereabove. Composition in which the propylene polymer(s) is (are) neither grafted nor cross-linked is usually preferred due to a better compromise of properties.

The composition according to the invention may be obtained by any known process.

According to a first example, a propylene polymer is obtained by using polymerization conditions such that it incorporates few volatile hydrocarbon compounds is used. Example of such conditions is the polymerization of the propylene and of the optional comonomer(s) in a hydrocarbon diluent chosen from liquid alcanes and cycloalcanes wherein the molar fraction of propylene is at most 0.9. The diluent is usually hexane or heptane. The temperature and pressure are those generally used by the man skilled in the art. Temperature between 50 and 90°C and pressures between 10 and 30 atmospheres are particularly suitable. The catalytic system may be any catalytic system known to be sufficiently productive and stereospecific permitting the propylene to be polymerized in isotactic form and capable of incorporating the desired comonomer(s). The propylene polymer issued from said polymerization reaction is then separated from the polymerization medium by any known techniques such as filtration or centrifugation and usually dried to eliminate the polymerization diluent. It is particularly advantageous that the polymer is dried until the content of polymerization diluent is less than 0.1, specifically less than 0.01 weight %. Preferably the drying is carried out at a temperature of from 70 to 110°C. The duration of the drying depends on the temperature. Its is usually from 0.1 to 2 h, preferably from 0.25 to 1 h. The dried propylene polymer is then usually melt blended with the additives mentioned hereabove. The melt blending is carried out by any known techniques. It may be for example first carried out at ambient temperature followed by a second mixing at a temperature above the melting point of the propylene polymer, for example in a mechanical mixer or in an extruder. The temperature of the second stage is generally from 100 to 300°C, in particular from 120 to 210°C. An alternative method consists in introducing the additives into the polymer that is already molten. It is preferred to proceed according to the first technique.

Other ways for the preparation of the composition according to the invention involve the removal of the volatile compounds. The operating conditions (temperature, duration and pressure) of such removal are such that the melting and/or the degradation of the propylene polymer are avoided.

Two embodiments give particularly good results.

According to the first one, a propylene polymer issued from the polymerization reaction and containing preferably, per 100 parts by weight, more than 0.025 part by weight of volatile compounds is brought into contact with an aliphatic or cycloaliphatic hydrocarbon diluent containing from 4 to 7 carbon atoms at a temperature in the range of 5 to 60°C for a period of 0.1 to 4 h in an amount of from 0.1 to 5 g of polymer per g of hydrocarbon diluent, and separated from said diluent. The contact of the propylene polymer and the hydrocarbon diluent is generally carried out at a pressure of 10^5 to 3 10^5 Pa. The temperature is preferably in the range of 10 to 60°C, temperatures of 15 to 40°C giving the best results. The relative amounts of the propylene polymer to the hydrocarbon diluent are often of from 0.2 to 1, ratio of from 0.25 to 0.75 giving the best results at reasonable costs. The duration of said treatment is advantageously from 0.2 to 2 h; duration of from 0.3 to 1 h giving the best compromise. The propylene polymer issued from said treatment is then usually separated from the hydrocarbon diluent by any known process such as filtration or centrifugation. The filtration is preferably chosen for economic reasons. The propylene polymer thus obtained is then preferably dried by any known process. Preferably the drying is carried out at a temperature of from 70 to 120 °C. The duration of the drying depends on the temperature. Its is usually from 0.1 to 2 h, preferably from 0.25 to 1.5 h. The treated propylene polymer is then advantageously mixed with the above mentioned additives. The mixing conditions (concentrations, temperature ...) are usually those described hereabove with respect to the propylene polymer issued from the polymerization.

According to the second embodiment, a propylene polymer issued from the polymerization reaction and containing preferably, per 100 parts by weight, more than 0.025 part by weight of volatile compounds is first melt mixed with the additive(s) described hereabove to obtain granules which are contacted with a gas stream which may be a stream of hot air, a gas stream containing steam or a stream of steam at a temperature of from 60 to 160 °C for a duration of from 30 minutes to 100 hours. Treatment of the propylene polymer with gas stream containing steam gives good results. In this specific case, the gas stream is generally composed of a gas such as nitrogen or air being preferred. The temperature of this process is generally at least 70°C and more precisely at least 80 °C, values of at least 90 °C being the most common. The temperature is furthermore preferably of at most 120°C. Good results are obtained when the temperature is of approximately 100°C. The duration of said treatment usually do not exceed 80 hours and more particularly 60 hours. When the treatment is carried out with a gas stream containing steam, the total amount of steam is generally of at least 0.01 kg, preferably at least 0.05 kg and more particularly at least 0.1 kg per kg of propylene polymer. In general, this amount is less than or equal to 20 kg, most often less than or equal to 10 kg and more particularly less than or equal to 5 kg of steam per kg of polymer.

According to the present invention a propylene polymer issued from the polymerization reaction and containing, per 100 parts by weight, more than 0.025 part by weight of volatile compounds treated with the hydrocarbon diluent, mixed with the additive and treated with the gas stream gives composition having particularly good fogging resistance.

Moreover compositions wherein the propylene polymer is obtained by using polymerization conditions wherein the content of volatile hydrocarbon compounds is low and mixed with the additives and treated with the gas stream are advantageously used.

Generally speaking the treatment of the polymer may be carried out continuously or batchwise. The device(s) used for the polymer treatments may be any receptacle into which the polymer issued from the polymerization, the hydrocarbon diluent or the gas stream are introduced.

The melt blending of the propylene polymer and the additives is carried out in any device known for that purpose giving sufficiently high shear rate to induce the melting of the propylene polymer. It is thus possible to work with external and internal mixers. Internal mixers are the most appropriate and amongst them extruders are the most preferred.

The additives used in the above described process are usually of, per 100 part by weight of the propylene polymer, at least from 0.03 to 0.5 part by weight of the phenol (b), and up to 0.3 part by weight of the phosphite (b).

The preferred quantities of these additives and the quantity of the other additives that may be used are in accordance with that described hereabove with respect of the composition according to the invention.

The compositions of the invention are particularly suitable for the manufacture of expanded granules and for the subsequent manufacture, starting from these latters, of cellular articles. Due to their improved resistance to fogging, the composition of the present invention may be successfully used in the automotive industry and particularly as interior trim such as dashboard, sunvisor, headrest, interior material of the ceiling, or of the doors. It should be furthermore noted that the improvement of the fogging resistance is obtained without loss and even without modifications in the mechanical and chemical properties of the propylene polymer.

The manufacture of the expanded granules and the subsequent moulding of the expanded granules into cellular articles are generally carried out in the conventional manner known by the man of ordinary skill in the art. Examples of the manufacture are described, for example, in documents EP-A-0317995 and US-A-4626555.

The following examples illustrate the invention.

Example 1 (given by way of comparison)

A random propylene copolymer containing 2.3 weight % of ethylene and having a MFI of 9.8 g/10 min and a melting temperature of 145.2 is melt blended with 2 g/kg of pentaerythrityltetrakis(3,5-ditertbutylparacresol)propionate (IRGANOX® 1010), 0.3 g/kg of 2,6-ditertbut-4-methylphenol, 1 g/kg of bis(2,4-ditertbutylphenyl)pentaerythritol diphosphite (ULTRANOX® 626), 0.6 g/kg or erucic acid amide (CRODAMINE ER) and 1 g/kg of calcium stearate has been pelletized on a ZSK 40 extruder at a temperature of 210 °C to obtain a composition containing, per 100 parts by weight, 0.075 part by weight of volatile compounds. Processed in expanded granules and cellular articles to be used for interior material for automobile said composition generates fogging.

Example 2 (given by way of comparison)

Example 1 is reproduced but by melting the random propylene copolymer with 2 g/kg of the IRGANOX 1010 product, 0.5 g/kg of tris(2,4-ditertbutylphenyl) posphite) (IRGAFOS® 168), 2 g/kg of distearylthiodipropionate, 0.6 g/kg of erucic acid amide and 1 g/kg of hydrotalcite to obtain a composition containing, per 100 parts, 0.055 part by weight of volatile compounds.

Interior materials for automobile made from expanded granules of said composition generate fogging.

Example 3 (according to the invention)

Example 2 is reproduced after having contacted the composition with a stream of steam at 100 °C during 15 h (0.5 kg of steam per kg of composition). The volatile compounds content of the obtained composition is, per 100 parts, of 0.019 part by weight.

Used as interior material for automobile, said composition generates dramatically less fogging than that of Examples 1 and 2.

Example 4 (according to the invention)

Example 2 is reproduced after having contacted the random copolymer with hexane at 30 °C during 1 h (0.5 kg of composition per kg of hexane). The volatile compounds content of the obtained composition is, per 100 parts, of 0.009 part by weight.

Used as interior material for automobile, said composition generates dramatically less fogging than that of Examples 1 and 2.

CLAIMS

1 - A composition for expanded granules based on semi-crystalline propylene polymer comprising, per 100 parts by weight, less than 0.025 part by weight of volatile compounds.

2 - The composition according to Claim 1, containing, per 100 parts by weight, less than 0.05 part by weight of sterically hindered phenol having a molecular weight lower than 500 daltons (phenol (a)) and less than 0.05 part by weight of organic phosphite of formula $P(OR)_3$ wherein each R represents, independently from each other, alkyl radicals or non sterically hindered aryl radicals (phosphite (a)).

3 - The composition according to Claim 1 or 2, wherein the propylene polymer is chosen from propylene homopolymers and copolymers of this latter containing at most 50 weight % of at least one comonomer chosen from ethylene and α-olefins containing from 4 to 8 carbon atoms and their mixtures.

4 - The composition according to Claim 3, wherein the propylene polymer is a propylene copolymer containing from 1 to 6 mol % of ethylene.

5 - The composition according to Claim 4, containing, per 100 parts by weight, from 0.03 to 0.5 part by weight of sterically hindered phenol having a molecular weight of at least 500 daltons (phenol (b)), and up to 0.3 part by weight of organic phosphite of formula $P(OR')_3$ wherein each R' represents independently from each other alkyl or aryl radicals providing that at least one R' represents a sterically hindered alkyl radical (phosphite (b)).

6 - The composition according to Claim 5, wherein the weight ratio of the phenol (b) to the organic phosphite (b) is from 2 to 10.

7 - Process for obtaining the composition for axpanded granules according to Claim 1, comprising the polymerization of propylene and of the optional comonomer(s) in a hydrocarbon diluent chosen from liquid alcanes and cycloalcanes wherein the molar fraction of propylene is at most 0.9.

8 - Process according to Claim 7, wherein the polymer issued from the polymerization reaction separated from the polymerization medium and dried is

finally melt mixed with, per 100 parts by weight of the final composition, from 0.03 to 0.5 part by weight of phenol (b), and up to 0.3 part by weight of phosphite (b).

9 - Process for obtaining the composition for expanded granules according to Claim 1, wherein a propylene polymer issued from the polymerization reaction and containing, per 100 parts by weight of the propylene polymer, more than 0.025 part by weight of volatile compounds is brought into contact with an aliphatic or cycloaliphatic hydrocarbon diluent containing from 4 to 7 carbon atoms at a temperature in the range of 5 to 60 °C for a period of from 0.1 to 4 h in an amount of from 0.1 to 5 g of propylene polymer per g of hydrocarbon diluent, and separated from said diluent.

10 - Process according to Claim 9, wherein the propylene polymer is finally melt mixed with, per 100 parts of final composition, at least from 0.03 to 0.5 part by weight of phenol (b), and up to 0.3 part by weight of phosphite (b).

11 - Process for obtaining a composition for expanded granules according to Claim 1, wherein a propylene polymer issued from the polymerization reaction and containing, per 100 parts by weight, more than 0.025 part by weight of volatile compounds is first melt mixed with, per 100 parts of the final composition, at least from 0.03 to 0.5 part by weight of phenol (b), and up to 0.3 part by weight of organic phosphite (b) to obtain granules which are brought into contact with a gas stream at a temperature in the range of 60 to 160 °C for a duration from 30 minutes to 100 hours.

12 - Use of a composition according to Claim 1 for the manufacture of expanded granules.

13 - Cellular articles fashioned by moulding expanded granules obtained from a composition according to Claim 1.

INTERNATIONAL SEARCH REPORT

Intern al Application No
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A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C08F10/06 C08K5/00

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)
IPC 7 C08F C08K

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, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 875 530 A (MONTELL NORTH AMERICA INC) 4 November 1998 (1998-11-04) the whole document ---	1-13
X	US 6 214 760 B1 (KRUPER JR WILLIAM J ET AL) 10 April 2001 (2001-04-10) column 21, line 55 -column 22, line 14 -----	7

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

☒ Patent family members are listed in 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

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"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

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"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

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INTERNATIONAL SEARCH REPORT

Information on patent family members

Intern[illegible]l Application No
PCT/EP 01/07017

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP 0875530 A	04-11-1998	US 5834541 A	10-11-1998
		AU 733943 B2	31-05-2001
		AU 6378298 A	05-11-1998
		BR 9801528 A	29-06-1999
		CA 2236634 A1	02-11-1998
		CN 1198448 A	11-11-1998
		CZ 9801330 A3	11-11-1998
		EP 0875530 A1	04-11-1998
		HU 9800914 A2	30-11-1998
		JP 10306178 A	17-11-1998
		NO 981917 A	03-11-1998
		PL 326099 A1	09-11-1998
		SK 48498 A3	04-11-1998
		TW 401441 B	11-08-2000
US 6214760 B1	10-04-2001	AU 4440499 A	06-03-2000
		AU 4564799 A	06-03-2000
		AU 4564899 A	06-03-2000
		EP 1104429 A1	06-06-2001
		EP 1104426 A1	06-06-2001
		EP 1104430 A1	06-06-2001
		WO 0009523 A1	24-02-2000
		WO 0009515 A1	24-02-2000
		WO 0009524 A1	24-02-2000
		US 6140521 A	31-10-2000
		US 6162935 A	19-12-2000

PCT REQUEST

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VIII-18	**Figure of the drawings which should accompany the abstract**	
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IX-1-1	Name (LAST, First)	DESTRYKER, Elise

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10-2-2	Not received	
10-3	**Corrected date of actual receipt due to later but timely received papers or drawings completing the purported international application**	
10-4	**Date of timely receipt of the required corrections under PCT Article 11(2)**	
10-5	**International Searching Authority**	ISA/EP
10-6	**Transmittal of search copy delayed until search fee is paid**	

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11-1	**Date of receipt of the record copy by the International Bureau**	24 JULY 2001 24. 07. 01