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(54) **Title:** AN EPOXY RESIN COMPONENT FOR AN ADHESIVE COMPOSITION

(57) **Abstract:** An epoxy resin component for an adhesive composition is disclosed. Said epoxy resin component comprises of an epoxy resin premix; and a filler composition comprising cenospheres and wollastonite in a w/w (weight / weight) ratio in a range of 1:0.01 - 1:5.

AN EPOXY RESIN COMPONENT FOR AN ADHESIVE COMPOSITION

Field of Invention

The present disclosure relates to epoxy resin components for adhesive compositions. Specifically, the present disclosure relates to low density epoxy resin components having desired rheological properties and suitable for use in structural adhesives.

Background

Structural adhesives find application in various industries such as automobile, wind mill, aerospace to produce a load-bearing joint. Structural adhesives or in general any adhesive composition comprises of any of the following elements: adhesive base, diluents, hardener/curatives, fillers, solvents and carriers. Adhesive base is the key ingredient of the adhesive composition which forms bonds between itself in the presence of a suitable hardener or curative. Fillers are added to modify or enhance the properties of the adhesive base such as: to improve the rheological properties such as viscosity, sag resistance and reduction in shrinkage upon curing. Fillers may also be added to increase the cured strength, impart or enhance electrical or thermal conductivity, increase the bulk etc. However, addition of filler leads to an increase in density and hence weight of the adhesive composition. The increase in density, particularly in case of structural adhesives, increases the structural loading of the structure to which the adhesive has been applied and thereby affects the performance of the structure. In view of the aforesaid, it is desirable to reduce the weight of the adhesive composition for their application as structural adhesives. It is also desirable that such adhesive compositions should have similar or improved rheological properties as compared to the conventional adhesive compositions.

Summary

An epoxy resin component for an adhesive composition is disclosed. Said epoxy resin component comprises of an epoxy resin premix; and a filler composition comprising cenospheres and wollastonite in a w/w (weight / weight) ratio in a range of 1:0.01 – 1:5.

- 5 An adhesive composition comprising said epoxy resin component and a hardener component is also disclosed.

Detailed Description

For the purpose of promoting an understanding of the principles of the invention,
10 reference will now be made to embodiments and specific language will be used to describe the same. It will nevertheless be understood that no limitation of the scope of the invention is thereby intended, such alterations and further modifications in the disclosed composition and method, and such further applications of the principles of the invention therein being contemplated as would normally occur to one skilled in the art to which the inven-
15 tion relates.

It will be understood by those skilled in the art that the foregoing general description and the following detailed description are exemplary and explanatory of the invention and are not intended to be restrictive thereof.

Reference throughout this specification to “one embodiment” “an embodiment” or
20 similar language means that a particular feature, structure, or characteristic described in connection with the embodiment is included in at least one embodiment of the present invention. Thus, appearances of the phrase “in one embodiment”, “in an embodiment” and similar language throughout this specification may, but do not necessarily, all refer to the same embodiment.

In its broadest scope, the present disclosure relates to an epoxy resin component for an adhesive composition. Specifically, the present disclosure relates to a low density epoxy resin component suitable for use in structural adhesives. Said epoxy resin component comprises of an epoxy resin premix; and a filler composition comprising cenospheres and wollastonite in a w/w ratio in a range of 1:0.01 – 1:5.

An adhesive composition comprising said epoxy resin component is also disclosed. The adhesive composition comprises about 65-75 % w/w of an epoxy resin component, and about 25-35% w/w of a hardener component. Herein, the epoxy resin component comprises of an epoxy resin premix and a filler composition comprising cenospheres and wollastonite in a w/w ratio in a range of 1:0.01- 1:5.

The epoxy resin component disclosed herein has a lower density but similar or improved rheological properties as compared to conventional epoxy resin components. Conventional epoxy resin component in context of the present disclosure refers to an epoxy resin component having the same ingredients as that used in the epoxy resin component of the present disclosure, except the filler composition. The conventional epoxy resin component includes conventional fillers such as milled glass fibres. Similarly, conventional adhesive composition refers to an adhesive composition comprising a conventional epoxy resin component.

The combination of cenospheres and wollastonite in the given ratio provides a synergistic effect of maintaining the rheological properties such as viscosity, thixotropic index in the desired range while reducing the density of the epoxy resin component and hence the cured adhesive composition. The present inventors observed that when cenospheres and wollastonite are used individually, the rheological properties such as viscosity and thixotropic index of the epoxy resin component increase beyond the desired range making the epoxy resin component unsuitable for use as a structural adhesive.

By applying the teachings of the present disclosure, an epoxy resin component having a viscosity in a range of 300-700 Pa.S at 25°C, a thixotropic index in a range of 3-5 can be obtained, while achieving a reduction in weight / density by 5-15 % . The density of the resultant adhesive composition is 8-10% lower than that of the conventional adhesive composition. Low density adhesive compositions aid in reducing the overall weight of the structure to which it is applied such as a windmill blade, thereby increasing its efficiency. Additionally, it may also lead to an improvement in other properties of the adhesive composition, such as sagging, peel strength and shrinkage.

Cenospheres used in the filler composition are lightweight, inert, hollow spheres made of one or more of SiO₂, Al₂O₃, CaO, K₂O, Fe₂O₃, TiO₂, MgO, Na₂O and filled with an inert air or gas such as carbon dioxide and nitrogen. Said cenospheres have a particle size in a range of 75- 300µ. Preferably, cenospheres have the particle size in the range of 100-150µ. In accordance with a further embodiment, said cenospheres have a bulk density in a range of 300-600 Kg/m³. Preferably, said cenospheres have the bulk density in the range of 400-500 Kg/m³. Said cenospheres may be manufactured by any known method or may be obtained from commercial sources. For example, cenospheres sold by Cenosphere India Pvt. Ltd. under the trade name CIL™ 500 may be used.

Wollastonite used in the filler composition is a naturally occurring calcium silicate having formula Ca₃Si₃O₉ and a needle-like shape. Said wollastonite has a particle size in a range of 75-300 µ. Preferably, the particle size is in the range of 100-200µ. In accordance with a related embodiment, said wollastonite has an aspect ratio in a range of 5:1 to 30:1. Preferably, wollastonite has an aspect ratio in the range of 10:1-20:1. In accordance with yet another embodiment, said wollastonite has a bulk density in a range of 200-500 Kg/m³. Preferably, said wollastonite has the bulk density in a range of 300-400 Kg/m³. Wollastonite may be manufactured by any known method or obtained from commercial sources. For

example, wollastonite sold by Wolkem India Ltd. under the trade name “KEMOLIT[®]” may be used. In accordance with a further embodiment, wollastonite is in surface modified form. The surface modification may be achieved with known silanes by any known method. Commercially available surface modified wollastonite may also be used. For example, 5 surface modified wollastonite sold by Wolkem India Ltd. under the trade name “FILL-LEX[®]” may be used.

In accordance with an aspect, the filler composition comprises cenospheres and wollastonite in a w/w ratio in a range of 1:0.01–1:5. In accordance with a preferred embodiment, the filler composition comprises cenospheres and wollastonite in a w/w ratio in 10 a range of 1:1 to 1:3.

In accordance with an embodiment, the filler composition is present in the epoxy resin component in an amount in a range of 10-30 % w/w of said epoxy resin component. In accordance with a preferred embodiment, the filler composition is present in the epoxy resin component in an amount in a range of 15-25 % w/w of said epoxy resin component.

15 In accordance with an embodiment, the epoxy resin premix comprises of at least one epoxy resin, at least one diluent and fumed silica.

In accordance with an embodiment, the epoxy resin may be a polyglycidyl ether of a polyhydric alcohol such as 1,4-butanediol or 1,3-propanediol or, preferably, a polyglycidyl ether of a polyhydric phenol, for example a bisphenol such as bis(4- 20 hydroxyphenyl)methane (bisphenol F) or 2,2-bis-(4-hydroxyphenyl)propane (bisphenol A) or a novolak formed from formaldehyde and a phenol such as phenol itself or a cresol, or a mixture of two or more such polyglycidyl ethers. In accordance with a preferred embodiment, said epoxy resin is selected from a group consisting of bisphenol A diglycidyl ether and bisphenol F diglycidyl ether. A combination of two or more epoxy resins may also be 25 used. Such epoxy resins may be prepared using known methods or may be obtained from

commercial sources. In accordance with a further embodiment, the epoxy resin forms at least 60-80%, and preferably 65-75% of the epoxy resin component.

A wide range of diluents can be used to prepare the epoxy resin premix. In accordance with an embodiment, the diluent is a reactive diluent which participates in a chemical
5 reaction with at least one other ingredient of the adhesive composition upon curing. The reactive diluent may be selected from a group consisting of epoxy families like di ols, mono ols, novolacs and epoxy urethane hybrids. In accordance with an alternate embodiment, the diluent is a non-reactive diluent. The non-reactive diluent may be selected from
10 a group consisting of Benzyl alcohol, Furfurylalcohol, Nonyl phenol, Dibutyl phthalate and hydrocarbon resins such as Necirez-products, Epodil-L, Pine-oil. The diluents which could modify certain properties of the epoxy resin component or the adhesive composition may also be used. Also, a combination of two or more diluents may be used. The diluent is added in an amount in a range of 0-20% w/ w of said epoxy resin component. The amount of diluent may vary in accordance with the application.

15 In accordance with an embodiment, aforesaid fumed silica may be hydrophilic or hydrophobic. Fumed silica is added in an amount in a range of 0.01-10% w/ w of said epoxy resin component. Fumed silica assists in improving thixotropy and anti-setting properties. Fumed silica used in the present disclosure may be selected from fumed silica known in the art; or may be obtained from commercial sources. In accordance with an
20 embodiment, a color may be additionally included in the epoxy resin premix.

Said epoxy resin premix may be prepared by any known method. Preferably, the epoxy resin premix is prepared by mixing the epoxy resin and the reactive diluents in a reactor, followed by the addition of fumed silica. Alternatively, the epoxy resin premix may be obtained from commercial sources. For example, an intermediate of Epotec®
25 YD1535 G, a commercially available epoxy resin component from Aditya Birla Chemi-

icals, Thailand may be used. The intermediate comprises of bisphenol A based diglycidyl ether, bisphenol F based diglycidyl ether and reactive diluents and is referred to as Epotec® YD1535G premix.

In accordance with an embodiment, the hardener component comprises of at least
5 one hardener and fumed silica. Any known hardener may be used. Any two or more hardeners may also be used depending on the application. In accordance with an embodiment, the hardener may be selected from a group consisting of polyetheramine, cycloaliphatic amines having an amino or aminoalkyl group attached to the ring, such as 3-aminomethyl-3,5,5-trimethylcyclohexylamine (isophoronediamine), an amino polyamide, tetraethylene-
10 pentamine having N-(2-aminoethyl)-N'-{2-[(2-aminoethyl)amino]ethyl}-1,2-ethanediamine, 2, 4-(2-aminoethyl)-N-(2-aminoethyl)-N'-{2-[(2-aminoethyl)amino]ethyl}-1,2-ethanediamine, 2,1-(2-aminoethyl)-4-[(2-aminoethyl)amino]ethyl]-piperazine, 5, 1-[2-[[2-[(2-aminoethyl)amino]ethyl]-
amino]ethyl]-piperazine) and mixtures thereof. In accordance with an embodiment, the
15 hardener is a mixture of polyetheramine, isophoronediamine(IPDA) and aminopolyamide. By way of a specific example example, the hardener is a mixture of IPDA, polyetheramine, polyamide and an aliphatic amine. The hardeners suitable for the presently disclosed hardener component may be prepared by any known method or obtained from commercial sources.

20 In accordance with an embodiment, fumed silica used in the hardener component may be hydrophilic or hydrophobic. Fumed silica is added in an amount in the range of 0.1-10 % w/ w of said hardener component. Said fumed silica may be selected from fumed silica known in the art; or may be obtained from commercial sources. In accordance with an embodiment, the hardener component may further comprise of conventional filler such

as milled glass fibres. In accordance with an embodiment, a color may be additionally included in the hardener component.

The hardener component may be prepared by any known method. In accordance with an embodiment, the hardener component is prepared by mixing one or more hardeners followed by addition of fumed silica. Alternatively, the hardener component may be obtained from commercial sources. For example, Epotec® TH7257G (commercially available from Aditya Birla Chemicals, Thailand) may be used. Alternatively, an intermediate of Epotec® TH7257G (or Epotec® TH7257G without the fillers) may be used as the hardener component. This intermediate is referred to as Epotec® TH7257G premix.

The hardener component may be mixed with the epoxy resin component in a quantity depending on formulation stoichiometry. In accordance with an embodiment, the hardener component is added in an amount in a range of 25-35 % w/w of the total weight of the adhesive composition. In accordance with an embodiment, the epoxy resin component and the hardener component are added in a w/w ratio in a range of 100: 40 to 100:50. Preferably, the epoxy resin component and the hardener component are mixed in a w/w ratio of 100:45.

In accordance with an embodiment, the adhesive composition may further comprise optional additives. Optional additives may include but are not limited to other resins, stabilizers, plasticizers, catalyst de-activators, dyes, pigments, thixotropic agents, photo initiators, latent catalysts, inhibitors, surfactants, solvents, fluidity control agents, diluents that aid processing, adhesion promoters, flexibilizers, toughening agents, fire retardants, and mixtures thereof.

Any known method may be used to mix the epoxy resin component, the hardener component and any optional additive. Preferably, the mixing is carried out in planetary mixer/stirrer.

The adhesive composition of the present disclosure can be applied to a surface of metal, plastic, fiberglass, or another material that the adhesive composition can bond to. The adhesive composition may be applied to one or between one or more surfaces and then cured. Any known method of applying the adhesive composition may be used. These
5 include, for example, static mixer, rolling coating, and the like. The curing of the adhesive composition may be carried out by heating. For heat curing, the adhesive composition is subjected to heating at a predetermined temperature for a predetermined period of time. The temperature and time required for curing would depend on the epoxy resins and the hardeners used in the adhesive composition.

10

Specific Embodiments are described below

Such an epoxy resin component for an adhesive composition, the epoxy resin component comprising an epoxy resin premix; and a filler composition comprising cenospheres and wollastonite in a w/w ratio in a range of 1:0.01–1:5.

15 Such an epoxy resin component, wherein the filler composition comprises cenospheres and wollastonite in a w/w ratio in a range of 1:1 to 1:3.

Such an epoxy resin component, wherein the filler composition is present in an amount in a range of 10-30 % w/w of said epoxy resin component.

20 Such an epoxy resin component, wherein the filler composition is present in an amount in a range of 15-25 % w/w of said epoxy resin component.

Such an epoxy resin component, wherein cenospheres have a particle size in a range of 100-150 μ .

Such an epoxy resin component, wherein wollastonite has a particle size in a range of 100-200 μ .

Such an epoxy resin component, wherein wollastonite has an aspect ratio in a range of 5:1- 30:1.

Such an epoxy resin component, wherein wollastonite has an aspect ratio in a range of 10:1- 20:1.

5 Such an epoxy resin component, wherein wollastonite is in surface modified form.

Such an epoxy resin component, having a viscosity in a range of about 300-700 Pa.S at 25°C.

Such an epoxy resin component, having a thixotropic index in a range of about 3-5.

Such an adhesive composition comprising about 65-75 % w/w of said epoxy resin compo-
 10 nent; and about 25-35 % w/w of a hardener component.

Examples

The following examples are provided to explain and illustrate the preferred embod-
 iments of the present disclosure and do not in any way limit the scope of the invention as
 15 described and claimed:

Example 1: Preparation of the adhesive composition in accordance with the present disclo-
 sure.

Composition of the adhesive composition has been illustrated in Table 1:

20

Table 1

Materials / Formulation	Ex 1 (g)
Epoxy Resin Component	
Epotec® YD1535G premix	71.1

Cenospheres (CIL150)	8.2
Wollastonite (KEMOLIT® KSV-51)	15.4
Fumed silica	5.3
Total	100
Hardener Component	
TH7257G premix	39.6
Fumed silica	3.6
Mixing Ratio (Epoxy Resin Component: Hardener Component)	100:43.2

The epoxy resin component was prepared using 71.1 parts by weight of epoxy resin premix (Epotec® YD1535G premix) to which 8.2 parts by weight of cenosphere (CIL150), 15.4 parts by weight of wollastonite (KEMOLIT® KSV-51) and 5.3 parts by weight of fumed silica were added and mixed in a blender at room temperature. The epoxy resin component and the hardener component were mixed at 25°C in a ratio of 100:43.2 (w/w) to obtain the adhesive composition. The adhesive composition was shaped to be a desired specimen, and was cured in the oven at specific conditions to determine the properties thereof. Weight reduction, sagging, viscosity were measured using conventional methods. Peel strength was measured by Standard ISO11339. Tensile lap shear and fracture toughness were determined by Standard DINEN1465 and ISO13586 respectively.

Example 2: Preparation of the adhesive composition in accordance with present disclosure
(Hardener contains milled glass fibres (MGF)).

Table 2

Materials / Formulation	Ex 2 (g)
Epoxy Resin Component	
Epotec® YD1535G premix	71.1
Cenospheres (CIL150)	8.2
Wollastonite (KEMOLIT® KSV-51)	15.4
Fumed Silica	5.3
Total	100
Hardener Component	
Epotec® TH7257G	43
Milled Glass Fibers	2
Mixing Ratio (Epoxy Resin Component: Hard- ener Component)	100:45

The epoxy resin component was prepared using 71.1 parts by weight of epoxy resin premix (Epotec® YD1535G premix) to which 8.2 parts by weight of cenosphere (CIL150), 15.4 parts by weight of wollastonite (KEMOLIT® KSV-51) and 5.3 parts by weight of fumed silica were added and mixed in a blender at room temperature. The hardener component was prepared by mixing Epotec® TH7257G and milled glass fibers. The epoxy resin component and the hardener component were mixed at 25°C in a ratio of 100:45 (w/w) to obtain the adhesive composition. The adhesive composition was shaped to be a desired specimen, and was cured in the oven at specific condition for each testing methods (specified in Example 1).

Comparative Example 1: The properties of the epoxy resin component and the adhesive composition prepared in Examples 1 and 2 were compared with that of a conventional adhesive composition (S1). The composition of conventional adhesive composition (S1) is provided in Table 3. The method illustrated in Example 1 was used to prepare the conventional adhesive composition.

Table 3

Materials / Formulation	S1 (g)
Epoxy Resin Component	
Epotec® YD1535G premix	71
Milled glass fibre + Fumed silica	29
Total	100
Hardener Component	

Epotec® TH7257G	45
Mixing Ratio	100:45

Properties of Ex 1 and Ex 2 were compared with S1 and have been tabulated in Table 4.

Table 4

5

Properties	S1	Ex 1	Ex 2
Epoxy resin component			
Viscosity of resin @ 25°C(Pa. S)	560	565±10	560±20
Thixotropic Index	4.27	4.1	4.1
Adhesive composition			
Weight reduction (%)	-	8.2	8.4
Shrinkage * (sq mm)	2104.63	2600	2300
Sagging or Slump**	Slump = 5.5	Sagging = 5	Sagging = 14±4
Tensile lap shear strength (FRP-FRP) Thickness 1 mm (MPa)	23.552	18.5±0.6	19.3±1.1
Peel strength (steel/steel) (N/mm)	6.10±0.15	8.16±0.12	8.30±0.12

Density of cured sample (g/cm ³)	1.190	1.093	1.090
Fracture toughness (60°Cx1.5h + 70°Cx7h)			
Critical stress intensity factor (K _{1c}) (Mpa•m ^{1/2})	2.53	2.05	2.11
Critical strain energy release rate (G _{1c}) (J/m ²)	2930	2500	2530

* Values less than 3000 are preferred.

** Values less than 20 are the optimum range

5 **Observations:** Weight reduction was observed in the adhesive composition of the present disclosure as compared to the conventional adhesive composition. It was further observed that the shrinkage, peel strength of the disclosed adhesive composition is improved as compared to the conventional adhesive composition.

10 Comparative Example 2: Various samples of adhesive compositions comprising solely cenospheres (C1, C2)/ wollastonite (W1, W2) were also prepared. The properties of these adhesive compositions were compared with that of the conventional adhesive composition (S1). The composition and the properties of said samples vis-à-vis the conventional adhesive composition have been tabulated in Table 5.

Table 5

Ingredients/ Properties	Unit	S1	C1 (CIL150)	C2 (CIL100)	W1 (KEMOLI T[®] KSV-51)	W2 (Fillex[®] 11-AF1)
Composition						
Epoxy Resin Component						
Epotec [®] YD1535G premix	g	71	71.7	71.7	71	71
Milled glass fi- bers + Fumed sil- ica	g	29				
Cenospheres	g	-	23.3	23.3	-	-
Wollastonite	g	-			23	23
Fumed Silica	g		5	5	6	6
Total	g	100	100	100	100	100
Hardener Component						
Epotec [®] TH7257G	g	45	46	46	45	45
Mixing Ratio	-	100:45	100:46	100:46	100:45	100:45

Properties						
Viscosity of epoxy resin component @ 25°C (by Rheometer)	Pa.S	560	1100	841	726	681
Thixotropic Index	-	4.27	4.49	4.1	7.32	6.15
Shrinkage of adhesive composition	MPa	2536, 2140	3000	3000	2078	-
Weight reduction of adhesive composition	%	-	23	21	1.7	1.5

Observations: The epoxy resin component comprising solely cenosphere (C1, C2)/ wollastonite (W1, W2) were observed to have reduced weight but much higher viscosity and / or thixotropic index as compared to the epoxy resin component of the conventional adhesive composition, making the former unsuitable for use in a structural adhesive. Also if the amount of cenosphere/ wollastonite is lowered, the sagging and shrinkage properties will suffer. However, cenospheres and wollastonite when used in a ratio in a range of 1:0.01- 1:5 provide an epoxy resin component having desired viscosity of 300-700 Pa.S at 25°C and thixotropic index of 3-5. This indicates that cenospheres and wollastonite when combined in the ratio of 1:0.01- 1:5 act in a synergistic manner to achieve the desired rheological properties.

Industrial Applicability

The adhesive composition comprising the filler composition of the present disclosure may be used to bond relatively large structures including but not limited to, aerodynamic wings, wind turbine blades, and automobile components. The filler composition
5 formed by cenospheres and wollastonite employed is cheap and easy to manufacture. An unexpected synergy between cenospheres and wollastonite, employed in accordance with the present disclosure, contributes to or provides a number of improvements in the adhesive composition, such as: viscosity and thixotropic index are in desirable range, the overall weight reduces, shrinkage and peel strength improves. Weight reduction, particularly in
10 windmill application plays an important role in reducing the weight of the blade and thereby improving the efficiency of windmill. Similar effect can be observed when the adhesive composition of the present disclosure is applied to other applications.

We Claim:

1. An epoxy resin component for an adhesive composition, the epoxy resin component comprising:
5 - an epoxy resin premix; and
 - a filler composition comprising cenospheres and wollastonite in a w/w ratio in a range of 1:0.01–1:5.
2. An epoxy resin component as claimed in claim 1, wherein the filler composition
10 comprises cenospheres and wollastonite in a w/w ratio in a range of 1:1 to 1:3.
3. An epoxy resin component as claimed in claim 1, wherein the filler composition is present in an amount in a range of 10-30 % w/w of said epoxy resin component.
- 15 4. An epoxy resin component as claimed in claim 3, wherein the filler composition is present in an amount in a range of 15-25 % w/w of said epoxy resin component.
5. An epoxy resin component as claimed in claim 1, wherein cenospheres have a particle size in a range of 100-150 μ .
20
6. An epoxy resin component as claimed in claim 1, wherein wollastonite has a particle size in a range of 100-200 μ .
7. An epoxy resin component as claimed in claim 1, wherein wollastonite has an aspect ratio in a range of 5:1- 30:1.
25

8. An epoxy resin component as claimed in claim 7, wherein wollastonite has the aspect ratio in the range of 10:1- 20:1.
9. An epoxy resin component as claimed in claim 1, wherein wollastonite is in surface
5 modified form.
10. An epoxy resin component as claimed in claim 1 having a viscosity in a range of about 300-700 Pa.S at 25°C.
- 10 11. An epoxy resin component as claimed in claim 1 having a thixotropic index in a range of about 3-5.
12. An adhesive composition comprising:
- about 65-75 % w/w of an epoxy resin component claimed in any of the preced-
 - 15 ing claims; and
 - about 25-35 % w/w of a hardener component.

INTERNATIONAL SEARCH REPORT

International application No
PCT/IB2016/057420

A. CLASSIFICATION OF SUBJECT MATTER
INV. C09J163/00 C08L63/00
ADD.
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)
C09J C08L

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 practicable, search terms used)
EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2008/251202 A1 (EAGLE GLENN G [US] ET AL) 16 October 2008 (2008-10-16) page 9; examples 8,9; table 5 page 9; example 11; table 7 paragraphs [0081] - [0087] paragraph [0061] page 4, paragraph 36	1-12
A	EP 0 488 949 A2 (CIBA GEIGY AG [CH]) 3 June 1992 (1992-06-03) claims 1-20 tables 5, 7	1-12
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Further documents are listed in the continuation of Box C.

See patent family annex.

* Special categories of cited documents :

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Date of the actual completion of the international search 31 March 2017	Date of mailing of the international search report 10/04/2017
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Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Authorized officer Mensah, Laure
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International application No
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