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(54) Title: WATER-BORNE PAINTS BASED ON EPOXY RESINS

(57) Abstract: An aqueous epoxy resin system AB is described comprising an aqueously dispersed epoxy resin A having, on the average, at least one epoxy group per molecule, and a water-soluble or water-dispersible curing agent B which comprises the reaction product of an amine B1 having at least one primary and/or at least one secondary amino group, an adduct B2 of a polyalkylene ether polyol B21 and an epoxide component B22, and an aromatic compound B3 having at least one acidic group selected from the group consisting of hydroxyl and carboxyl groups, which system can be applied by rolling, spraying or brushing to provide corrosion protection on base metals.

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Water-Borne Paints Based on Epoxy Resins

The present invention relates to water-borne paints based on epoxy resins. Specifically, it relates to paints comprising a combination of a water-borne epoxy resin binder and a water-borne epoxy resin-based curing agent.

Water-borne paints comprising a water-dilutable epoxy resin and a water-dilutable curing agent based on reaction products of epoxy resins and amines have been described, i. a., in EP 0 000 605 B1. Such systems provide good corrosion resistance especially on base metal substrates such as steel sheets.

It is an object of this invention to improve the corrosion resistance further, particularly against brine as evidenced in the salt spray test.

This object has been achieved by providing an aqueous epoxy resin system **AB** comprising an aqueously dispersed epoxy resin **A** having, on the average, at least one epoxy group per molecule, and a water-soluble or water-dispersible curing agent **B** which comprises the reaction product of an amine **B1** having at least one primary and/or at least one secondary amino group, an adduct **B2** of a polyalkylene ether polyol **B21** and an epoxide component **B22**, and an aromatic compound **B3** having at least one acidic group selected from the group consisting of hydroxyl and carboxyl groups.

The invention also relates to a water-soluble or water-dispersible curing agent composition **B** which comprises the reaction product of an amine **B1** selected from the group consisting of amines having at least one primary amino group and amines having at least one

secondary amino group, an adduct **B2** of a polyalkylene ether polyol **B21** and an epoxide component **B22**, and an aromatic compound **B3** having at least one acidic group selected from the group consisting of hydroxyl and carboxyl groups.

The invention also relates to coating compositions comprising the said aqueous epoxy resin system **AB** and at least one of pigments, fillers, further binders, and additives.

The invention further relates to a process of coating metal based substrates with the said aqueous epoxy resin system **AB**, which process comprises applying the said aqueous epoxy resin system **AB** to a metal based substrate by spraying, brushing, or rolling.

The water-soluble or water-dispersible curing agent **B** comprises the reaction product of an amine **B1** selected from the group consisting of amines having at least one primary amino group and amines having at least one secondary amino group, an adduct **B2** of a polyalkylene ether polyol **B21** and an epoxide component **B22**, and an aromatic compound **B3** having at least one acidic group selected from the group consisting of hydroxyl and carboxyl groups.

While reaction products of novolak-based epoxy resins with mononuclear aromatic compounds having at least two hydroxyl groups have been described in DE 14 70 785 A1, such reaction products merely serve to transform the otherwise liquid epoxy resins to solid ones. Besides dicarboxylic aromatic acids and dihydroxy aromatic compounds, also aromatic hydroxycarboxylic acids such as salicylic acid are disclosed. Reaction products of polyalkylene glycol modified epoxy resins, amines and the said aromatic compounds are neither mentioned nor made obvious.

The amines **B1** are preferably purely aliphatic amines, i. e. those where only aliphatic organic groups which may be linear, branched or cyclic are present in the molecule. They may, however, also comprise such amines where the amine nitrogen atom is bound to an aliphatic carbon atom, while other parts of the molecule may also comprise aromatic moieties. It is preferred that the amine **B1** is a diprimary diamine. Such amines are, e. g. the isomeric xylylene diamines, preferably meta-xylylene diamine. Preferred aliphatic amines are 1,4-diaminobutane, hexamethylene diamine, the isomeric trimethyl hexamethylene diamines, the isomeric bis-(aminomethyl) cyclohexanes, isophorone diamine, diethylene triamine, triethylene tetramine, tetraethylene pentamine, the higher diaminopolyethylene imines, 1-(2-aminoethyl)piperidine, 1-(2-aminoethyl)piperazine, N-(2-aminoethyl)-1,3-propane diamine, and N,N-bis(2-aminoethyl)-1,3-propane diamine as well as mixtures of two or more of these amines. Especially preferred are meta-xylylene diamine, isophorone diamine, diethylene triamine, triethylene tetramine, tetraethylene pentamine, and mixtures thereof.

The polyether polyols **B21** are preferably addition products of alkylene oxides to polyfunctional alcohols or mixtures thereof, such as the diols ethylene and propylene glycol, neopentyl glycol, 1,4-butane diol, triols like glycerol, and higher polyfunctional alcohols such as pentaerythritol or sorbitol. It is also preferred that the mass fraction of such polyvalent (i. e. trifunctional or higher) alcohols does not exceed 10 % of the mass of all alcohols. Especially preferred are polyethylene glycol and copolymers comprising oxyethylene and oxypropylene moieties, where at least a mass fraction of 20 %, more preferably at least 30 %, is constituted of oxyethylene groups.

The epoxy compounds **B22** are preferably glycidyl ethers of dihydric alcohols or phenols, or novolaks, or glycidyl esters of dicarboxylic acids. Among the phenols, resorcinol, hydro-

quinone, 2,2-bis-(4-hydroxyphenyl)-propane (bisphenol A), mixtures of isomers of dihydroxydiphenylmethane (bisphenol F), tetrabromo bisphenol A, 4,4'-dihydroxydiphenyl cyclohexane, 2,2-bis-(4-hydroxy-3-methylphenyl)-propane, 4,4'-dihydroxydiphenyl, 4,4'-dihydroxybenzophenone, 1,1-bis-(4-hydroxyphenyl)-ethane, 2,2-bis-[4-(2'-hydroxypropoxy)-phenyl]-propane, 1,1-bis-(4-hydroxyphenyl)-isobutane, 2,2-bis(4-hydroxy-3-tert.-butyl-phenyl)-propane, bis-(2-hydroxynaphthyl)-methane, 1,5-dihydroxy naphthalene, tris-(4-hydroxyphenyl)-methane, bis-(4-hydroxyphenyl)ether, bis-(4-hydroxyphenyl)sulphone, as well as products of halogenation and hydration of the compounds mentioned. Epoxy resins based on bisphenol A are especially preferred.

They preferably have a specific epoxide group content (amount of substance of epoxide groups in the said epoxy compounds divided by the mass of the said epoxy compounds) of from 0.5 mol/kg to 10 mol/kg, especially preferably from 1.0 mol/kg to 7.0 mol/kg. They can be saturated or unsaturated, aliphatic and mixed aliphatic-aromatic compounds and have at least one, preferably at least two epoxide groups per molecule, on the average. They can also have hydroxyl groups in their molecules. It is also possible to use diepoxy alkanes made by epoxidation of diolefins. Particularly preferred are the diglycidyl ether of bisphenol A (BADGE) and bisphenol F, and epoxy resins based on advancement products of BADGE and bisphenol A. These BADGE based resins are commonly referred to as type 1, type 5, type 7 etc. resins, according to their degree of polymerisation. It is particularly preferred to use a mixture of BADGE and type 1 epoxy resins.

The adduct is made by reacting the polyether polyols **B21** and the epoxy componds **B22** in the presence of a catalyst selected from the group consisting of Lewis acids such as boron trifluoride and complexes thereof with ethers or amines.

The aromatic compound **B3** having at least one acidic group selected from the group consisting of hydroxyl and carboxyl groups is preferably derived from benzene, diphenyl, or naphthalene, or homologues thereof, i. e., lower alkyl substituted benzenes, diphenyls or naphthalenes such as toluene, xylene, 1-or 2-methyl naphthalene, 1,2- or 1,3- or 1,4- or 1,8- or 2,3-dimethyl naphthalene, or the corresponding ethyl-, propyl- or butyl-substituted benzenes, diphenyls, or naphthalenes, where linear, branched and mixed substituents are also encompassed. It is preferred to have at least two of the said acidic groups, with at least two hydroxyl groups or at least one hydroxyl group and at least one carboxyl group being especially preferred. Preferably, the aromatic compound **B3** has at least two acidic groups whereof at least one is a hydroxyl group. Also preferred are aromatic compounds **B3** having at least two acidic groups whereof at least one is a hydroxyl group and one is a carboxyl group. Compounds **B3** having no hydroxyl groups have been found to be less satisfactory. Particularly preferred are resorcinol, salicylic acid, p-hydroxybenzoic acid, 2,4-dihydroxybenzoic acid (beta-resorcylic acid), 2,5-dihydroxybenzoic acid (gentisic acid), 2,6-dihydroxybenzoic acid (gamma-resorcylic acid), 3,4-dihydroxybenzoic acid (proto-catechuic acid), 3,5-dihydroxybenzoic acid (alpha-resorcylic acid) 2-hydroxy-3-naphthoic acid, 2-hydroxy-6-naphthoic acid and alkyl derivatives of these such as 2-hydroxy-3-, -4-, and -5-methyl-benzoic acid (cresotic acids).

The epoxy resin **A** can be a non-modified or hydrophilically modified epoxy resin, selected from the same group as stated under **B22**. Hydrophilic modification of the epoxy resins is made according to the usual practice, by adding an adduct such as those described as **B2** as an emulsifier to the epoxy resin, and dispersing the mixture, or by dispersing an unmodified epoxy resin in an aqueous dispersion of the emulsifier.

The aqueous epoxy resin system **AB** of the present invention is preferably made in a multi-step process, comprising

- as the first step, preparing an adduct **B2** of a polyalkylene ether polyol **B21** and epoxy resins or compounds **B22**, using Lewis acids or complex salts thereof as catalysts,
- adding **B2** to the amine **B1** in which the aromatic compound **B3** has been dissolved, the amino groups of said amine **B1** reacting with the epoxy groups of **B2**, preferably by consuming at least 90 % of these epoxy groups, more preferred, at least 95 %, and especially preferred, at least 98 %, of the epoxy groups of **B2**,
- optionally adding an aliphatic polycarboxylic acid **B4** which reacts with residual amine **B1** at elevated temperature to form a polyamide or amide oligomer,
- removing the water formed by the reaction, preferably by azeotropic distillation,
- optionally adding a monoepoxy compound or a mixture of monoepoxy compounds **B5**,
- and optionally adding a polyamine, preferably a diamine **B6**,
- optionally, neutralisation of the resulting product with acid to convert at least 20 % of the amino groups into the respective cations, and
- dispersing the optionally neutralised reaction product in water, and
- optionally adding further water to adjust the mass fraction of solids to between 40 % and 80 %.

The aromatic compound **B3** is preferably dissolved in the amine **B2** so that there results a clear solution free from undissolved solid material remaining from **B3**. An alternative route for the second step is to add the amine **B1** in which the aromatic compound **B3** has been dissolved to the adduct **B2**.

The curing agent **B** made in accordance with the steps as detailed supra is either admixed

to an aqueous dispersion of an epoxy resin **A** which may be self-emulsified, such as by reaction of an epoxy resin with polyoxyethylene glycol, or externally emulsified by addition of an appropriate emulsifier, or an unmodified epoxy resin **A** is emulsified in water comprising the curing agent **B**, all variants leading to the epoxy resin system **AB**.

The polycarboxylic, preferably dicarboxylic, aliphatic acid **B4** is optionally added to consume residual amine. It has surprisingly been found that the small portion of polyamide or amide oligomer formed in this reaction further enhances the properties of the aqueous epoxy resin system of the present invention. The acid **B4** is preferably selected from the group consisting of adipic acid, glutaric acid, succinic acid, mixtures thereof, and also, other aliphatic dicarboxylic acids such as C_7- to C_{12}- acids, and fatty acid dimers having up to 50 carbon atoms.

It has also been found advantageous to include a reaction step where residual amine or also, amide oligomer is consumed by reaction with a monoepoxide **B5**, such as butyl glycidyl ether, hexyl glycidyl ether, 2-ethylhexyl glycidyl ether, or also, aromatic mono-epoxy compounds such as cresyl or xylenyl glycidyl ethers. It is especially preferred to employ mixtures of aliphatic and aromatic glycidyl ethers.

It is also preferred to add further primary amine **B6**, especially diamine, in the last step before neutralisation, to enhance the hydrophilicity of the reaction product formed. Such diamines are preferentially chosen from diamines having two primary amino groups, namely cycloaliphatic diamines such as isophorone diamine, branched aliphatic amines such as trimethylhexane diamine isomers, and mixed aromatic-aliphatic amines having the amino groups bound to aliphatic carbon atoms such as meta-xylylene diamine and tetramethyl xylylene diamine.

It is preferred to include at least one of the optional steps of adding an aliphatic polycarboxylic acid **B4**, adding a monoepoxide **B5**, and adding a further amine **B6**.

The aqueous epoxy resin systems of the present invention, preferably made according to the process outlined supra, can be used in any epoxy resin based coatings. It has been found that such aqueous systems are especially suited for coating of base metals where they exhibit a markedly improved corrosion protection, particularly in contact with brine.

The invention is further illustrated in the following examples.

Example 1 Preparation of a polyalkylene glycol-modified epoxy resin

642 g of bisphenol A diglycidyl ether were mixed with 356 g of polyethylene glycol (number average molar mass 600 g/mol) and heated to 125 °C whereafter 2.4 g of a boron trifluoride benzylamine complex (®Anchor 1040, Air Products) were added, and the reaction mixture was kept at 140 °C until the specific content of epoxide groups had reached a value of approximately 2.4 mol/kg.

Example 2 Preparation of a Curing Agent

19 g of salicylic acid were dissolved in 38.4 g of meta-xylylene diamine which had been heated to about 50 °C. Stirring was continued until a clear solution had formed. This solution was further heated to about 80 °C, and 17.5 g of the modified epoxy resin of Example 1 were added. Deionised water (75 g) was then added in two portions. A resin solution having an amine number of 219 mg/g and a mass fraction of solids of about 50 % was obtained.

Example 3 Preparation of Further Curing Agents

Example 2 was repeated, while replacing the salicylic acid in turn with 5.0 g of phthalic anhydride (Example 3.1), 5.0 g of isophthalic acid (Example 3.2), 5.0 g of resorcinol (Example 3.3), 5.0 g of benzoic acid (Example 3.4), 5.0 g of bisphenol A (Example 3.5), 10.0 g of the monohydrate of gallic acid (Example 3.6), 10.0 g of isophthalic acid (Example 3.7), 10.0 g of resorcinol (Example 3.8), 10.0 g of benzoic acid (Example 3.9), 10.0 g of bisphenol A (Example 3.10), 5.0 g of tetrahydrophthalic acid anhydride (Example 3.11), by nothing (Example 3.12, comparative), and replacing the amine (meta-xylylene diamine) by 4.8 g of isophorone diamine while keeping the same amount of salicylic acid in the mixture (Example 3.13).

Example 4 Preparation of Coating Compositions

Clearcoats were prepared from 50 g of an aqueous epoxy resin dispersion of type 1 solid resin (®Beckopox EP 384w/53 WAMP, mass fraction of solids of 53 %, Cytec Surface Specialties Germany GmbH & Co. KG) and the mass of curing agents of Example 3 as stated in table 1. The quantities of the curing agents were adjusted to have the same amount of substance of active amine hydrogen atoms.

Table 1 Composition of Clearcoats and Development of Pendulum Hardness

Example	Curing Agent of Example	Mass of Curing Agent in g	Pendulum Hardness* (König) in s
4.1	3.1	3.8	105
4.2	3.2	3.8	126
4.3	3.3	3.8	150
4.4	3.4	3.8	153
4.5	3.5	3.8	153
4.6	3.6	3.8	153
4.7	3.7	4.1	73
4.8	3.8	4.1	160
4.9	3.9	4.1	163
4.10	3.10	4.1	167
4.11	3.11	3.8	150
4	2	7	171

*as tested after seven days' drying at room temperature (23 °C), 50 μm dry film thickness, on glass plates

As can be seen, with the exception of Examples 4.1, 4.2 and 4.7 (where aromatic compounds with only carboxyl groups have been used), the pendulum hardness has a good level of from 150 to 170 seconds.

Example 5 Coatings Test

In a further test, the corrosion resistance of pigmented coatings comprising the aqueously dispersed epoxy resin as used in Example 4 and the curing agents of Example 2 (where salicylic acid as compound **B3** is present, and the amine **B1** is meta-xylylene diamine) and

of Examples 3.12 (no compound **B3**) and 3.13 (salicylic acid as compound **B3** and isophorone diamine as amine **B1**).

A pigmented binder dispersion was prepared using the following recipe:

A pigmented paint was made by charging a mixture of 36.2 g of ®Beckopox EP 384w/53 WAMP (as in Example 4), 1.3 g of ®Additol VXW 6208/60 (a polymeric non-ionic wetting and dispersing agent, Cytec Surface Specialties Austria GmbH), 0.3 g of ®Additol VXW 6393 (mineral oil based defoamer, Cytec Surface Specialties Austria GmbH), and 7.4 g of deionised water, adding thereto 21.6 g of a white titanium dioxide pigment (®Kronos 2190, Kronos International Inc.), 0.25 g of a yellow iron oxide pigment (®Bayferrox 3920, Lanxess Deutschland GmbH), 0.8 g of a black iron oxide pigment (®Bayferrox 306, Lanxess Deutschland GmbH), 2.5 g of barium sulphate (®EWO, Dr. Rudolf Alberti GmbH & Co.), 4.20 g of zinc phosphate (®Heucophos ZPO, Heubach GmbH), and 11.3 g of micronised talc (®Microtalc IT extra, Norwegian Talc AS), dispersing the mixture in a dissolver for about thirty minutes, and then letting down by adding further 10.8 g of the epoxy resin dispersion, 0.65 g of a coalescing agent (2,2,4-trimethyl-1,3-pentanediol-monoisobutyrate, ®Texanol, Eastman Chemical Company), 0.7 g of a polyurethane thickener (®Additol VXW 6388, Cytec Surface Specialties Austria GmbH), and 2 g of deionised water. 100 g each of this pigmented binder dispersion was mixed with 5 g of the curing agent of Example 2 (paint 5.1), 5.3 g of the curing agent of Example 3.13 (paint 5.2), and for a comparative test, 4.3 g of the curing agent of Example 3.12 (paint 5.3). These coating compositions were applied by spraying on non-treated steel panels (®Gardobond OC) and dried for seven days at room temperature (23 °C) and 50 % relative humidity to yield dry film thicknesses of between 50 µm and 60 µm. These were treated in a salt spray test in accordance with EN ISO 9227 (NSS Test), each sample having been tested with and without central scratch. The

surface appearance was assessed according to EN ISO 4628.

The results are listed in Table 2.

Table 2 Salt Spray Test

Paint	5.1		5.2		5.3	
Scratch	yes	no	yes	no	yes	no
Dry Film Thickness in µm	55	55	58	61	51	61
Blistering **after 24 hours**	OK	OK	OK	OK	OK, 2 rp	OK, 9 rp
Blistering at scratch	(S1) p		(S2) p		(S2) p	
creep from scratch in mm	1.5		1.5		4.5	
Blistering **after 48 hours**	OK	OK	OK	OK	OK, 2 rp	OK, 9 rp
Blistering at scratch	(S1) p		2 (S2)		1 (S3)	
creep from scratch in mm	1.5		1.5		6	
Blistering **after one week**	OK	OK	OK	OK	2 (S4)	1 (S2)
Blistering at scratch	2 (S2)		2 (S3)			
creep from scratch in mm	5		4		delam.	
Blistering **after two weeks**	OK	OK	OK	OK		5 (S3)
Blistering at scratch	3 (S2)		2 (S3)			
creep from scratch in mm	8		7			
Blistering **after three weeks**	1 (S3)	1 rp	(S2) p	(S2)		
Blistering at scratch	3 (S3)		2 (S4)			
creep from scratch in mm	13		13			

OK: no blisters

p: partially

rp: rust points

delam.: complete delamination

Similarly favourable results have been obtained particularly for other aromatic hydroxycarboxylic acids as compounds **B3.** As can be seen from this table, the addition of a compound **B3** markedly improves the corrosion resistance and extends the time to failure, while the nature of the amine component in the curing agent is not critical in this context.

Patent Claims

1. An aqueous epoxy resin system **AB** comprising an aqueously dispersed epoxy resin **A** having, on the average, at least one epoxy group per molecule, and a water-soluble or water-dispersible curing agent **B** which comprises the reaction product of an amine **B1** having at least one primary and/or at least one secondary amino group, an adduct **B2** of a polyalkylene ether polyol **B21** and an epoxide component **B22**, and an aromatic compound **B3** having at least one acidic group selected from the group consisting of hydroxyl and carboxyl groups.

2. The aqueous epoxy resin system **AB** of claim 1 wherein the aqueously dispersed epoxy resin **A** has, on the average, at least two epoxy groups per molecule.

3. The aqueous epoxy resin system **AB** of claim 1 wherein the aromatic compound **B3** has at least two acidic groups whereof at least one is a hydroxyl group.

4. The aqueous epoxy resin system **AB** of claim 1 wherein the aromatic compound **B3** has at least two acidic groups whereof at least one is a hydroxyl group and one is a carboxyl group.

5. The aqueous epoxy resin system **AB** of claim 1 wherein the aromatic compound **B3** is salicylic acid.

6. The aqueous epoxy resin system **AB** of claim 1 wherein the amine **B1** is a diprimary

diamine.

7. The aqueous epoxy resin system **AB** of claim 1 wherein the water-dispersible curing agent **B** has an amine number of from 120 mg/g to 300 mg/g.

8. A process for the preparation of the water-dispersible curing agent **B** as claimed in claim 1 comprising the steps of

- preparing an adduct **B2** of a polyalkylene ether polyol **B21** and epoxy resins or compounds **B22**, using Lewis acids or complex salts thereof as catalysts,
- adding **B2** to the amine **B1** in which the aromatic compound **B3** has been dissolved, the amino groups of said amine **B1** reacting with the epoxy groups of **B2**, preferably by consuming at least 90 % of these epoxy groups, more preferred, at least 95 %, and especially preferred, at least 98 %, of the epoxy groups of **B2**,
- optionally adding an aliphatic polycarboxylic acid **B4** which reacts with residual amine **B1** at elevated temperature to form a polyamide or amide oligomer,
- removing the water formed by the reaction, preferably by azeotropic distillation,
- optionally adding a monoepoxy compound or a mixture of monoepoxy compounds **B5**,
- and optionally adding a polyamine, preferably a diamine **B6**,
- optionally, neutralisation of the resulting product with acid to convert at least 20 % of the amino groups into the respective cations, and
- dispersing the optionally neutralised reaction product in water, and
- optionally adding further water to adjust the mass fraction of solids to between 40 % and 80 %.

9. A process for the preparation of the aqueous epoxy resin system **AB** of claim 1

wherein a water-dispersible curing agent **B** is made by the process of claim 8, and wherein either the said water-dispersible curing agent **B** is admixed to an aqueous solution or dispersion of an epoxy resin **A** having a specific epoxide group content of from 0.5 mol/kg to 10 mol/kg, or an aqueous dispersion of the said curing agent **B** is charged, and the epoxy resin **A** is emulsified in the said dispersion of the curing agent.

10. A method of use of the aqueous epoxy resin system as claimed in claim 1 for coating of base metals substrates comprising applying the said epoxy resin system to the said substrate by rolling, brushing or spraying.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2008/059898

A. CLASSIFICATION OF SUBJECT MATTER
INV. C09D163/00 C08G59/62 C08G59/50 C08G59/18

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)
C09D C08G

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, CHEM ABS 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 1 518 875 A (COGNIS DEUTCHLAND GMBH & CO KG [DE]) 30 March 2005 (2005-03-30) abstract page 3, paragraph 15 - paragraph 17 page 4, paragraph 24 - paragraph 25 page 4, paragraph 29 - page 5 page 6, paragraph 39 - paragraph 43 page 9, paragraph 62; examples B1-B5; table 1	1-3,6,7
Y		1-10
X	EP 1 647 584 A (CYTEC SURFACE SPECIALTIES AUST [AT]) 19 April 2006 (2006-04-19) claim 2 page 3, paragraphs 13,16 page 5, paragraph 20	1,2

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[X] Further documents are listed in the continuation of Box C.

[X] See patent family annex.

* Special categories of cited documents :

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Date of the actual completion of the international search	Date of mailing of the international search report
28 October 2008	07/11/2008

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INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2008/059898

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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