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(54) **Title:** PARTICULATE FILLER FOR REACTIVE PLASTICS BASED ON POLYESTER RESINS

(57) **Abstract:** Particulate filler for polyester resin-based reactive plastics is composed of a pulp fraction of the finely ground waste of a glass fibre filled polyester resin-based reactive plastic having a particle size in the range from 0.005 to 0.1 mm, especially of the finely ground waste of a glass fibre filled unsaturated polyester resin.

PARTICULATE FILLER FOR REACTIVE PLASTICS BASED ON POLYESTER RESINS

Field of the invention

The invention relates to a particulate filler for polyester resin-based reactive plastics from waste polyester resin-based reactive plastics filled with glass fibres, in particular from the production of glass laminates. Further, the invention relates to a blend of reactive plastic polyester resin containing this filler.

State of the art

Nowadays, polyester resin based reactive plastics filled with glass fibre are widely used for the production of automotive parts and components in order to reduce their weight and thus their fuel consumption.

Reactive plastics are mainly used in applications where products are demanding in terms of elevated temperature, resistance, non-flammability, low shrinkage and require good surface properties.

The production of such parts is both structurally and technologically demanding and the production itself and the subsequent finishing operations inevitably result in waste. This waste cannot be further sorted and consists of a polyester resin matrix and glass fibre reinforcement. Since these two fractions are already irreversibly linked by the curing process, they cannot be separated. The material thus becomes waste with a predetermined disposal method - as mixed waste. It is therefore a product that hardly finds a place in the industrial or consumer sphere anymore.

This is a waste that is inherently unprocessable by conventional technologies typical of thermoplastics and/or reactive plastics. It is therefore largely landfilled or used as part of mixtures of solid alternative fuels in incinerators.

Existing solutions for the recycling or recovery of such waste are highly specific and difficult to apply on a larger scale from a technological or economic point of view.

One of the well-known solutions towards the recovery of waste of reactive plastics based on polyester resins is offered by Chinese patent CN104446144, which relates to a method for recycling waste based on unsaturated polyester resin reinforced with fibre in the production of artificial marble using this waste as a secondary raw material. The recycling method according to the invention consists in crushing the waste and mixing it into an artificial marble mixture

based on unsaturated polyester resin of the following composition: 60 to 65 parts by weight of marble aggregate, 10 to 13 parts by weight of a mixture of unsaturated polyester resin and hardener, 5 to 12.5 parts by weight of FRP waste, 7.5 to 15 parts by weight of filler and 8 to 10 parts by weight of silica sand. In the production of artificial marble, a technological process involving room temperature casting, mechanical vibration, compaction, and heating and curing is used. The forming time is short, the production efficiency is high, the comprehensive performance of the product is good, and the obtained artificial marble has excellent mechanical properties and corrosion resistance. However, it is a highly specific way of using waste based on unsaturated fibre-filled polyester resin, the practical applicability of which is limited to the application at hand.

Another Chinese patent, CN102432771, relates to a method of recycling unsaturated polyester resin from waste polyester films from the production of unsaturated polyester resin based composites reinforced with glass fibre. The main technological scheme of the solution is as follows: 2-methyl-1,3-propylene glycol, waste polyester film and catalyst are added to the reaction vessel for the degradation reaction. Fumaric acid, resinol, diethylene glycol, propylene glycol and a polymerization inhibitor are then added, after which the resulting product is mixed and reacts with monomeric styrene to prepare an unsaturated polyester resin. It is therefore again a use with limited capacity, which is also quite demanding from a technological point of view.

The subject of Polish patent PL302203 is a method of processing waste from hardened unsaturated polyesters, in particular glass fibre reinforced polyesters. The method consists in passing the waste through a methylene chloride extraction process with a weight ratio of methylene chloride to waste of at least 1:1 (preferably 5:1) at room temperature and elevated pressure. The mass after extraction is first filtered and then a portion of the methylene chloride is evaporated. The post-extraction mass containing at least 20% adsorbed methylene chloride is usable in subsequent continuous mixing of resins as a plasticizer or other component of composites (with a boiling point higher than that of methylene chloride, and which do not react with it). The whole is mixed, the solvent is slowly evaporated and then the mixture is subjected to mechanical processing. Again, this is a use with a limited capacity and is technologically and energetically very demanding.

The subject-matter of Czech utility model No 37113 is a functional particulate filler for thermoplastics, which consists of a pulp of finely ground waste reactive plastic (epoxy polyester or phenol-formaldehyde resin) filled with glass fibres, having a particle size ranging from 0.1 to 1.0 mm. This particle size not only allows sufficient homogenization with the polymer

matrix, but also, due to the particle size range and its composition, the particulate filler also acts as a reinforcing (functional) part of the final product. The utility model further discloses polymer compositions comprising mentioned functional particulate filler in a thermoplastic polymer matrix based on polypropylene, high-density polyethylene, acrylonitrile-butadiene-styrene copolymer or polyvinyl chloride. To improve the bonding between polar polymers and glass fibres, a polypropylene-based compatibilizer can be used here. The problem is that the use of this functional particulate filler is limited to thermoplastic polymer matrices only. It is not practically applicable for polyester resin-based reactive plastics due to the particle size range.

The essence of the technical solution

To eliminate the aforementioned shortcomings in the recycling of glass fibre filled polyester resin-based reactive plastics, the particulate filler for polyester resin based reactive plastics according to the presented technical solution contributes to a large extent. The essence of the solution is that this particulate filler is made up of a fraction of finely ground glass fibre-filled polyester resin-based reactive plastic waste, which has a particle size ranging from 0.005 to 0.1 mm. It is advantageous if the fraction of the finely ground reactive plastic waste pulp based on polyester resin is a fraction of the reactive plastic pulp based on unsaturated polyester resin filled with glass fibre.

The essence of the polyester resin reactive plastic blend according to the technical solution comprising a particulate filler is that in a polymer matrix based on a polyester resin, in particular an unsaturated polyester resin, contains 15 to 50% by weight of a particulate filler consisting of a fraction of the pulp of finely ground waste reactive plastic of the same composition as the polymer matrix, filled with glass fibre, where the particle size of the pulp ranges from 0.005 to 0.1 mm.

Compared to most of the known solutions, the solution according to the presented technical solution of waste recycling on the basis of polyester resin filled with glass fibre is advantageous from the technological, energy and therefore economic point of view. In contrast to the known use of the pulp of reactive plastic waste as a functional particulate filler for thermoplastics, there is a difference in principle, which lies in the identified need to use a pulp fraction of finely ground reactive plastic waste based on polyester resin filled with glass fibre, which has a particle size in the range from 0.005 to 0.1 mm. The 0.1 to 1 mm particle size fraction, which is applied as a particle functional filler for thermoplastics, is not applicable in a

polymer matrix based on unsaturated polyester resin due to unsatisfactory technological and utility properties of the prepared blend of reactive plastic polyester resin.

Examples

Example 1

The particulate filler for reactive plastics in the exemplary embodiment consists of a pulp fraction of finely ground waste reactive plastic (Menzolit BMC 3100, BMC TETRADUR TD 492/2) based on an unsaturated glass fibre filled polyester resin having a particle size in the range from 0.005 to 0.1 mm.

In an exemplary embodiment, the blend of reactive plastic polyester resin comprises, in a polymer matrix based on unsaturated polyester resin, 15% by weight of a particulate filler comprising a fraction by weight of finely ground reactive plastic waste of the same composition as the polymer matrix, filled with glass fibre, wherein the pulp particle size ranges from 0.005 to 0.1 mm. Specifically, this particulate filler was used in the mixture of unsaturated polyester resin menzolite BMC 3100, where it replaced the originally contained mineral filler.

This blend is suitable for the production of automotive headlight visor parts using BMC (Bulk Moulding Compound) technology, where a polymer composite material is prepared by mixing a glass fibre reinforcement with a polyester resin and then processed by pressing or injection moulding.

Example 2

The particulate filler for reactive plastics in the exemplary embodiment consists of a pulp fraction of finely ground waste reactive plastic (menzolite BMC 3100) based on an unsaturated glass fibre filled polyester resin having a particle size in the range from 0.005 to 0.1 mm

In an exemplary embodiment, the blend of reactive plastic polyester resin comprises, in a polymer matrix based on unsaturated polyester resin, 50% by weight of a particulate filler comprising a fraction by weight of finely ground reactive plastic waste of the same composition as the polymer matrix, filled with glass fibre, wherein the pulp particle size ranges from 0.005 to 0.1 mm. Specifically, this particulate filler was used in the mixture of unsaturated polyester resin menzolite BMC 3100, where it replaced the originally contained mineral filler.

This blend is suitable for the production of automotive non-visual headlight parts using BMC (Bulk Moulding Compound) technology, where a polymer composite material is prepared

by mixing a glass fibre reinforcement with a polyester resin and then processed by pressing or injection moulding.

Industrial applicability

A particulate filler based on a pulp of finely ground waste of reactive plastic based on unsaturated glass fibre filled polyester resin, particularly from the manufacture of laminate composites, is usable as a filler for reactive plastic polyester resin blends. Especially for the blend of unsaturated polyester resin for BMC technology, where it successfully replaced the mineral filler (talc, calcium carbonate) originally included.

CLAIMS

1. Particulate filler for polyester resin-based reactive plastics, characterized in that it is composed of a pulp fraction of the finely ground waste of a glass fibre filled polyester resin-based reactive plastic having a particle size in the range from 0.005 to 0.1 mm.

2. The particulate filler according to claim 1, characterized in that the pulp fraction of the finely ground waste polyester resin-based reactive plastic is the pulp fraction of the unsaturated glass fibre-filled polyester resin reactive plastic.

3. A blend of a reactive polyester resin, containing a particulate filler, characterized in that the polymer matrix is based on a polyester resin, in particular an unsaturated polyester resin, comprising 15 to 50% by weight of a particulate filler consisting of a pulp fraction by weight of a crumb fraction of finely ground waste of reactive plastics of the same composition as the polymer matrix, filled with glass fibre, the pulp fraction having a particle size in the range from 0.005 to 0.1 mm.

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A. CLASSIFICATION OF SUBJECT MATTER

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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)

C08J 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 practicable, search terms used)

EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	CZ 37 113 U1 (D A T L COMPLET S R O [CZ]) 8 June 2023 (2023-06-08) cited in the application claim 3; examples 3,4	1,2
A	DIDIER PERRIN ET AL: "Treatment of SMC Composite Waste for Recycling as Reinforcing Fillers in Thermoplastics", MACROMOLECULAR SYMPOSIA, vol. 221, no. 1, 1 January 2005 (2005-01-01), pages 227-236, XP055194977, ISSN: 1022-1360, DOI: 10.1002/masy.200550323 page 229; figure 1	1-3

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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 :

"A" document defining the general state of the art which is not considered to be of particular relevance

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"&" document member of the same patent family

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1

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C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DE 39 22 740 A1 (BASF AG [DE]) 24 January 1991 (1991-01-24) column 1, lines 25-41 column 1, lines 45-48,61-64 column 2, lines 3-8,29-34	1-3
A	CN 1 554 493 A (UNIV TONGJI [CN]) 15 December 2004 (2004-12-15) claims 2-10; examples	1-3
A	DE 197 15 418 C2 (NADAY PETER [DE]) 18 June 2003 (2003-06-18) claims; example 1	1-3

1

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/CZ2024/050037

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
CZ 37113 U1	08-06-2023	NONE	
DE 3922740 A1	24-01-1991	DE 3922740 A1	24-01-1991
		EP 0407925 A2	16-01-1991
		ES 2059905 T3	16-11-1994
CN 1554493 A	15-12-2004	NONE	
DE 19715418 C2	18-06-2003	NONE	