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**(54) LACES AND GRANULES OF THERMOPLASTIC POLYMERS
AND THEIR PRODUCTION**

- (71) We, IMPERIAL CHEMICAL INDUSTRIES LIMITED, Imperial Chemical House, Millbank, London SW1P 3JF, a British Company, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it to be performed, to be particularly described in and by the following statement:—
- 10 This invention relates to laces and granules of thermoplastic polymers and their production. In particular it relates to the production of laces and granules reinforced with long fibre.
- 15 Many thermoplastic compositions are compounded by mixing various additives, e.g., stabilizers, fillers and pigments, with a thermoplastic polymer and extruding the mixture by means of a screw extruder. The
- 20 thermoplastic is heated to above its softening point as it passes through the screw and while in the screw the melt is mixed. After mixing the melt passes through a die so as to produce what is usually known as a
- 25 "lace". In most cases the lace is chopped into granules, usually after it has been cooled, e.g., by passing the lace through a cold water bath.
- 30 Laces may also be produced by extrusion through a die without the use of a screw feed. For example U.K. patent specification no. 1,334,702 describes a process for making fibre reinforced thermoplastic material. In this process a roving of the fibres, preferably
- 35 glass fibre, is passed through a bed of powdered thermoplastic material and while the roving is in the bed it is separated into bundles each of which consist of one or more filaments. The roving is allowed to
- 40 regain its original state and powder is trapped between the fibres so that the result is a roving which is impregnated with the powder. The impregnated roving is heated
- so as to melt the thermoplastic powder and while the powder is molten the roving is drawn through a die to produce a consolidated lace. 45
- It is sometimes desirable to make small changes in the composition of a lace, e.g., a change of pigment. In the past this has normally required thorough cleaning of the whole of the equipment used to produce the lace and it is an object of this invention to facilitate the making of small changes in the composition of the lace. 50
- According to the method of the invention the composition of a thermoplastic polymer lace is modified by exposing the lace while its surface is tacky to a powdered thermoplastics polymer composition whereby the powder adheres to the surface of the lace to form a granular coating. 55
- In a preferred form of the invention the granular coating is heated so that the coating melts and becomes more uniform. Most suitably the lace is passed through a second die in order to make the molten coating more uniform. 60
- The coating is conveniently applied to the lace by passing the lace above the surface of a bath of the powdered thermoplastics composition while a current of air is directed at the lace from below the surface of the bath. During its passage through the bed of powdered material the air picks up powder which impinges on the surface of the lace. 65
- Fibre reinforced laces in particular are highly resistant to bending and this technique is particularly desirable in that it allows the powder to be applied to the surface of the lace without bending the lace. 70
- The surface of the lace can be made tacky by applying a solvent or adhesive but it is preferred to make the surface tacky by heating at least the surface to or main- 75
- 80
- 85

taining at least the surface at a temperature at which the surface is tacky.

In most cases laces are hot during the manufacturing process and they are tacky if the powder is applied before the lace has time to cool; laces which contain long fibre have sufficient tensile strength to pass through a die.

If a non-reinforced lace is to pass through a die it is necessary to cool the lace because the hot lace has little or no tensile strength. Since the lace is cooled from the surface inwards it will usually be necessary to reheat only the surface in order to make it sufficiently tacky for the powder to stick.

The method described above is particularly suitable for making small changes in the composition of a lace. For example a colour masterbatch containing a very high proportion of pigment can be coated onto a lace. When the granules produced by cutting the coated lace are remelted to produce the final product the pigmented layer becomes uniformly dispersed in the whole composition so that a uniformly coloured article is produced. Thus the coated lace is equivalent to a uniformly coloured lace. However producing a uniformly coloured lace would necessitate passing the pigment through the whole of the lace forming process and when it is desired to change the colour it is necessary to clean the whole equipment very thoroughly. In the method according to the invention it is necessary to change only the coating equipment which represents a very small proportion of the total. Thus the invention facilitates the production of a series of different coloured compounds.

It will be appreciated that any additives, e.g., stabilizers, fillers with small particle size such as ballotini glass or talc and foaming agents, can be applied in the same manner as the colour masterbatch. In some cases it is undesirable or impossible to pass an additive through the main lace forming process, e.g., a foaming agent might decompose or a liquid additive might have an undesirable effect on the powder bath of the process described in U.K. patent specification no. 1,334,702. The process according to the invention enables said additives to be incorporated since the conditions during the coating stage can be adjusted to suit the additive.

It will also be appreciated that the polymer used in the coating need not be the same as that in the lace.

When laces reinforced with long fibres are chopped the cutter sometimes fails to sever fibres at or near the surface of the lace. When this happens the granules separate by pulling out the unsevered fibre to leave a "whisker" which protrudes from the end of the granules. It has been found

that the presence of an unfilled sheath reduces the number of granules which have whiskers.

Thus a method of making granules of fibre, preferably glass fibre, thermoplastic polymer preferably comprises:

(a) Impregnating a fibrous, preferably glass, roving with a powdered thermoplastic;
(b) Heating the impregnated roving from (a) to a temperature at which the thermoplastic melts;

(c) While the thermoplastic is molten drawing the roving through a die to produce a consolidated lace;

(d) Passing the lace from (c), while its temperature is still high enough for its surface to be tacky, above the surface of a bath of a powdered thermoplastics composition while a current of air is directed at the lace from below the surface of the bath whereby powder from the bath is caused to adhere to the surface of the lace;

(e) Heating the granular coating applied in (d) so that it melts;

(f) Passing the lace with the molten coating through a second die so that the coating becomes more uniform.

(g) Cooling the lace from (f) and cutting it into granules.

The polymer of the sheath is most suitably the same as the polymer used to form the lace. On the other hand there is the possibility of operating a dual purpose process in which coating are used as described above to modify the composition of the lace and the coating also serves to reduce the incidence of whiskers.

A coating of several layers may be applied by repeated applications of the method described above.

A lace produced as described above is usually chopped to give granules.

The invention includes laces and granules which have been prepared as described above.

Two methods of carrying out the process according to the invention will now be described by way of example with reference to the accompanying diagrammatic drawings in which:—

Figure 1 illustrates the method of applying coatings to a glass fibre reinforced lace, and

Figure 2 illustrates a modification of the process of Figure 1 which is applicable to laces which are not reinforced.

Figure 3 illustrates two granules made according to the invention and two other granules.

Figure 1 illustrates the application of a coating to a fibre reinforced lace 10 which has been produced by the method described in U.K. patent Specification no. 1,334,702. The method of producing the lace is not

part of this invention and it is not illustrated in Figure 1.

The lace 10 passes through a die 11 which is the last step in the process of the preparation of the lace. Before it has time to cool and while the surface is still tacky the lace 10 passes over a tray 12 which contains a fluidised bed of powdered polymer 13. A nozzle 14 having orifice 15 situated in the bed and a blast of air from the nozzle produces a curtain which contains the powdered polymer. As the lace passes through this curtain the particles adhere to its tacky surface so that the surface becomes coated with the particles. The particles which do not contact the lace fall back into the tray 12 so that they are recirculated. The tray may be shielded to prevent scattering of the polymer but the shield is not shown in any drawings.

After passing over the tray 12 the lace enters a hot tube 16. The temperature of the tube is such that the coating (but not the body of the lace) melts and the molten coating passes through a second die 17 so that a uniform sheath is produced. After leaving the second die 17 the lace is cooled and chopped in conventional manner to produce granules.

Figure 2 shows a modification of the process for use with laces which are not reinforced with fibre. The differences are that the lace 10 is produced by a conventional screw extruder process and on leaving the die 11 of the extruder the lace passes through a water bath 18 which cools it so that it has sufficient tensile strength to withstand the rest of the process. On leaving the water bath the lace 10 passes through a small heater 19 which dries and heats the surface until it is tacky. On leaving the heater 19 the lace is coated with powder as described with reference to Figure 1. When the lace passes through the hot tube 16 it is necessary to heat it sufficiently to melt the adherent powder but care must be taken to avoid heating it so much that it loses its tensile strength.

The application of three coatings to glass fibre reinforced laces as described in Figure 1 will now be described. In each case the original lace was produced by passing glass roving at 5m per minute through a bed of powdered polypropylene. During its passage through the bed the roving was spread out and allowed to reform as an impregnated roving. The impregnated roving was passed through a hot tube to melt the polypropylene powder and then through a die to consolidate the roving into a lace. On leaving the die the surface temperature of the lace was approximately 230°C.

The lace was chopped (using a conventional chopper) into granules 1 cm long some of which had elongated whiskers

protruding from one end. The proportion of such granules varied between about 25% and about 35%.

EXAMPLE 1

A glass reinforced polypropylene lace, containing 23.3% by weight of continuous glass filaments running parallel to the lace axis and having a surface temperature of approximately 230°C, was pulled at a rate of 5m per minute through an atmosphere of air and polypropylene powder. The powder adhered to the hot lace surface and the granular coating so formed was melted and consolidated by its passage through a 12cm length of heated tube maintained at an average temperature of 250°C and fitted with a die. The resultant lace was chopped into 1cm long granules and it was found that the glass fibre concentration of the coated lace had been reduced to 21.2% by weight. The number of granules having elongated glass fibres protruding from their ends was only 3%, resulting in a much improved flow and making feeding of the granules into injection moulding machines easier by eliminating bridging.

EXAMPLE 2

Production of a Pigmented Lace

A glass fibre reinforced lace as described in Example 1 was pulled at a rate of 5m per minute through an atmosphere of air and polyethylene powder, the polyethylene powder containing 5% by weight of carbon black pigment. The granular covering of powder and pigment which adhered to the lace surface was melted and consolidated by its passage through a 12cm length of heated tube, maintained at an average temperature of 250°C and fitted with a die. The resulting lace had a glass fibre concentration of 21.2% by weight and on chopping it was found that the number of chips having glass fibres protruding from their ends had been reduced to 2%. When the material was injection moulded the specimen obtained had an overall pigment concentration of 0.6% by weight based on the total weight of polymer, and showed excellent colour dispersion.

EXAMPLE 3

Incorporation of Extra Additives into the Surface Coating

A glass fibre reinforced lace as described in Example 1, but having a glass fibre concentration of 27.0% by weight, was pulled at a rate of 5m per minute through an atmosphere of air and polypropylene powder, the polypropylene powder containing 5% by weight of a liquid heat stabilising agent. The granular covering adhering to the lace surface was melted and consolidated by its passage through a 12cm length of heated tube, maintained at an average

temperature of 250°C and fitted with a die. The resulting lace had a glass fibre concentration of 25.0% by weight and an overall heat stabilizer concentration of 0.5% by weight based on the total polymer weight. When the lace was chopped the number of granules having long glass fibres protruding from their ends had been reduced to 2%. Subsequent injection moulding of these granules yielded a specimen which showed a significantly increased resistance to high temperature deformation.

EXAMPLE 4

15 *Production of lace incorporating a foaming agent*

A glass fibre reinforced lace as described in Example 1 was pulled at the rate of 5m per minute through an atmosphere of air and low density polyethylene powder, the polyethylene powder containing 4% by weight of a blowing agent (azo-dicarbonamide). The granular covering of powder and blowing agent which adhered to the lace surface was consolidated by its passage through a 12cm length of heated tube, maintained at an average temperature of 160°C and fitted with a die. The resulting lace had a glass fibre concentration of 20.9% by weight and was chopped to form a granule suitable for moulding. About 3% of the granules had long fibres protruding from the cut ends.

These granules were fed to a 350 tons Bone-Craven injection moulding machine, fitted with a tray shaped mould, the cavity of which was 35cm x 25cm x 5cm deep with a wall thickness of 6mm. The shot size was adjusted so that a moulding of a final density of 0.76g/cm³ was achieved, by virtue of the foaming action of the decomposing blowing agent.

An important effect of the coating will now be described with reference to Figure 3.

A satisfactory granule without a coating (i.e., a granule not according to the invention) is shown in Figure 3a. The granule has a wall 20a (which in most cases is cylindrical but can be any shape which can be applied by pulling through a die) and a flat face 22a which was formed by the chopper. The granule contains fibres which are parallel to one another and which extend the whole length of the granule. There may be (at the most) a small tuft 23a of fibre which stands proud of the surface 22a. Figure 3b shows an unsatisfactory granule. Instead of, at the most, a small tuft there is a whisker 23b of fibre which protrudes from the flat face 22b; the whisker is about as long as the rest of the granule.

A satisfactory granule according to the invention is shown in Figure 3c. The granule has a coating 24c covering the surface 20c. As can be seen from the drawing the com-

position of the coating differs from the composition of the bulk of the granule. The bulk contains the fibres 21c whereas the coating does not. (Example 1 illustrates the case where this is the only difference. Examples 2, 3 and 4 show that there can also be other differences.) The coated granules may have, at the most, a small tuft 23c of fibre standing proud of the flat face 22c; granules having the elongated whiskers as shown at 23d are unsatisfactory because the elongated whiskers (but not the short tufts) have an adverse effect upon the bulk properties, e.g., feeding from a hopper may be difficult. It has been found that compositions which contain at least 90% by weight, preferably 95% by weight, of granules as shown in Figure 3c give satisfactory bulk properties even though the remainder of the granules of the composition have elongated whiskers. (Note: For the purposes of comparison and Claim 19, granules without any tuft are deemed to be as shown in Figure 3c).

Examples 1 to 4 show the production of coated granules with 2% to 3% having whiskers; in the comparative example there were 25% to 35% of such granules. Clearly the application of the coating substantially improves the bulk properties.

It is emphasised that where tufts of whiskers occur they protrude through the cut face 22 and no coating is applied to this face. It is surprising that the application of a coating to the surface 20 has a beneficial effect upon the whiskers which protrude through an uncoated surface.

Glass fibres are the most commonly used but the reduction of whiskering according to the invention can be applied to other types of fibre, e.g., carbon fibre.

WHAT WE CLAIM IS:—

1. A method of modifying the composition of a thermoplastic polymer lace which comprises exposing a lace the surface of which is tacky to a powdered thermoplastic polymer composition whereby the powder adheres to the surface of the lace to form a granular coating.

2. A method according to Claim 1, in which the lace is exposed to the powdered thermoplastics composition by passing it above the surface of a bath of the powdered thermoplastics composition while a current of air is directed at the lace from below the surface of the bath.

3. A method according to either Claim 1 or Claim 2 in which at least the surface of the lace is at a temperature at which the surface is tacky while it is exposed to the powdered thermoplastic.

4. A method of modifying the composition of a thermoplastics lace, which comprises carrying out a method as defined in any one of Claims 1 to 3 and thereafter

temperature of 250°C and fitted with a die. The resulting lace had a glass fibre concentration of 25.0% by weight and an overall heat stabilizer concentration of 0.5% by weight based on the total polymer weight. When the lace was chopped the number of granules having long glass fibres protruding from their ends had been reduced to 2%. Subsequent injection moulding of these granules yielded a specimen which showed a significantly increased resistance to high temperature deformation.

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These granules were fed to a 350 tons Bone-Craven injection moulding machine, fitted with a tray shaped mould, the cavity of which was 35cm x 25cm x 5cm deep with a wall thickness of 6mm. The shot size was adjusted so that a moulding of a final density of 0.76g/cm³ was achieved, by virtue of the foaming action of the decomposing blowing agent.

An important effect of the coating will now be described with reference to Figure 3.

A satisfactory granule without a coating (i.e., a granule not according to the invention) is shown in Figure 3a. The granule has a wall 20a (which in most cases is cylindrical but can be any shape which can be applied by pulling through a die) and a flat face 22a which was formed by the chopper. The granule contains fibres which are parallel to one another and which extend the whole length of the granule. There may be (at the most) a small tuft 23a of fibre which stands proud of the surface 22a. Figure 3b shows an unsatisfactory granule. Instead of, at the most, a small tuft there is a whisker 23b of fibre which protrudes from the flat face 22b; the whisker is about as long as the rest of the granule.

A satisfactory granule according to the invention is shown in Figure 3c. The granule has a coating 24c covering the surface 20c. As can be seen from the drawing the com-

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Glass fibres are the most commonly used but the reduction of whiskering according to the invention can be applied to other types of fibre, e.g., carbon fibre.

WHAT WE CLAIM IS:—

1. A method of modifying the composition of a thermoplastic polymer lace which comprises exposing a lace the surface of which is tacky to a powdered thermoplastic polymer composition whereby the powder adheres to the surface of the lace to form a granular coating.

2. A method according to Claim 1, in which the lace is exposed to the powdered thermoplastics composition by passing it above the surface of a bath of the powdered thermoplastics composition while a current of air is directed at the lace from below the surface of the bath.

3. A method according to either Claim 1 or Claim 2 in which at least the surface of the lace is at a temperature at which the surface is tacky while it is exposed to the powdered thermoplastic.

4. A method of modifying the composition of a thermoplastics lace, which comprises carrying out a method as defined in any one of Claims 1 to 3 and thereafter



heating the granular coating so that it melts.

5. A method of modifying the composition of a thermoplastics lace, which comprises carrying out a method as defined in Claim 4, and thereafter passing the lace with the molten coating through a die so that the coating becomes more uniform.

6. A method of modifying the composition of a lace according to any one of Claims 1 to 5 in which the thermoplastics lace is fibre reinforced.

7. A method of modifying the composition of a lace as hereinbefore described in any one of Examples 1, 2, 3 or 4.

8. A method of producing granules which comprises modifying the composition of a lace by a process according to any one of Claims 1 to 7 and thereafter cutting the lace into granules.

9. A method of making granules of a fibre filled theroplastics polymer, which method comprises:

(a) Impregnating a fibrous roving with a powdered thermoplastic;

(b) Heating the impregnated roving from (a) to a temperature at which the thermoplastic melts;

(c) While the thermoplastic is molten drawing the roving through a die to produce a consolidated lace;

(d) Passing the lace from (c), while its

temperature is still high enough for its surface to be tacky, above the surface of a bath of powdered thermoplastics composition while a current of air is directed at the lace from below the surface of the bath whereby powder from the bath is caused to adhere to the surface of the lace;

(c) Heating the granular coating applied in (d) so that it melts;

(f) Passing the lace with the molten coating through a second die so that the coating becomes more uniform;

(g) Cooling the lace from (f) and cutting it into granules.

10. A process according to Claim 9, in which the roving is a glass fibre roving.

11. A lace which has been produced by a method according to any one of Claims 1 to 7.

12. Granules which have been produced by a method according to any one of Claims 8 to 10.

13. A granular composition containing granules according to Claim 12 in which at least 90% by weight of the granules, based on the total composition, have only small tufts of fibres protruding from the granules substantially as illustrated in Figure 3c or are tuftless.

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