

PATENT SPECIFICATION

DRAWINGS ATTACHED

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COMPLETE SPECIFICATION

Rigidified Polythene Structures and Method of Producing them

We, VAESSEN-SCHOEMAKER HOLDING N.V., a Dutch limited liability company, organised and existing under the laws of the Netherlands, of Deventer, Province of Overijsel, the Netherlands, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to moulded polythene structures having valuable combinations of physical and chemical properties, and which can be produced economically in the configurations required to constitute the wall structures of containers, boats, wash basins, panel and other articles. The invention also relates to a method of producing such structures.

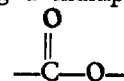
Polythene is well known to be a synthetic thermoplastic material having excellent properties of chemical inertness and water resistance and a very low modulus of elasticity (Young's modulus) or low rigidity.

The chemical inertness of polythene, which is attributable to its marked non-polar character, makes layers of this material excellently suited for use as the wall structure or lining of containers and other articles to be employed in contact with substances that attack other plastics or metals or both.

On the other hand, the low modulus of polythene severely limits the rigidity and thus the size, economy or serviceability of containers and other articles made of the material. As the size of such containers is increased it becomes necessary either to make them with very thick walls or to support them in rigid frames which themselves are not sufficiently resistant to attack by chemicals. In either case the complexities and costs involved cause otherwise desirable articles that

might be made of the material to be commercially unattractive.

It is also well known that polyester resins possess a relatively high modulus of elasticity, or rigidity, but have a much lower resistance to chemicals than that of polythene. Polyester resins in general are macromolecular substances carrying a multiplicity of



groups in the chain. Typical examples are copolymers of dicarboxylic acid polyesters of polyhydric alcohols and vinyl monomers. They often are compounded with reinforcing materials, especially glass fibre, but even then they have a lower resistance to chemicals than polythene. Other materials which may be incorporated in them include vegetable fibres such as cotton or shredded fabrics, mineral fibres such as asbestos, and finely divided fillers or pigments.

The relatively low chemical resistance of polyesters resins is related to their polarity. Due to their relatively high polarity, it has not been possible hitherto to cause coatings or layers of this material to adhere directly to layers of polythene by known coating or moulding techniques without resort to costly treatment of the polythene layer, such, for example, as a flame treatment to oxidize its outer surface. Another possible way of incorporating the two materials into a single laminated structure has required the bonding of a layer of jute or asbestos to a moulded wall of polythene followed by the application of a layer of glass fibre reinforced polyester to the jute or asbestos layer; but this, too, involves objectionably complex and costly manufacturing procedures.

According to the present invention, it has been discovered that a strong interfacial co-

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hesion between polythene and polyester layers can be obtained, notwithstanding the widely different chemical and physical properties of the two materials, by forming a foamed layer of polythene and applying a coating or layer of a polyester resin to and setting it upon the foamed polythene layer.

By the use of this discovery, reinforced polythene structures can be readily produced in the form of containers, boat hulls, wash basins, panels, and various other desired articles, with the wall-like body of each article comprising a layer of a reinforced polyester resin set in direct adhesion to a foamed layer of polythene. Where properties of the polyester resin are desired on both sides of the articles, polyester layers may be formed over both sides of the foamed polythene layer. More advantageously, a layer of reinforced polyester resin is formed and set in direct adhesion to one side of a foamed layer of polythene having a layer of unfoamed polythene in direct adhesion to its other side, so that the articles obtained will have a sandwich-like laminar wall structure possessing great strength together with the valuable properties of polythene.

Articles made with such a laminar wall structure can be produced economically in any of a great variety of configurations and sizes. The cohesion between the resin layers is so strong that they do not become separated even when the articles are subjected to severe deflection tests, as by squeezing them in a vice. Although the polyester layer may be cracked or broken in such tests, the polythene remains strongly joined to it everywhere.

Moreover, it has been discovered that this laminar wall structure, because of its sandwich construction together with its other properties, has even greater rigidity than the same thickness of the reinforced polyester alone. It is therefore extremely well suited for the manufacture of large hollow articles such as boat hulls or large size containers for chemicals.

The production of articles according to the invention can be carried out by the use of various combinations of moulding operations, or of moulding and coating operations, suited to the properties of the materials to be formed into the adhering layers. When large size containers or other large hollow articles are to be produced it is advantageous to make them by sintering a mixture of a finely divided polythene and a blowing agent in an externally heated thin-walled mould, so as to form against the mould wall a foamed layer of polythene having the general configuration of the desired wall structure of the article, and thereafter spraying a liquid glass fibre polyester resin composition onto the foamed layer so as to form thereover a

polyester layer that will set to a hardened state in adhesion to the foamed polythene.

The mould and associated apparatus used to form the foamed layer may also be used to form a sintered layer of unfoamed polythene in adhesion to one side of the foamed layer, before the application of the polyester resin composition to its other side. For example, a charge of a finely divided polythene can be sintered in the same mould to form an unfoamed layer therein either immediately before or immediately after the sintering operation by which the foamed layer is formed, so that the structure removed from the mould will be composed of a foamed layer of polythene united with an unfoamed polythene layer covering either its outer or its inner side, whichever may be desired.

While such a laminar structure of polythene layers alone lacks the rigidity and serviceability that can be attained according to the invention, it acquires these qualities upon the application and setting of a layer of reinforced polyester resin over the exposed side of the foamed layer of polythene as herein set forth. By moulding the polythene layers to the configuration of a container or other hollow article with the unfoamed layer of polythene at the inner side thereof and forming the polyester layer over the outer side thereof, a self-supporting container can be obtained which will exhibit the full chemical resistance of polythene to its contents and yet will have a rigidity and usefulness not attainable at comparable cost in any other way.

The polyester resin composition used according to the invention may be any of those which are commercially available or known from the literature relating to self-hardening polyester coating compositions. It is often advantageous to use a polyester liquid compounded with glass fibres to reinforce the polyester layer formed on the foamed polythene layer. Various other additions, including other reinforcing materials, hardeners, polymerization accelerators or catalysts, and filler or pigments of various kinds, may also be employed in the liquid polyester composition, as will readily be understood by persons skilled in the art. Among suitable commercial compositions are the polyester resins used in boat-building or for coating the hulls of boats.

The foamed polythene layer may be formed by the use of various mixtures of polythenes and blowing agents in various moulding techniques. Methods suitable for producing it by the sinter-moulding of powder mixtures, and also for joining it with an unfoamed sintered layer of polythene are disclosed more fully in Patent Specification No. 46328/65 (Serial No. 1,115,639).

The following Examples illustrate the in-

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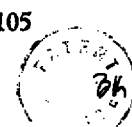
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vention, with reference to the accompanying diagrammatic drawings, in which:

Fig. 1, is a sectional elevation of a moulding apparatus for use in the method of the invention,

Fig. 2, is a fragmentary sectional view of a moulded polythene structure formed using such a moulding apparatus,

Fig. 3 is an elevation illustrating a method of applying polyester resin to the structure of Fig. 2,

Fig. 4 is a fragmented perspective view of a completed container formed in accordance with the invention,

Fig. 5 is a fragmentary sectional view of the container shown in Fig. 4,

Fig. 6 is a sectional view of another mould for use in the method of the invention, and

Fig. 7 is a similar view of the corresponding moulded article produced in accordance with the invention.

EXAMPLE 1

A rectangular sheet metal mould having a length of 100 cm. and a cross section of 50 cm. x 50 cm. was filled to about 3/4 of its volume with a mixture of a finely divided polythene (density 0.918, melting index 2) and 2% by weight of a powdered blowing agent known as Porofoor T.R. (Registered Trade Mark). This is a nitrogen-liberating blowing agent having a density of 1.5, a stated blowing (decomposition) temperature of 115°C., and a theoretical gas generation of 130 ml. per gram.

The mould so filled was heated for 3 minutes in an oven having a temperature of 325°C., while being rotated slowly about its longitudinal axis (at about 2 r.p.m.) to distribute the moulding powder continually over the entire inner surface of the mould.

In the course of this heating, a foamed sintered layer of polythene was formed to a thickness of about 2 mm. upon the heated inner surface of the mould.

Then the mould was removed from the oven and emptied of unsintered powder by removing a heat-insulating closure from one end of the mould and pouring out the surplus powder mixture.

The mould was then filled again to about 3/4 of its volume with a finely divided polythene of the nature above noted, without a blowing agent, and returned to the oven for a further heating period of 6 minutes at 325°C.

At the end of that period, the mould was removed from the oven, again emptied of unsintered powder, and then returned to the oven for 4 minutes to smooth the inner surface of the polythene structure contained in it. Then it was removed from the oven and cooled, and the moulded polythene structure was removed from the mould.

The structure thus formed was a hollow

container having the configuration of the mould and composed of a foamed polythene layer approximately 2 mm thick united over its inner side with a dense or non-foamed layer of polythene approximately 2.5 mm thick. This structure was so flexible and lacking in rigidity that it would not support itself in an upright position.

Then a mixture of 40 parts by weight of glass fibre with 60 parts of a self-polymerizable polyester liquid was sprayed over the outer side of the moulded polythene structure by means of a spray gun, until a polyester layer 5 mm thick had been formed in adhesion to the foamed layer of polythene. The polyester used was the type K 572 boat-building resin produced by the firm Synress, Hook of Holland. The glass fibre was a product of the A.K.U. firm of Arnheim, Holland.

After the hardening of the polyester layer, a rigid self-supporting container, excellently suited for the storage and transportation of chemicals was obtained.

Referring to Figs. 1—5 of the drawings, Fig. 1 indicates a sheet metal mould 10 largely filled with finely divided polythene or with a mixture of finely divided polythene and a blowing agent as above described. The mould has a heat-insulating closure 11 fitting into an opening in one end wall. It is mounted between clamps 12 and 13 for rotation with a shaft 14 in a frame 15 with which the mould is movable into and out of an oven 18.

Fig. 2 is an enlarged diagrammatic sectional view of a portion of the moulded laminar structure removed from the mould. A foamed polythene layer 1 forms the outer side of this structure and is lined by the dense polythene layer 2 sintered integrally over its inner side.

Fig. 3 indicates a way of supporting the moulded polythene structure on a slowly rotating hanger 20 while a liquid polyester composition is sprayed from a gun 21 over the outer side of the foamed polythene layer 1.

Fig. 4 is a perspective view, partly broken away, of the laminar container C finally obtained. The wall structure of this container, as seen in the fragmentary sectional view constituting Fig. 5, consists of the foamed polythene layer 1 integrally joined over its entire inner side with the smooth dense polythene layer 2, and joined over its entire outer side with a hardened glass fibre reinforced polyester layer 3.

EXAMPLE 2

The process of Example 1 was carried out with variations of the concentration of the blowing agent (Porofoor T.R.) (Registered Trade Mark) in the mixture sintered to form the foamed layer. It was found that when as little as 0.1% of the blowing agent was used

the polyester would no longer adhere to the foamed polythene layer, while when as much as 10% of the blowing agent was used the foamed layer would no longer adhere to the wall of the mould and became deformed.

The most effective of these mixtures were found to be those containing about 1 to 5% of the blowing agent, although other concentrations in the range from 0.25% to 10% gave foamed layers to which the polyester layer would adhere.

EXAMPLE 3

The method of the invention substantially according to Example 1 was carried out with the use of various grades of finely divided polythene for the formation of the foamed layer and also for the formation of the dense polythene layer constituting the inner surface of the container. Among the grades successfully used were polythenes having melting indices of 1, 7 and 20 and densities of 0.918, 0.940 and 0.960. Those having the lower melting indices are generally to be preferred for the manufacture of containers for use in chemical industries.

EXAMPLE 4

A wash-basin of sheet metal, having generally the internal configuration shown at 30 in Fig. 6 of the drawings, was used as the mould in this example.

The mould was charged with a weighed quantity of powdered polythene (melting index 7, density 0.918) and was covered by a heat-insulating asbestos plate as indicated at 31 in Fig. 6.

Then the mould was clamped in a mechanism of known character which rotated it about two axes simultaneously so as to distribute the powder continually over its entire inner surface; and while being so rotated it was heated for 12 minutes in an oven at 325°C.

The mould was then removed from the oven, opened, and charged with a weighed quantity of a mixture of powdered polythene (density 0.918, melting index 2) and 3% by weight of Porofoor T.R.; whereupon, it was reclosed and rotated and heated in the oven as before, for a further period of 10 minutes.

Upon removing the mould from the oven, cooling it and removing its contents, a laminar polythene structure was obtained which, when inverted, had generally the configuration of the original wash-basin, as indicated in Fig. 7 of the drawings. This structure was composed of a centrally depressed top layer 32 of densely sintered polythene united with an underlying layer 33 of foamed polythene.

The under side of the foamed layer was then spray-coated with a liquid glass fibre polyester composition of the nature described in Example 1, to form a polyester layer indicated at 34 in Fig. 7. After the hardening

of the polyester layer it was finished with a gel coating.

The article so obtained was a very rigid wash-basin having a smooth water-impervious top structure of polythene. It was capable of being connected to a wall by means of consoles fitted into the recesses A and B (Fig. 7) beneath its side panels. The layer thicknesses as shown in Fig. 7 are greatly exaggerated in relation to the size of the article, the actual thicknesses being, for example, about 2.5 mm for layer 32, about 3 mm for layer 33 and about 2.5 mm for layer 34.

EXAMPLE 5

A sheet metal mould was made in the form of a miniature boat having a length of 60 cm and a beam of 10 cm.

A mixture of 200 g of polythene powder (density 0.950, melting index 3) and 4% by weight of a blowing agent known as Porofoor S 44 (Registered Trade Mark) was placed in the mould, and the mould was closed by an asbestos cover confining the powder to its inner surface. Porofoor S 44 (Registered Trade Mark) has a density of 1.6, a blowing (decomposition) temperature of about 175°C, and a theoretical gas generation of about 120 ml per gram.

The mould was then rotated about its longitudinal axis while being rocked on a cross axis and heated at 300°C, for 4 minutes.

Then it was opened and 1,000 g of polythene powder (density 0.940, melting index 6) was placed in it; whereupon it was reclosed, rotated and rocked as before, and heated at 300°C for 14 minutes.

Then the mould was cooled, the moulded polythene structure was removed from it, and a layer of glass fibre polyester resin was applied to the outside of that structure and set thereon in cohesion with the foamed polythene layer. After the hardening of the polyester, a gel coating about 0.3 mm thick was applied to it.

The reinforced moulded boat which resulted was extremely strong and durable. Its laminar wall structure consisted of an inner layer of densely sintered polythene about 2 mm thick, a middle layer of foamed polythene about 2 mm thick, and an outer layer of polyester resin about 2 mm thick.

According to another important feature of the present invention, it has been found practicable to make containers and other articles formed of the new laminar wall structure which have heating elements such as pipes or heating strips embedded in their wall structure. These articles will serve for heating liquids or other substances held in contact with their wall structure, thus eliminating the necessity for separate heating elements. A wall structure suitable for this purpose preferably comprises a thin densely sintered layer of polythene, only about 1 to 2 mm thick, at

its inner side, a foamed polythene layer approximately 1 mm thick in cohesion with the outer side of the dense layer, then the heating elements in contact with the foamed layer, and finally the polyester layer which is applied and set over the heating elements and the foamed layer in strong adherence to them.

- 5 It is also practicable to embed iron, steel or other metal reinforcing strips in the laminar wall structure in the same manner as the heating elements mentioned above. By making structures according to the invention with such reinforcements embedded in them, the invention can be used for the manufacture of very large rigid polythene containers having, for example, capacities of 1,000 litres or more.

WHAT WE CLAIM IS:—

- 20 1. A reinforced polythene structure comprising a layer of a polyester resin adhering directly to a foamed layer of polythene.
- 25 2. A structure as claimed in claim 1, including a layer of unfoamed polythene adhering directly to the layer of foamed polythene on the opposite surface to that bearing the polyester resin.
- 30 3. A structure as claimed in claim 1 or 2, in which the polyester resin is reinforced with glass fibre.
- 35 4. A structure as claimed in any one of claims 1—3, in which the layer of foamed polythene is a foamed sintered layer and the layer of polyester resin is sprayed on.
- 40 5. A structure as claimed in claim 2 or claim 3 or 4 as appended ~~therein~~ *thereto*, in which the layer of unfoamed polythene is a sintered layer.
- 45 6. A structure as claimed in claim 5, being a self-supporting container, the sintered layer of unfoamed polythene providing a chemically resistant inner surface, and the polyester resin layer providing a reinforcing outer layer of greater rigidity than the polythene layer.
7. A reinforced polythene structure substantially as described herein with reference to Fig. 4, Fig. 5 or Fig. 7 of the accompanying drawings.

8. A method of making a structure as claimed in claim 1, wherein a foamed layer of polythene is formed having substantially the configuration of the desired structure, and a layer of polyester resin is applied to a surface of the foamed layer and caused to set in place thereon.

9. A method as claimed in claim 8, wherein the polyester resin is reinforced with glass fibre.

10. A method as claimed in claim 8 or 9, wherein the polyester resin is sprayed onto the foamed layer, in liquid form.

11. A method as claimed in any one of claims 8—10, wherein the foamed polythene layer is initially formed upon an unfoamed polythene layer of the desired configuration and caused to set in place thereon, the polyester resin layer then being applied to the free surface of the foamed layer.

12. A method as claimed in any one of claims 8—11, wherein the foamed polythene layer is formed by sintering a mixture of finely divided polythene and a blowing agent.

13. A method as claimed in claims 11 and 12, wherein the unfoamed polythene layer is formed by sintering finely divided particles of polythene.

14. A method as claimed in claim 12 as appended to any one of claims 8—10, wherein the foamed polythene layer is the first layer of the structure to be formed, an unfoamed layer of polythene then being formed upon one surface of said foamed layer by sintering finely divided particles of polythene into a covering layer adhering to the foamed layer, and the layer of polyester resin being applied to the other surface.

15. A method of making a rigidified polythene structure substantially as described in any one of Examples 1—5 herein.

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its inner side, a foamed polythene layer approximately 1 mm thick in cohesion with the outer side of the dense layer, then the heating elements in contact with the foamed layer, and finally the polyester layer which is applied and set over the heating elements and the foamed layer in strong adherence to them.

It is also practicable to embed iron, steel or other metal reinforcing strips in the laminar wall structure in the same manner as the heating elements mentioned above. By making structures according to the invention with such reinforcements embedded in them, the invention can be used for the manufacture of very large rigid polythene containers having, for example, capacities of 1,000 litres or more.

WHAT WE CLAIM IS:—

1. A reinforced polythene structure comprising a layer of a polyester resin adhering directly to a foamed layer of polythene.

2. A structure as claimed in claim 1, including a layer of unfoamed polythene adhering directly to the layer of foamed polythene on the opposite surface to that bearing the polyester resin.

3. A structure as claimed in claim 1 or 2, in which the polyester resin is reinforced with glass fibre.

4. A structure as claimed in any one of claims 1—3, in which the layer of foamed polythene is a foamed sintered layer and the layer of polyester resin is sprayed on.

5. A structure as claimed in claim 2 or claim 3 or 4 as appended ~~thereto~~, in which the layer of unfoamed polythene is a sintered layer.

6. A structure as claimed in claim 5, being a self-supporting container, the sintered layer of unfoamed polythene providing a chemically resistant inner surface, and the polyester resin layer providing a reinforcing outer layer of greater rigidity than the polythene layer.

7. A reinforced polythene structure substantially as described herein with reference to Fig. 4, Fig. 5 or Fig. 7 of the accompanying drawings.

8. A method of making a structure as claimed in claim 1, wherein a foamed layer of polythene is formed having substantially the configuration of the desired structure, and a layer of polyester resin is applied to a surface of the foamed layer and caused to set in place thereon.

9. A method as claimed in claim 8, wherein the polyester resin is reinforced with glass fibre.

10. A method as claimed in claim 8 or 9, wherein the polyester resin is sprayed onto the foamed layer, in liquid form.

11. A method as claimed in any one of claims 8—10, wherein the foamed polythene layer is initially formed upon an unfoamed polythene layer of the desired configuration and caused to set in place thereon, the polyester resin layer then being applied to the free surface of the foamed layer.

12. A method as claimed in any one of claims 8—11, wherein the foamed polythene layer is formed by sintering a mixture of finely divided polythene and a blowing agent.

13. A method as claimed in claims 11 and 12, wherein the unfoamed polythene layer is formed by sintering finely divided particles of polythene.

14. A method as claimed in claim 12 as appended to any one of claims 8—10, wherein the foamed polythene layer is the first layer of the structure to be formed, an unfoamed layer of polythene then being formed upon one surface of said foamed layer by sintering finely divided particles of polythene into a covering layer adhering to the foamed layer, and the layer of polyester resin being applied to the other surface.

15. A method of making a rigidified polythene structure substantially as described in any one of Examples 1—5 herein.

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