

PATENT SPECIFICATION

NO DRAWINGS

Inventors: PAUL JOHNSON GARNER and DAVID FAIRCLOUGH OXLEY

L156,217

L156,217



Date of filing Complete Specification (under Section 3 (3) of the Patents Act 1949): 19 April, 1968.

Two Applications Date: 3 May, 1967. Nos. 20590/67 & 20591/67.

Two Applications Date: 15 Dec., 1967. Nos. 57082/67 & 57083/67.

Complete Specification Published: 25 June, 1969.

© Crown Copyright 1969.

Index at acceptance: —B5 A(1R14B, 1R14C1C, 1R14C1X, 3DX)

Int. Cl: —B 29 d 27/00, B 29 g 3/00

COMPLETE SPECIFICATION

Process for the Production of Cellular Articles

We, IMPERIAL CHEMICAL INDUSTRIES LIMITED, of Imperial Chemical House, Millbank, London, S.W.1., 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 is to be performed, to be particularly described in and by the following statement:—

The present invention relates to a process for the production of injection moulded articles having a foamed core and an unfoamed surface skin and to the articles produced thereby.

It had been proposed that such articles may be made by injecting a thermoplastic synthetic resin containing a blowing agent into a mould cavity, retaining the composition within the mould for long enough to allow any desired setting of the resin adjacent to the walls to take place and then allowing the mould to open by a predetermined amount to allow the core of the resin to foam. The mould surfaces may, if desired, be cooled to enhance the setting of the resin adjacent to the walls. The expansion of the mould cavity may be instantaneous or may be at a controlled rate and may be effected by the pressure generated by the decomposition of the blowing agent and the relaxation of external pressures on the mould and/or by positive opening of the mould.

A disadvantage of this process, especially in the manufacture of large mouldings, is that due to the presence of the blowing agent in the surface material sink marks and voids tend to appear in the surface skin; furthermore, the gases generated by the decomposition of the blowing agent tend to prevent perfect contact between the thermoplastic material and the mould surface which also leads to surface imperfections in the

mouldings. It is an object of the present invention to provide a process for the production of articles having a foamed core and an unfoamed surface skin in which the tendency for sink marks or voids to occur in the surface skin is reduced.

According to the present invention we provide a process for the production of articles having a cellular core and an unfoamed surface skin comprising injecting a predetermined amount of an unfoamable thermoplastic resin into a mould cavity and subsequently, and before the central portion of said unfoamable thermoplastic resin has set, injecting a thermoplastic resin composition containing a blowing agent at a temperature at or above the activation temperature of the blowing agent into the charge of the unfoamable thermoplastic resin that is in the mould cavity, and maintaining the composition within the mould for sufficient time to allow or cause the foamable composition to foam and the thermoplastic materials to set.

The walls of the mould may, if desired, be cooled to enhance the setting of the thermoplastic material adjacent thereto. The moulds used in the process of the present invention may have a mould cavity of constant volume or the cavity may be enlargeable to allow the foamable composition to expand. When a constant volume mould cavity is used the amounts of the unfoamable thermoplastic material and the thermoplastic resin containing a blowing agent that are injected into the mould must be controlled to allow the required amount of foaming to produce the desired foam density. The amounts of material injected will of course also depend upon the size of the mould cavity.

Where a mould having an enlargeable cavity is used a predetermined amount of ex-

Price 5s. 0d.
[Price 4s. 6d.]

5 pansion of the mould cavity is preferably
 allowed to take place after the foamable
 composition has been injected into the mould,
 so as to allow the foamable composition to
 expand. This controlled timing of the ex-
 10 pansion allows the formation of particularly
 uniform foams. If desired, this expansion
 may be delayed to allow some of the foamable
 thermoplastic material adjacent to the un-
 foamable composition to set and so become
 15 part of the surface skin of the mouldings.
 The mould cavity may be enlarged by in-
 stantaneously relieving the pressure holding
 opposed surfaces of the mould together and
 allowing the gas pressure generated by the
 20 activation of the blowing agent to open the
 mould, thus allowing the foamable composi-
 tion to expand to fill the larger mould cavity,
 alternatively the pressure holding opposed
 surfaces of the mould together may be re-
 duced slowly thus allowing a controlled ex-
 pansion of the mould. The expansion of the
 25 mould may be effected entirely by the pres-
 sure of the gases generated by the blowing
 agent or external forces may be applied to
 help enlarge the mould cavity.

We have found that by first injecting an
 unfoamable thermoplastic resin into the
 30 mould cavity the tendency for voids or sink
 marks to be formed in the surface of the
 article is reduced. Furthermore, if the foam-
 able and the unfoamable materials are ob-
 tained from separate sources, the process of
 the present invention has the added advan-
 35 tage that if any pigments, fillers or other
 additives which improve the surface finish
 of the article are to be included in the mould-
 ing they need only be included in the un-
 foamable resin. For example, additives which
 40 improve the resistance of the article to
 moisture or light, such as ultra-violet light
 stabilisers need only be included in the un-
 foamable resin. This may result in consider-
 able financial saving as smaller amounts of
 45 these additives or compositions containing
 them may be used and many of these addi-
 tives are expensive. Examples of fillers which
 may be included to improve the stiffness of
 the moulding include glass and asbestos
 50 fibres or glass and mica plates optionally
 coated with a resin to improve adhesion be-
 tween the filler and the thermoplastic. If
 desired, different additives may be included
 in the foamable and unfoamable composi-
 55 tions. Additives that may be etched to im-
 prove the adhesive properties of the surface
 may be included in the unfoamable material,
 for example, titanium dioxide may be in-
 cluded and may subsequently be etched with
 60 chromic acid.

The amount of unfoamable thermoplastic
 resin that is injected into the mould will
 depend upon the size of the mould and the
 required thickness of the unfoamed surface
 65 skin. Any thermoplastic which may be in-

jection moulded may be used as the unfoam-
 able resin and examples of suitable thermo-
 plastics include polymers of α -olefines such as
 polyethylene, polypropylene, polybutene,
 70 and poly-4-methyl pentene-1 and copoly-
 mers of such olefines; homo-polymers and
 copolymers of vinyl chloride optionally con-
 taining a plasticiser; homo-polymers and
 copolymers of styrene; homo-polymers and
 75 copolymers of methyl methacrylate and
 linear polyesters such as polyethylene tereph-
 thalate. The choice of the resin will depend
 upon the use to which the article is to be put,
 for example, if a stiff panel is required to
 80 form, for example, part of the body of a
 motor car then a stiff outer skin is required
 and polypropylene is a suitable resin. How-
 ever, if a flexible moulding is required, for
 example, for the interior trim of a motor
 85 car then a plasticised vinyl chloride polymer
 will be eminently suitable as the unfoamable
 thermoplastic resin.

The thermoplastic resin which contains
 the blowing agent may be the same as or
 different from the unfoamable thermoplastic
 resin and may conveniently be a resin
 90 selected from those resins described above as
 being suitable as the unfoamed thermoplastic
 resin.

In one embodiment of the invention at
 95 least part of one or more walls of the mould
 cavity may be lined with a removable insert
 prior to the injection of the thermoplastic
 materials into the mould. The thermoplastic
 material will then adhere to the insert when
 it is injected into the mould and thus this
 100 technique may be used to provide a rigid
 backing to the cellular article having the un-
 foamed skin. This is particularly useful in
 the production of flexible mouldings such as
 the interior trim of a motor vehicle where it
 is desirable to have a resilient trim with a
 pleasing surface which can be rigidly mounted
 105 in the motor vehicle; in this case one of the
 walls of the mould may be lined to provide
 the backing to mount the trim whereas the
 other is not lined thus providing the pleasing
 surface. Examples of suitable lining materials
 include wooden sheets such as plywood and
 110 hardboard, metal sheets or sheets of thermo-
 plastic materials which do not melt at the
 moulding temperatures. The lining could if
 desired be of a flexible material.

There are several methods by which the
 process of the present invention may be
 120 operated, one of which uses a conventional
 injection moulding machine in which a pre-
 determined amount of the unfoamable
 material is introduced into the front of the
 barrel of the injection moulding machine
 125 and the foamable composition introduced into
 the barrel behind the unfoamable material.
 This method however suffers from the dis-
 advantage that if the process is operated re-
 130 petitively, which is desirable, the second and

subsequent shots of unfoamable material are being introduced into a region of the barrel of the machine which already contains some of the foamable composition and, thus, mixing of the two occurs. A more convenient, and our preferred method is to use two injection barrels, both of which feed material through a common sprue into the mould. The operation of the machine is synchronised so that the required quantity of unfoamable material is first introduced into the nozzle, the required quantity of foamable material introduced behind the unfoamable material and the total amount injected into the mould so that the nozzle is emptied and further unfoamable material may be introduced into the sprue without it mixing with any of the foamable composition. Alternatively two injection moulding machines may be used, each of which injects material through separate nozzles into the mould, the timing of the operation of the machines being correlated so that the desired amount of unfoamable material is first introduced into the mould and is then followed by the desired amount of the foamable composition; some foamable composition may be introduced before all the unfoamable material has been introduced. In operation of the present invention it is essential that the foamable material be introduced into the charge of unfoamable material that is in the mould. In this way the injection and expansion of the foamable material urges the unfoamable material onto the interior surface of the mould somewhat like the blowing up of a balloon. Thus, it is preferred that both the unfoamable and the foamable material be injected into the mould through the same nozzle and if this is not the case the points at which the two nozzles introduce material into the mould should be sufficiently close that the foamable material is introduced into the charge of the unfoamable material.

Another method which may be used comprises using a reciprocating screw type injection moulding machine which is provided with an inlet port either at some position along the extruder screw or at some position in the barrel wall through which a blowing agent may be injected into the thermoplastic material for convenience we prefer to use a liquid or gaseous blowing agent. So that in operation a thermoplastic material is introduced to the injection moulding machine and carried along the barrel of the machine by the screw so that a plug of the material builds up at the ejection end of the machine, simultaneously forcing the screw of the machine back. After a plug of the required size has been formed a gaseous or liquid blowing agent is injected into the thermoplastic material in the barrel of the machine until the required quantity of thermoplastic material containing the blowing agent has

built up behind the original plug of material. The supply of blowing agent is then discontinued and the thermoplastic material injected into the mould. This process may be operated repetitively using a steady supply of blowing agent free polymer.

In a further embodiment of the present invention a second amount of the unfoamable thermoplastic material may be injected into the mould after the thermoplastic resin composition containing the blowing agent. An advantage of using this technique is that the second amount of the unfoamable material removes residual foamable material there may be in the injection nozzle and thus leaves the nozzle clean for the next moulding operation.

The surface of the moulds used in the present invention may be textured to impart any desired surface finish to the moulded article. For example, the mould surface may be smooth, grained, matt or have a regular pattern thereon so as to impart a corresponding surface finish to the unfoamed skin of the article. Alternatively, the mould may be coated with a high temperature resistant material, for example, silicone rubber, to impart a desired surface finish.

The blowing agent which may be used to foam the thermoplastic materials may be solids which decompose on heating to yield a gas which effects foaming, may be liquids which vaporise on heating or on the reduction of the pressure to effect foaming or may be gaseous. The liquids are preferably organic liquids which are inert to and may be absorbed by the thermoplastic material. The gas liberated by the solid type of blowing agent should preferably be inert to the thermoplastic material. By the activation temperature of the blowing agent we mean that the temperature of the thermoplastic resin composition containing a blowing agent as it leaves the injection nozzle is sufficiently high for the thermoplastic resin to foam under the conditions to which it is subjected in the mould. This temperature is determined by the nature of the blowing agent; for example if the blowing agent is a solid that liberates a gas on heating the temperature of the thermoplastic as it leaves the nozzle should be above the temperature at which the gas is liberated and is with some solid blowing agents dependent on the pressure. If, however, the blowing agent is a liquid which vapourises on heating the conditions under which the gas is liberated will depend on both the temperature of the thermoplastic and the pressure to which it is subjected; in this situation the temperature of the resin as it leaves the nozzle should be sufficiently high that, when a constant volume mould cavity is used the gas will be formed under the pressures to which the material is subjected in the mould and when a mould

with an enlargeable cavity is used the gas will be formed when the pressure on the mould cavity is relieved to allow it to expand. The thermoplastic resin may be brought to this temperature either by heating it to the required temperature as it passes along the barrel although the pressure inside the barrel should be sufficiently high to prevent foaming or by maintaining the temperature in the barrel below the activation temperature and relying on frictional heating of the material as it is ejected through the nozzle to raise it to the activation temperature. This second method is our preferred method. Examples of suitable liquid blowing agents include hydrocarbons such as pentane and the halo- and polyhalo-hydrocarbons examples of suitable solid blowing agents include the nitrogen liberating blowing agents, for example the azo and hydrazo compounds, and also the carbon dioxide liberating blowing agents for example carbonates and bicarbonates. Examples of gases which may be used as blowing agents include nitrogen and carbon dioxide. In some instances it may be necessary to include a nucleating agent in the foamable material to provide sites for bubble formation.

The process of the present invention may be used to produce a wide variety of articles having a foamed core with an unfoamed skin. Articles having a rigid or a flexible skin may be prepared; examples of articles with a rigid skin include pieces of luggage—for example suitcases, articles of furniture for example chair seats and television cases, and panels which may be building panels or panels to form the bodies of motor vehicles and trains. Examples of articles with a flexible surface skin include parts for the interior trim of motor cars, railway carriages, caravans, aeroplanes, seat covers, for example car seat covers, and a wide variety of other applications. Positions such as ribs or bosses where the article may be attached to a supporting member may be integrally moulded with the article and the process of the present invention has the added advantage that these relatively thick positions may be moulded without the formation of voids or sink marks in the surface of the article at that point. If such thick positions are moulded without a foamed core sink marks tend to appear because of the contraction of the relatively large amount of material at that point.

The present invention is illustrated but in no way limited by reference to the accompanying examples.

EXAMPLE 1

60 An 18 inch diameter disc mould was fitted on an 18 oz. injection moulding machine which had a 350 ton hydraulic lock on the mould.

A charge of a plasticised polyvinyl chloride

composition (of B.S. Softness 40) sufficient to fill 1/3 of the barrel of the injection moulding machine was placed in the machine and allowed to reach the front of the barrel. The remainder of the barrel was then filled with a plasticised polyvinyl chloride composition containing

Polyvinyl chloride	100 parts by weight
Di-iso-octyl phthalate (plasticiser)	80 parts by weight
Epoxidised Soya Bean Oil	2 parts by weight
Diphenyl decyl phosphite (Kicker)	1 part by weight
Stearic acid	0.3 part by weight
Azodicarbonamide paste (blowing agent)	3 parts by weight

The "kicker" activates the decomposition of the blowing agent.

The azodicarbonamide paste was made up by mixing 1 part by weight of azodicarbonamide with 1 part by weight of di-iso-octyl phthalate on a mill. The temperature along the barrel of the machine was gradually increased from 160°C to 200°C just before the nozzle, the nozzle itself being at 190°C. The complete charge was injected into the mould which had a cavity 1/16 inch wide and allowed to cool therein for 1 second. The pressure holding the mould closed was then reduced and the mould allowed to open so that its cavity was 5/16 inch wide and the composition allowed to cool. Finally the mould was completely opened and the sample removed.

A moulding having a surface skin 1/32 inch thick and a foamed core 1/4 inch thick and density of 0.4 grams/cc. was obtained. The moulding had a pleasing appearance and the surface of the moulding had fewer imperfections than a moulding produced by repeating the moulding cycle with a charge composed entirely of foamable material. The process was repeated using a red unfoamable composition and a blue foamable composition and a moulding having an unfoamed, red surface skin and a blue foamed core with a sharp linear distinction between the two was obtained.

EXAMPLE 2

A charge of a propylene copolymer containing 15% by weight of asbestos fibres sufficient to fill one half of the barrel of the injection moulding machine used in Example 1 was fed to the machine and allowed to reach the front of the barrel. The remainder of the barrel was then filled with a propylene copolymer containing 0.35% by weight of azodicarbonamide (as blowing agent). The temperature along the barrel ranged from 215°C just before the nozzle to 160°C at the rear of the screw.

The complete charge was injected into the mould which had a cavity 3/32 inch wide

and allowed to cool therein for 3 seconds. The pressure holding the mould closed was then reduced and the mould allowed to open until its cavity was 1/4 inch wide; the sample was then allowed to cool before the mould was completely opened and the sample removed.

A moulding having a surface skin of asbestos filled propylene copolymer 1/32 inch thick and a foamed core of density 0.4 grams/cc was obtained. The moulding obtained had fewer surface imperfections than a moulding produced by repeating the moulding cycle with a charge composed entirely of asbestos filled propylene copolymer containing 0.35% by weight of azodicarbonamide.

EXAMPLE 3

A mould 12 1/2 inches in diameter and 2 1/2 inches deep was fitted to the injection moulding machine used in the previous examples and a charge of propylene copolymer containing red pigment and sufficient to fill 1/4 of the barrel of the injection moulding machine was introduced to the machine and allowed to reach the front of the barrel. The remainder of the barrel was then filled with a propylene copolymer containing .35% by weight of azodicarbonamide (as blowing agent). The temperature along the barrel of the machine was gradually increased from 160°C to 195°C before the nozzle; the nozzle itself being at 190°C.

The complete charge was then injected into the mould which had a cavity 1/8 inch wide and allowed to cool therein for 1 second. The pressure holding the mould closed was then reduced and the mould allowed to open so that the cavity became 1/4 inch wide. The sample was allowed to cool before the mould was opened completely and the sample removed.

A moulding having a surface skin 1/32 inch thick and a foamed core 3/16 inch thick and of density of 0.4 grams/cc was obtained. The moulding was formed with a rib 1 inch deep and 1/16 inch thick and, as well as having a blemish free surface, the moulding was free from sink marks in its surface opposite the point where this rib was formed.

WHAT WE CLAIM IS:—

1. A process for the production of articles having a cellular core and an unfoamed surface skin comprising injecting a predetermined amount of an unfoamable thermoplastic resin into mould cavity and subsequently, and before the central portion of said unfoamable thermoplastic resin has set, injecting a thermoplastic resin composition containing a blowing agent at a temperature at or above the activation temperature of the blowing agent into the charge of the unfoamable thermoplastic resin that is in the mould

cavity and maintaining the composition within the cavity for sufficient time to allow or cause the foamable composition to foam and the thermoplastic materials to set.

2. A process according to claim 1 in which the walls of the mould cavity are cooled.

3. A process according to claim 1 or claim 2 in which the mould cavity may be enlarged to allow the foamable composition to expand.

4. A process according to claim 3 in which a predetermined amount of expansion of the mould cavity is allowed to take place after the foamable composition has been injected into the mould.

5. A process according to claim 4 in which the mould cavity is permitted to enlarge by instantaneously relieving the pressure holding opposed surfaces of the mould together.

6. A process according to claim 4 in which the pressure holding opposed surfaces of the mould together is gradually reduced, thus allowing a controlled expansion of the mould.

7. A process according to claim 5 or claim 6 in which the enlargement of the mould is effected by the pressures generated within the mould by the gas generated by the blowing agent.

8. A process according to any of the preceding claims in which at least part of one or more walls of the mould cavity is lined with a removable insert prior to the injection of the thermoplastic materials into the mould.

9. A process according to any of the preceding claims in which the unfoamable thermoplastic resin and the thermoplastic resin composition containing a blowing agent are introduced into an injection nozzle from two separate injection barrels, operation of the machine being synchronised so that the required quantity of unfoamable material is first introduced into the mould.

10. A process according to any of claims 1 to 8 in which a reciprocating screw type injection moulding machine is used which is provided with an inlet port either at some position along the extruder screw or at some position in the barrel wall and wherein when a plug of the required amount of unfoamable material has build up adjacent the injection orifice of the machine a blowing agent is injected into the thermoplastic material through said inlet port until the required quantity of thermoplastic material containing the blowing agent has built up behind the original plug of material the supply of blowing agent is then discontinued and the thermoplastic materials injected into the mould.

11. A process according to any of the preceding claims in which a second amount of unfoamable thermoplastic material is injected into the mould after the thermoplastic material containing the blowing agent.

12. A process according to any of the pre-

and allowed to cool therein for 3 seconds. The pressure holding the mould closed was then reduced and the mould allowed to open until its cavity was 1/4 inch wide; the sample was then allowed to cool before the mould was completely opened and the sample removed.

A moulding having a surface skin of asbestos filled propylene copolymer 1/32 inch thick and a foamed core of density 0.4 grams/cc was obtained. The moulding obtained had fewer surface imperfections than a moulding produced by repeating the moulding cycle with a charge composed entirely of asbestos filled propylene copolymer containing 0.35% by weight of azodicarbonamide.

EXAMPLE 3

A mould 12 1/2 inches in diameter and 2 1/2 inches deep was fitted to the injection moulding machine used in the previous examples and a charge of propylene copolymer containing red pigment and sufficient to fill 1/4 of the barrel of the injection moulding machine was introduced to the machine and allowed to reach the front of the barrel. The remainder of the barrel was then filled with a propylene copolymer containing .35% by weight of azodicarbonamide (as blowing agent). The temperature along the barrel of the machine was gradually increased from 160°C to 195°C before the nozzle; the nozzle itself being at 190°C.

The complete charge was then injected into the mould which had a cavity 1/8 inch wide and allowed to cool therein for 1 second. The pressure holding the mould closed was then reduced and the mould allowed to open so that the cavity became 1/4 inch wide. The sample was allowed to cool before the mould was opened completely and the sample removed.

A moulding having a surface skin 1/32 inch thick and a foamed core 3/16 inch thick and of density of 0.4 grams/cc was obtained. The moulding was formed with a rib 1 inch deep and 1/16 inch thick and, as well as having a blemish free surface, the moulding was free from sink marks in its surface opposite the point where this rib was formed.

WHAT WE CLAIM IS:—

1. A process for the production of articles having a cellular core and an unfoamed surface skin comprising injecting a predetermined amount of an unfoamable thermoplastic resin into mould cavity and subsequently, and before the central portion of said unfoamable thermoplastic resin has set, injecting a thermoplastic resin composition containing a blowing agent at a temperature at or above the activation temperature of the blowing agent into the charge of the unfoamable thermoplastic resin that is in the mould

cavity and maintaining the composition within the cavity for sufficient time to allow or cause the foamable composition to foam and the thermoplastic materials to set.

2. A process according to claim 1 in which the walls of the mould cavity are cooled.

3. A process according to claim 1 or claim 2 in which the mould cavity may be enlarged to allow the foamable composition to expand.

4. A process according to claim 3 in which a predetermined amount of expansion of the mould cavity is allowed to take place after the foamable composition has been injected into the mould.

5. A process according to claim 4 in which the mould cavity is permitted to enlarge by instantaneously relieving the pressure holding opposed surfaces of the mould together.

6. A process according to claim 4 in which the pressure holding opposed surfaces of the mould together is gradually reduced, thus allowing a controlled expansion of the mould.

7. A process according to claim 5 or claim 6 in which the enlargement of the mould is effected by the pressures generated within the mould by the gas generated by the blowing agent.

8. A process according to any of the preceding claims in which at least part of one or more walls of the mould cavity is lined with a removable insert prior to the injection of the thermoplastic materials into the mould.

9. A process according to any of the preceding claims in which the unfoamable thermoplastic resin and the thermoplastic resin composition containing a blowing agent are introduced into an injection nozzle from two separate injection barrels, operation of the machine being synchronised so that the required quantity of unfoamable material is first introduced into the mould.

10. A process according to any of claims 1 to 8 in which a reciprocating screw type injection moulding machine is used which is provided with an inlet port either at some position along the extruder screw or at some position in the barrel wall and wherein when a plug of the required amount of unfoamable material has build up adjacent the injection orifice of the machine a blowing agent is injected into the thermoplastic material through said inlet port until the required quantity of thermoplastic material containing the blowing agent has built up behind the original plug of material the supply of blowing agent is then discontinued and the thermoplastic materials injected into the mould.

11. A process according to any of the preceding claims in which a second amount of unfoamable thermoplastic material is injected into the mould after the thermoplastic material containing the blowing agent.

12. A process according to any of the pre-



ceding claims in which the blowing agent comprises a volatile organic liquid.

5 13. A process according to any of the preceding claims in which the blowing agent comprises a solid which decomposes on heating to yield a gas which is inert to the thermoplastic material.

10 14. A process according to any of the preceding claims in which the surface of the mould is textured to impart a surface finish to the moulded article.

15 15. A process according to any of the preceding claims in which the unfoamable thermoplastic resin contains additives which improve the resistance of the article to moisture, light or heat.

20 16. Articles having a cellular core and an unfoamed surface skin whenever produced by a process according to any of the preceding claims.

17. Car panels whenever produced by a process according to any of claims 1 to 15.

18. Articles of furniture whenever produced by a process according to any of claims 1 to 15.

25 19. A process for the production of articles having a cellular core and an unfoamed surface skin substantially as hereinbefore described with particular reference to Example 1.

30 20. A process for the production of articles having a cellular core and an unfoamed surface skin substantially as hereinbefore described with particular reference to Examples 2 and 3.

25

30

35

C. GRATWICK,
Agent for the Applicants.

Printed for Her Majesty's Stationery Office by the Courier Press, Leamington Spa, 1969.

Published by the Patent Office, 25, Southampton Buildings, London, W.C.2, from which copies may be obtained.

