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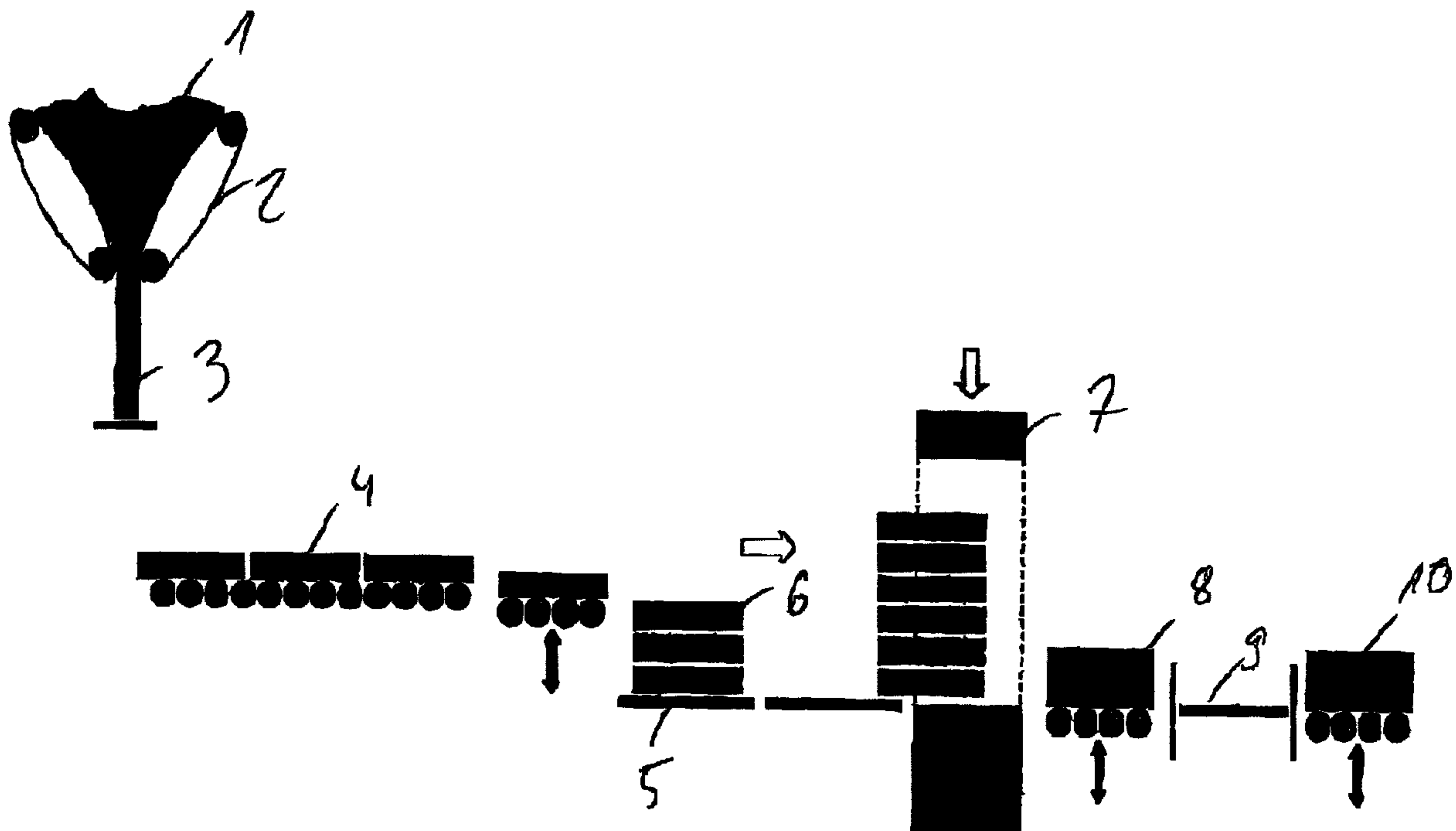
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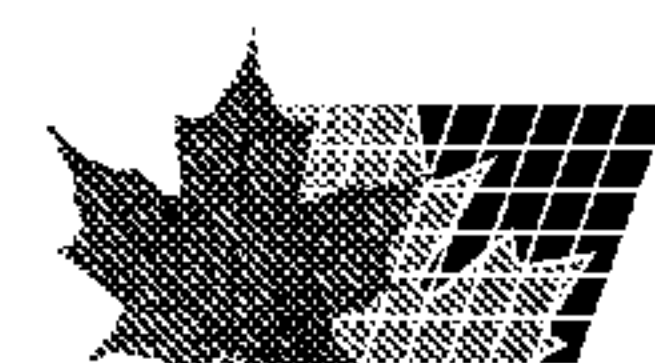
(54) Titre : METHODE DE FABRICATION DE CORPS PROFILES EN GRAPHITE EXPANSE

(54) Title: METHOD FOR MANUFACTURE OF SHAPED BODIES OF EXPANDED GRAPHITE



(57) Abrégé/Abstract:

The present invention relates to a process for the preparation of mouldings comprising compressed expanded graphite, to the mouldings prepared by this process and to the use thereof. The three-dimensional mouldings comprising compressed graphite expandate are prepared by bonding together two-dimensional pre-formed pieces comprising pre-compressed expanded graphite under the influence of pressure to give single-piece mouldings.



Abstract

The present invention relates to a process for the preparation of
5 mouldings comprising compressed expanded graphite, to the mouldings
prepared by this process and to the use thereof. The three-
dimensional mouldings comprising compressed graphite expandate are
prepared by bonding together two-dimensional pre-formed pieces
comprising pre-compressed expanded graphite under the influence of
10 pressure to give single-piece mouldings.

Process for the preparation of mouldings comprising expanded graphite

Description

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The present invention relates to a process for the preparation of mouldings comprising expanded graphite, in particular mouldings having a thickness of at least 50 mm, to the mouldings prepared according to this process, and to the use thereof.

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Patent Applications DE 41 17 077 A, DE 41 17 074 A and DE 40 167 10 propose the preparation of three-dimensional mouldings directly from pulverulent exfoliated graphite, in which either

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- exfoliatable graphite is expanded incompletely with the supply of moderate heat in a fluidised bed or actually in the final form, and the expansion is then brought to completion at elevated temperature in the mould, or
- exfoliatable graphite which has been incompletely expanded (pre-expanded) with the supply of moderate heat is pressed in a mould to give a pre-formed piece, and the expansion is then brought to completion in the mould at a higher temperature, or
- a moist preparation of exfoliatable graphite is expanded in a mould with the supply of heat.

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The final stage of the expansion therefore takes place in each case in the final form, and no compacting is provided for after completion of the expansion. The mould must on the one hand be largely closed so that the geometry of the graphite body does not change during expansion, but must on the other hand permit the escape of air. The mouldings obtained by this process should be dimensionally stable and have a homogeneous density. In the preferred variants of the aforementioned processes various additives and auxiliary substances, in particular binders, are added to the exfoliatable graphite.

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Handling of partially expanded graphite powder is necessary in the process which proceeds by way of a pre-expansion step. The low bulk density (30 to 100 g/l in the Embodiment Examples of DE 40 167 10 A) and the bulkiness of the particles make it difficult to fill moulds
5 completely with it, and dust pollution is high.

It is proposed according to the invention to prepare three-dimensional mouldings comprising compressed graphite expandate by way of the intermediate stage of two-dimensional semi-finished
10 products or pre-formed pieces comprising incompletely compressed expanded graphite.

The process according to the invention includes the steps:

- preparation of two-dimensional semi-finished products comprising
15 graphite expandate, as an option having further additives, wherein the graphite expandate in the semi-finished product is compressed only slightly,
- stacking one on top of the other the desired number (at least two) of semi-finished products or planar pre-formed pieces cut
20 out from a semi-finished product,
- bonding of the semi-finished products or pre-formed pieces by the influence of pressure to give a single-piece moulding,
- final shaping and further post-working as an option.

"Two-dimensional semi-finished products" or "planar pre-formed
25 pieces" are understood hereinafter to be formed bodies whose thickness is substantially less than their superficial extent, for instance flat slabs, disks, webs and similar planar formed bodies. Such two-dimensional semi-finished products can be created simply in a known manner by pressing the pulverulent starting material to give
30 a planar formed body, for example a flat strand, a web or a slab.

In accordance with the process according to the invention the transition from the formless powder to the complex three-dimensional moulding need not be completed in one step. In terms of processing technology it is substantially less costly to convert a formless
35 material, for example a powder or a slurry, initially into a simple

form, for example that of a flat slab, than directly into a complex three-dimensional form. Moreover, in the process according to the invention the difficulties encountered in the prior art when charging dies with dusty powder, bulky particles or viscous masses do not occur.

A further advantage of the process according to the invention resides in the fact that, compared with the prior art single-stage process, the process according to the invention offers more opportunity for variation and optimisation of the product parameters than the substantially single-stage prior art process, precisely because it includes a plurality of steps. Thus different semi-finished products prepared in the first step can be combined in a multitude of ways, so that a multitude of different products is obtainable, for example by combining semi-finished products of different density and/or thickness and/or composition and/or coating.

Further details, variants and advantages of the process according to the invention, the products obtainable by this process, and the use thereof are contained in the detailed description which follows, and in the Figures which show:

- Figure 1 Flow chart of the process according to the invention;
Figure 2 Flow chart of the process according to the invention,
 with the semi-finished product comprising graphite
 expandate partially coated with a heat-storing material.

Figure 1 shows schematically how the process according to the invention is carried out.

The starting material of the process according to the invention is graphite expandate 1 (also designated as exfoliated graphite). Fibrous or particulate additives or fillers of metal, carbon, ceramic materials or mineral materials can be added to the graphite expandate, for example carbon black or other forms of carbon particles to optimise the thermal and electrical conductivity, fibres, for example carbon fibres or metallic fibres, and/or ceramic particles or mineral particles, for example comprising heat-storing

substances. The selection of these materials is dictated by the intended use of the mouldings which are to be prepared, the invention is not bound to the use of any particular fibrous or particulate additives.

5 The preparation of expanded graphite (graphite expandate, exfoliated graphite) is known from, inter alia, US-A 3 404 061. In order to prepare expanded graphite, graphite intercalation compounds or graphite salts, for example graphite hydrogensulfate or graphite
10 nitrate, are subjected to thermal shock. Here, the volume of the graphite particles increases by a factor of from 200 to 400, and the bulk density falls to 2 to 20 g/l. The so-called graphite expandate thus obtained consists of worm-shaped or concertina-shaped, bulky aggregates. If these particles are compressed under pressure, they
15 become entangled and interlocked with one another. Owing to this effect, self-supporting planar formed bodies, for example continuous webs, sheetings or slabs, can be prepared without the addition of binder. Compression to give sheetings having thicknesses of from 0.15 to 3 mm and densities of from 0.1 to 2 g/cm³, preferably 0.7 to 1.8 g/cm³, is in particular known. These sheetings are used, for
20 example, as a sealing material.

However, in the semi-finished products for the process according to the invention the graphite expandate is less strongly compressed than in the known graphite sheetings. It is crucial to the process according to the invention that the graphite expandate in the semi-
25 finished product is compressed only to an extent such that, on the one hand, a cohesion is brought about between the particles that is sufficient for the preparation of a planar semi-finished product, but on the other hand there still exists the possibility of an appreciable further compression. The graphite expandate, which has
30 a density of 2 to 20 g/l, preferably 3 to 15 g/l, is compressed in the semi-finished product to a density of from 0.01 to a maximum of 0.2 g/cm³, preferably 0.02 to 0.1 g/cm³. Because of this relatively low compression it can be assumed that the expandate particles are still in part worm-shaped, so that in a subsequent compression step
35 they are able to continue to become entangled and interlocked. Because of the low compression of the expandate the semi-finished products have rough surfaces, unlike the smooth surfaces of the known graphite sheetings.

The semi-finished products are preferably prepared continuously by pressing between two textile conveyor belts 2 in the form of long stretched-out continuous webs 3. Pre-formed pieces 4 of appropriate size are then cut or stamped out from the planar semi-finished product thus obtained. The size, that is to say, the superficial extent, as well as the surface geometry of the pre-formed pieces 4 are substantially dictated by the product to be prepared; the process according to the invention sets no limits in these respects. For practical reasons square slabs having a surface measuring from 100 mm x 100 mm to 2000 mm x 2000 mm are preferably used as the pre-formed pieces. However, circular, rectangular or differently shaped pre-formed pieces can naturally also be utilised.

A semi-finished product having the dimensions of the desired pre-formed piece 4, for example in the shape of a slab of a certain surface area and thickness, can naturally also be pressed directly from the graphite expandate. However, this variant is less preferable because of the discontinuous method of working as compared with the continuous preparation of flat strands or webs.

Suitable semi-finished products and pre-formed pieces prepared therefrom for the process according to the invention are between 4 and 45 mm, preferably between 5 and 40 mm thick. The use of thinner semi-finished products is not very economic, because the thinner the semi-finished product is, the more pre-formed pieces from this semi-finished product must be stacked one on top of the other in order to obtain a moulding of a specified size. Semi-finished products substantially thicker than 55 mm are likewise unsuitable. During the subsequent pressing together when the latter were stacked one on top of the other, the very thick pre-formed pieces obtained from such a semi-finished product would themselves predominantly be deformed and compressed into themselves, and would in that case barely bond with the adjacent pre-formed pieces. There is consequently a risk that only a stack of thinner deformed pre-formed pieces of a higher density that barely adhere to one another would be obtained from a stack of thick pre-formed pieces of lower density that lie loosely one on top of the other, instead of the desired single-piece moulding. Moreover, the marked deformation of thick pre-formed pieces leads to material breaking out at the sides during the pressing together.

The desired three-dimensional component is built up from these flat pre-formed pieces 4 by stacking the necessary number of flat pre-formed pieces 4 one on top of the other on a multi-deck collecting device 5. At least two pre-formed pieces are laid one on top of the other. This stack 6 is then pressed together in a press 7, wherein the pre-formed pieces lying one on top of the other bond together to give a single-piece raw moulding 8 under the influence of pressure. The compression reserve still available in the semi-finished products is exploited during the pressing together, so that the density rises to values of between 0.025 and 0.4 g/cm³, preferably 0.03 to 0.25 g/cm³.

The thickness (height) of the raw moulding 8 obtainable by pressing together the stack 6 of pre-formed pieces 4 lying one on top of the other is dictated by the number and thickness of the pre-formed pieces stacked one on top of the other as well as by the pressure applied during pressing. The number of pre-formed pieces to be stacked one on top of the other must take account of the volumetric shrinkage resulting from the compression, that is to say, for example, when compression takes place to approximately half of the initial volume during pressing, the volume of the stack of pre-formed pieces must be approximately double that of the desired end product.

A particular advantage of the process according to the invention is that mouldings having a thickness (in the direction of pressing) of more than 50 mm can also be prepared.

The pre-formed pieces stacked one on top of the other that are to be bonded together must not necessarily be identical as to thickness, density and composition. On the contrary, the process according to the invention includes the possibility of combining pre-formed pieces of different thickness, density and/or composition in accordance with the desired properties of the moulding to be prepared. In this way, for example, mouldings can be prepared having a tailor-made spatial variation in composition.

The surfaces of individual pre-formed pieces or of all the pre-formed pieces can be provided in part with a coating, a cover, a deposit or an overlay. It is crucial to the invention that the graphite particles of the two pre-formed pieces are able to contact

one another and interact, that is to say become entangled and interlocked with one another, over at least 50 % of each interface between two pre-formed pieces.

5 Some of the pre-formed pieces to be bonded may, if this is necessary for the intended function of the moulding to be prepared, be provided with an impregnation, for example with furan resin. This is advisable, for example, if the desired moulding is to have layers having lower porosity and hence lower permeability, for example layers that act as a vapour barrier.

10 For the application of mouldings according to the invention as heat storage means, individual pre-formed pieces may be impregnated with heat-storing substances, for example with phase-change materials that store latent heat, for example with paraffins or with aqueous solutions of salts which form hydrates and thus act as phase-change
15 materials. Such phase-change materials and the action thereof are known to those skilled in the art. From 30 to 40 % of the pore volume of the pre-formed piece is preferably filled by the impregnation.

No additional supply of heat is necessary for the pressing
20 operation. The pressures applied are within the region 10 to 500 bar (1 to 50 MPa). Stationary platen presses 7 can be used for pressing the pre-formed pieces together. The air displaced during compression can escape laterally from the platen press. However, continuously operating installations having pairs of rolls can also
25 be utilised, for example calendering mills.

The raw mouldings 8 thus obtained are durable and stable, and the individual flat pre-formed bodies are bonded together non-detachably. This is attributable to the fact that during the pressing together of the pre-formed pieces stacked one on top of the
30 other the worms of the expandate from the adjacent pre-formed pieces, which are compressed only slightly, become entangled and interlocked across the interfaces between the individual pre-formed pieces. However, the invention is not tied to this theory. In any case, when the moulding is cut open perpendicularly to the planes of
35 the stacked pre-formed pieces, the boundaries between the originally individual pre-formed pieces are no longer visible at the cut faces in the finished moulding. The number of pre-formed pieces from

which it has been prepared is therefore no longer discernible from the finished moulding.

If the pressing together takes place in an open platen press, build-ups form on the lateral faces of the raw moulding as a result of displacement of the material under the influence of the pressing force. These are subsequently removed by means of mechanical processing in the cutting station 9, and the raw mouldings 8 are brought to the desired final form. It has emerged that the raw mouldings 8 can be brought very simply to the desired shape, for example the shape of a block 10, by processing with a saw or a carpet knife.

In the post-working shaping operation the mouldings can furthermore be provided with, for example, recesses and grooves, and undercuts can be worked from them, or patterns can be impressed into the surface, inter alia.

The operations of pressing and cutting can, however, also be undertaken in a combined installation, for example in a press having movable jaws at the front faces, which after pressing are moved down and thus cut off the edge build-ups.

Pressing close to final contours in a ventilatable die is also possible; however, this variant is less preferred on account of the high tooling-up cost.

Different mouldings obtained by the process according to the invention can be bonded together by means of adhesives in order to obtain components having complex geometry. All the known and generally widely used adhesives are suitable for this purpose.

Functional components can be pressed into the surfaces of the mouldings, for example heating tubes for heating and air-conditioning technology applications.

Further finishing processes as options include, for example, the application of coatings to the surfaces of the mouldings, for example by painting or by lamination with planar materials, for example wovens or other textile materials, and the impregnation of the mouldings with certain substances, depending on the intended function of the mouldings.

The conductivity of the mouldings for heat and electrical current depends on the compression. The greater the compression, the higher are the electrical and thermal conductivity and the more pronounced is the anisotropy of the thermal and electrical conductivity. It is therefore possible to control the isotropy/ anisotropy of the thermal and electrical conductivity by way of the compression.

An important field of application of the mouldings according to the invention, in particular blocks, is air-conditioning and temperature control technology. On account of the thermal conductivity of graphite the first application which suggests itself in this respect is as thermal conductors, for example as heat dissipation bodies for electronic equipment, or as heat exchangers.

In the mouldings according to the invention, by introducing a heat-storing material into the mouldings according to the invention the thermal conductivity of graphite can furthermore be coupled with the heat storage capacity of a material which stores latent or sensible heat. As a result of its thermal conductivity the graphite structure facilitates, respectively, the supplying of heat on charging, and the removal of heat on discharging, the storage means.

The mouldings according to the invention can be readily impregnated or infiltrated with liquids. Heat storage means can thus be prepared from the mouldings obtained by the process according to the invention, by impregnation or infiltration with a heat storage or phase-change material, such as salt hydrate, dissolved in a solvent, preferably water, or with a phase-change material which can be converted into the liquid phase, such as paraffin. From 30 to 40 % of the pore volume of the moulding are preferably filled by the impregnation.

Alternatively, individual pre-formed pieces can be impregnated, as already described, and combined with non-impregnated pre-formed pieces to give one moulding.

However, heat-storing materials that are present in solid form, for instance as a powder or granules, can also be introduced into the

blocks according to the invention. Figure 2 illustrates a method which is suitable for this.

Graphite expandate 1 is pressed by the same method as in Figure 1 between textile conveyor belts 2 to give a web 3, on the upward-
5 facing surface whereof the pulverulent or granular heat-storing material 11, for example magnesite or barite, is then distributed from a metering device 12 in planar manner to give a thin, loose layer. The grains or granule particles typically have diameters of between 0.5 and 5 mm. Slab-shaped pre-formed pieces 4' are then cut
10 out from the sprinkled web 3' in the cutting station, which are stacked one on top of the other on a multi-deck collecting device 5 to give a stack 6' and are pressed together in a stationary press 7. Alternatively, slabs 4 can also be cut out initially from the web 3, and their upward-facing surfaces can then be sprinkled with the
15 heat-storing material 11.

It is crucial to the cohesion of the composite that the sprinkled-on layer is not so thick and compact that in the stack it separates the surfaces of the adjoining graphite slabs completely from one another. An interaction between the slab surfaces must still be
20 possible through the powder bed, so that during pressing, the entanglement and interlocking of the graphite expandate particles among one another, which is crucial to the mutual adhesion of the slabs, occurs. The covering of the surface of each pre-formed piece with the sprinkled-on particles must not therefore exceed 50 %. The
25 strewing is therefore preferably not applied so as to be distributed over the total surface of the pre-formed piece, but only on certain regions, for example in channels or other indentations previously introduced into the surface and arranged in a suitable pattern. These channels or indentations can be cut out of the surface with a
30 cutting tool or pressed in with a stamp-type tool.

When the stack 6' is pressed together the sprinkled-on particles are pressed from the slab surface into those regions of the slabs which are near the surface, so that no sharply separated discrete layers of graphite or of the poured-on powder are discernible in the cross-
35 section of the body.

The separation between graphite layers and layers containing heat-storing material can be further reduced if the thinnest possible

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- pre-formed pieces are used, because the thinner the pre-formed pieces, the thinner the graphite layers between the layers which contain the heat-storing material. Pre-formed pieces having a thickness of from 10 to 20 mm are preferably used for the
- 5 preparation of blocks having heat-storing intermediate layers. In this way a less heterogeneous product having transitional regions between the individual layers can be obtained from heterogeneous semi-finished products built having discrete layers (graphite pre-formed piece with sprinkled-on powder or granules).
- 10 In the same way as the mouldings 8, the raw mouldings 8' charged with the heat-storing material which are thus obtained are then brought to the desired shape, for example the shape of a block 10', in the cutting station 9.
- It is clear that the process in accordance with Figure 2 is not
- 15 confined to the preparation of heat-storing intermediate layers. Mouldings having other functional layers can also be prepared by applying particles of a corresponding functional material to the upward-facing surface of the semi-finished product or the pre-formed pieces. It is crucial for the preparation of the moulding according
- 20 to the invention that a maximum of 50 % of the upward-facing surface of each pre-formed piece is covered by the particles of the functional material.
- Further variants of the process according to the invention consist
- 25 of inserting into the stacks 6 or 6' between the individual pre-formed pieces 4 or 4' plies of perforated plate, punched sheeting, open woven, knit or other wide-meshed planar formed bodies of textile. Wovens having mesh sizes of between 3 mm x 3 mm and 20 mm x 20 mm are, for example, suitable. Plies of wovens introduced
- 30 between the graphite layers bring about an improvement in the mechanical stability, in particular the flexural strength, of the mouldings.
- It is crucial that the intermediate plies do not cover the surfaces of the pre-formed pieces completely, in order that an interaction
- 35 between the slab surfaces is still possible, so that during pressing the graphite expandate particles become entangled and interlocked with one another across the slab boundaries. The extent to which

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each boundary surface between the pre-formed pieces is covered by a overlay or other types of coatings or deposits should not therefore exceed 50 % of the total surface area.

Where the substances introduced between the pre-formed pieces are
5 fusible or thermoplastic (for example punched plastics sheeting) or contain fusible or thermoplastic constituents, for example plastics fibres, the pressing operation can be undertaken at elevated temperature in order to achieve a softening, partial melting or melting of these substances. According to the invention, however,
10 this is not necessary in order to bring about adhesion between the slab surfaces. However, as a result of melting, those layers of the pre-formed pieces which are close to the surface can be infiltrated with the molten substance, so that the moulding obtained has continuous transitions between the graphite layers and the
15 intermediate layers.

It should, finally, be emphasised again that impregnation, infiltration and the introduction of intermediate layers serve only to impart to the mouldings according to the invention certain
20 functions necessary for their use, for example for heat storage. The materials which are introduced as intermediate layers or by means of impregnation do not have to fulfil any gluing, binding or adhesive function, because in the mouldings according to the invention the pre-formed pieces adhere to one another without
25 binders or glues owing to the effects of mutual entanglement and interlocking of the graphite expandate particles across the boundary surfaces of the pre-formed pieces, which effects are achieved by the pressing together.

30 Embodiment Examples

In the Embodiment Examples of the process according to the invention slab-shaped pre-formed pieces are stacked one on top of the other, pressed together in a platen press to give a raw moulding which is cut to give a block. Block is understood here to be a cubic or
35 cuboid body. The pre-formed pieces were prepared from graphite expandate having a density of 3 g/l. Unless otherwise indicated, the pressing operation took place at room temperature. The

"thickness" of the pre-formed pieces or the "thickness/height" of the blocks always means the dimension in the direction of pressing.

Examples 1 to 4: Preparation of 50 mm thick blocks

- 5 A block (Example 1) having a height (thickness) of 50 mm and a density of 0.15 g/cm³ was prepared by stacking one on top of the other 8 slab-shaped pre-formed pieces each having a thickness of 20 mm, a density of 0.025 g/cm³ and an area of 500 mm * 250 mm, and pressing them together at a pressure of 200 bar (20 MPa). The
- 10 built-up side faces were then cut off the raw formed piece. As a result, the length and breadth of the basal area of the block were reduced in each case by 20 mm by comparison with the area of the semi-finished products. The block was cut open in the direction of pressing, the cut faces were homogeneous, the original interfaces
- 15 between the pre-formed pieces were no longer discernible.
- In the Examples 2 to 4 which follow, blocks having in each case a height (thickness) of 50 mm were made from slab-shaped semi-finished products having an area of 400 mm x 600 mm. In each case a plurality of semi-finished products of the same thickness and
- 20 density were stacked one on top of the other and pressed together to make the blocks. The built-up side faces were then cut off the raw formed piece. As a result, the length and breadth of the basal area of the block were reduced in each case by 20 mm by comparison with the area of the semi-finished products.
- 25 Table 1 indicates the thickness and density of the blocks obtained by the process according to the invention, dependent on the number of semi-finished products stacked one on top of the other, their thickness and density.

30 Table 1

No.	Number of slab-shaped pre-formed pieces	Thickness of slab-shaped pre-formed pieces/mm	Density of graphite in slab-shaped pre-formed pieces/g/cm ³	Thickness (=height) of block / mm	Density of graphite in block / g/cm ³
2	2	35	0.055	50	0.084
3	5	35	0.088	50	0.220

4	4	35	0.027	50	0.095
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It is apparent from Table 1 that the material density in the block is dependent on the number of semi-finished products and their density. It is thus possible to achieve certain densities in a targeted manner by appropriate selection of the semi-finished products. This is attractive for the adjustment of certain values and anisotropies of the thermal conductivity, because these depend on the compression.

A comparison between blocks Nos. 2 and 3 shows that, for the same block thickness, as well as the same density and thickness of the semi-finished products, the density of the graphite in the block is greater the more semi-finished products were stacked one on top of the other and pressed together.

Example 5: blocks having heat-storing intermediate layers

A block was prepared by the process according to the invention from four slabs stacked one on top of the other, each having an area of 300 mm x 300 mm, a thickness of 40 mm and a density of 0.02 g/cm³. The upward-facing surfaces of three of the four slabs were each sprinkled with 100 g granular barite (grain size approx. 1 mm) so that a loose planar distribution of the grains was obtained on the slab surface, i.e. the surface was not covered completely. Barite serves as a storage material for sensible heat.

The three sprinkled slabs were then stacked one on top of the other, the fourth slab, which was not sprinkled, was laid uppermost on the stack, and the stack was pressed together to a height of 40 mm at a pressure of 150 bar (15 MPa) at room temperature. The block thus obtained contains three heat-storing intermediate layers, with the transition to the respectively adjacent graphite layers being continuous, because the grains of the heat-storing material are pressed into the slab surfaces during pressing together. The high thermal conductivity of the graphite layers between the heat-storing layers permits rapid heating and cooling, respectively, of the heat storage means.

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Example 6: blocks having reinforcing inserts between the semi-finished products

A block was prepared by the process according to the invention from five slabs stacked one on top of the other, each having a surface of 200 mm x 200 mm, a thickness of 40 mm and a density of 0.02 g/cm³. Plies of a fusible plastics fibre woven material, having a mesh size of 3 mm x 3 mm, were inserted in each case between the slabs when stacking them one on top of the other, so that a stack having an alternating arrangement of graphite slabs and woven plies was obtained. It was pressed together at a temperature of 180 °C at a pressure of 200 bar (20 MPa) to give a block having a thickness of 40 mm.

Example 7: Density-dependence of the level and the anisotropy of the thermal and electrical conductivity of the semi-finished products

It is apparent from a comparison of different pre-formed pieces (Table 2) that both the level and also the anisotropy of the thermal and electrical conductivity of the semi-finished products are dependent on density. The greater the compression, the flatter and more planar are the graphite particles and the more parallel their orientation to the slab plane. For this reason the ratio of the conductivity in the slab plane to the conductivity at right angles to the slab plane rises with increasing density.

Examples 8 and 9: Density-dependence of the level and the anisotropy of the thermal and electrical conductivity of the blocks

The dependence of the level and the anisotropy of the electrical and thermal conductivity on the compression of the graphite, already demonstrated in the previous Example using semi-finished products of differing density, can also be observed in the case of the blocks prepared by the process according to the invention. The comparison of two blocks of different density in Table 3 shows this. Each block was prepared by the process according to the invention from semi-finished products of the same density and thickness stacked one on top of the other.

Table 2

Semi-finished product No.	1	2	3	4	5
Thickness/mm	6	30	25	20	10
Density/ g/cm ³	0.02	0.03	0.04	0.06	0.13
Thermal conductivity/ W/mK					
• in the x-direction		4.2	6.1	6.8	9.9
• in the y-direction		3.5	4.1	5.8	11.1
• in the z-direction		3.0	3.2	3.6	5.1
Electrical resistance/ Ohm*μm					
• in the x-direction	843	475	405	268	148
• in the z-direction	957	640	480	500	233

Table 3

Block No.	8	9
Block thickness (= height) / mm	140	43
Density of graphite in the block/ g/cm ³	0.03	0.14
Number of semi-finished products	5	2
Thickness per semi-finished product/ mm	40	40
Density of graphite in the semi-finished products/ g/cm ³	0.02	0.024
Thermal conductivity / W/mK		
• in the x-direction	3.3	12.6
• in the y-direction	3.0	15.5
• in the z-direction	2.4	4.2
Electrical resistance / Ohm*μm		
• in the x-direction	500	105
• in the z-direction	575	340

Claims

1. Process for the preparation of mouldings comprising compressed graphite expandate, including the steps
 - 5 • preparation of planar pre-formed pieces having a density of from 0.01 g/cm³ to 0.2 g/cm³ from graphite expandate having a density of from 2 g/l to 20 g/l,
 - stacking of at least two pre-formed pieces one on top of the other,
 - 10 • bonding of the pre-formed pieces by the influence of pressure to give a single-piece moulding, wherein the graphite is compressed under the influence of the pressure to a density of from 0.025 g/cm³ to 0.4 g/cm³.
- 15 2. Process according to Claim 1, characterised in that the pre-formed pieces stacked one on top of the other are pressed together at a pressure within the range 1 to 50 MPa.
3. Process according to one of the preceding claims,
20 characterised in that the thickness of the pre-formed pieces is at least 4 mm and a maximum of 45 mm.
4. Process according to one of the preceding claims,
25 characterised in that the density of the graphite expandate from which the pre-formed pieces are prepared is from 3 to 15 g/l.
5. Process according to one of the preceding claims,
30 characterised in that the density of the graphite in the pre-formed pieces is from 0.02 to 0.1 g/l.
6. Process according to one of the preceding claims,
35 characterised in that the density of the graphite in the mouldings is from 0.03 to 0.25 g/l.

7. Process according to one of the preceding claims, characterised in that the thickness of the moulding in the direction of pressing is at least 50 mm.
- 5 8. Process according to one of the preceding claims, characterised in that the graphite expandate contains fibrous or particulate additives comprising metal, carbon, ceramic materials or mineral materials.
- 10 9. Process according to Claim 8, characterised in that particles of a heat-storing material are added to the graphite expandate.
- 15 10. Process according to one of the preceding claims, characterised in that the surfaces of individual pre-formed pieces or of all the pre-formed pieces are provided in partial manner with a coating, a cover, a deposit or an overlay, so that a maximum of 50 % of each interface between the pre-formed pieces is covered by the coating, cover, deposit or overlay.
- 20 11. Process according to Claim 10, characterised in that the upward-facing surfaces of individual pre-formed pieces or of all the pre-formed pieces in the stack are covered with a powder or granules, wherein the powder particles or granule particles cover a maximum of 50 % of the upward-facing surface of the pre-formed piece.
- 25 12. Process according to Claim 11, characterised in that the upward-facing surfaces of individual pre-formed pieces or of all the pre-formed pieces in the stack are covered with a pulverulent or granular heat-storing material, wherein the powder particles or granule particles cover a maximum of 50 % of the upward-facing surface of the pre-formed piece.
- 30 13. Process according to Claim 11, characterised in that indentations are let into the upward-facing surfaces of
- 35

- individual pre-formed pieces or of all the pre-formed pieces in the stack, into which indentations the pulverulent or granular material is introduced, wherein the powder particles or granule particles cover a maximum of 50 % of the upward-facing surface of the pre-formed piece.
- 5
14. Process according to Claim 10, characterised in that the surfaces of individual pre-formed pieces or of all the pre-formed pieces are overlaid with a planar textile material, a perforated plate or a punched sheeting, so that a maximum of 50 % of each interface between the pre-formed pieces is covered with the textile material.
- 10
15. Process according to Claim 10, characterised in that the textile material is a woven material having a mesh size of between 3 mm x 3 mm and 20 mm x 20 mm.
- 15
16. Process according to one of Claims 10 to 15, characterised in that the coating, cover, deposit or overlay contains fusible or thermoplastic constituents and that the operation of bonding the pre-formed pieces takes place under pressure at or above the softening or melting point of this substance.
- 20
17. Process according to one of the preceding claims, characterised in that the semi-finished products stacked one on top of the other differ from one another in their density and/or thickness and/or composition and/or coating.
- 25
18. Process according to one of the preceding claims, characterised in that in alternating manner pre-formed pieces of graphite expandate and overlays of planar textile materials, perforated plate or punched sheeting are stacked one on top of the other, wherein a maximum of 50 % of each interface between the pre-formed pieces is covered by the coating, cover, deposit or overlay, and are then bonded by the influence of pressure to give a single-piece moulding.
- 30
- 35

19. Process according to one of the preceding claims,
characterised in that some of the pre-formed pieces stacked
one on top of the other have an impregnation.
- 5 20. Process according to Claim 19, characterised in that some of
the pre-formed pieces stacked one on top of the other are
impregnated with furan resin.
- 10 21. Process according to Claim 19, characterised in that some of
the pre-formed pieces are impregnated with a heat-storing
material.
- 15 22. Process according to one of the preceding claims,
characterised in that the bonding of the pre-formed pieces
under pressure takes place in a platen press, in a
ventilatable pressing die or between pairs of rolls.
- 20 23. Process according to one of the preceding claims,
characterised in that the moulding obtained by the bonding of
the pre-formed pieces under the influence of pressure is then
brought to the desired final form by mechanical processing.
- 25 24. Process according to one of the preceding claims,
characterised in that the moulding obtained by the bonding of
the pre-formed pieces under the influence of pressure is
impregnated.
- 30 25. Process according to Claim 24, characterised in that the
moulding is impregnated with a heat-storing material.
26. Process according to Claim 25, characterised in that the
moulding is impregnated with a paraffin which acts as a phase-
change material.
- 35 27. Process according to one of the preceding claims,
characterised in that the surfaces of the moulding are coated

with a finish or with a lamination comprising a planar textile material.

28. Use of the mouldings prepared by the process according to
5 Claims 1 to 26 for the conduction or the exchange of heat.
29. Use of the mouldings prepared by the process according to
Claims 9, 12, 21, 25 or 26 for the storage of heat.
- 10 30. Process for the preparation of a heat storage means, which
includes the following steps:
- preparation of planar pre-formed pieces having a thickness
of from 10 to 20 mm and a density of from 0.01 g/cm³ to 0.2
15 g/cm³ from graphite expandate having a density of from 2 g/l
to 20 g/l,
 - coating of the upward-facing surfaces of the pre-formed
pieces with particles of a heat-storing material, so that a
maximum of 50 % of the upward-facing surface of each pre-
formed piece is covered with the particles of the heat-
20 storing material,
 - stacking of the pre-formed pieces one on top of the other,
so that the coated surfaces face upward,
 - laying of an uncoated pre-formed piece onto the stack,
 - bonding of the pre-formed pieces by the influence of
25 pressure to give a single-piece raw moulding, wherein the
graphite expandate is compressed to a density of from
0.025 g/cm³ to 0.4 g/cm³,
 - cutting of the raw moulding to the shape of a block.

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Figure 1

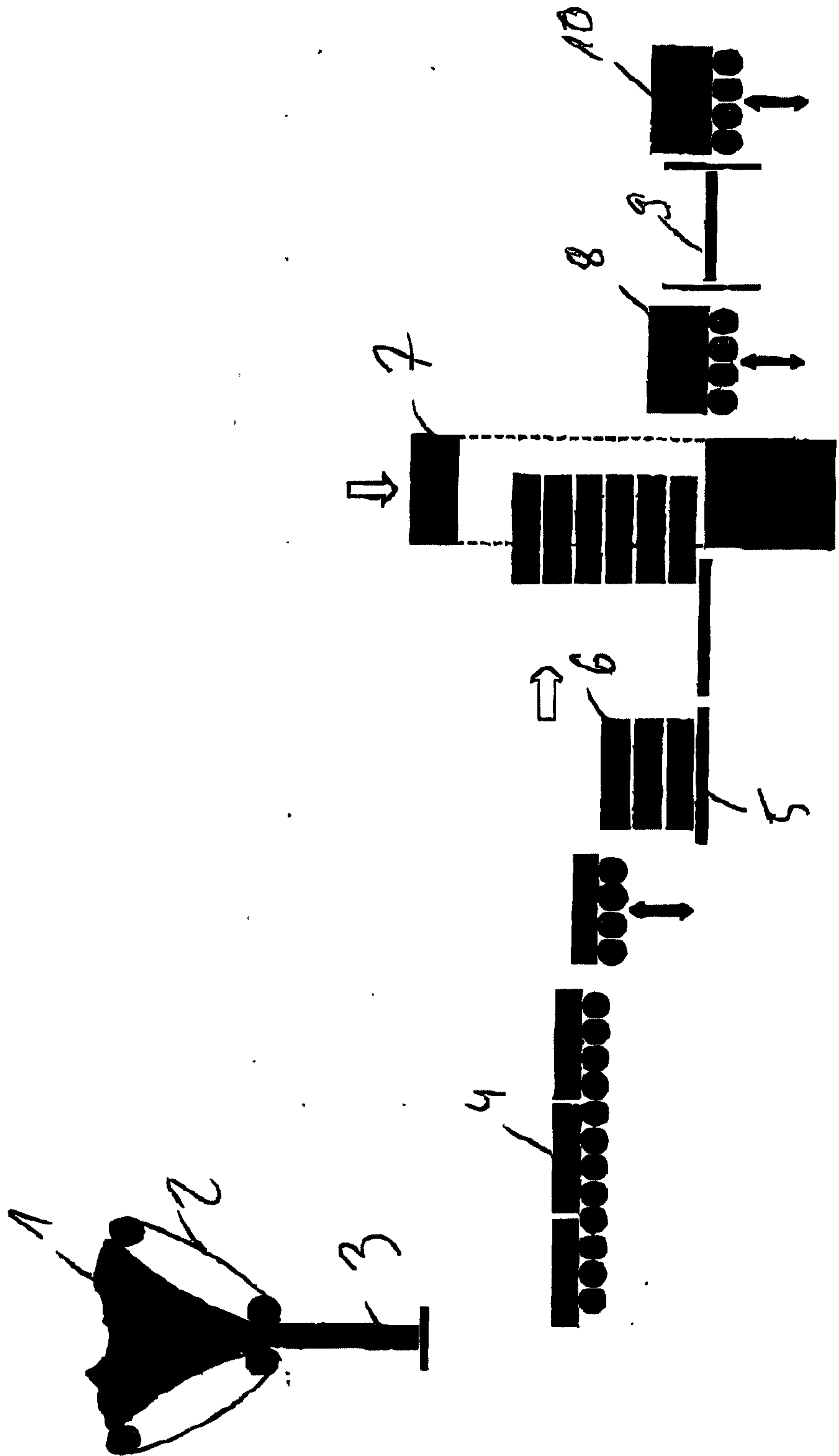


Figure 2

