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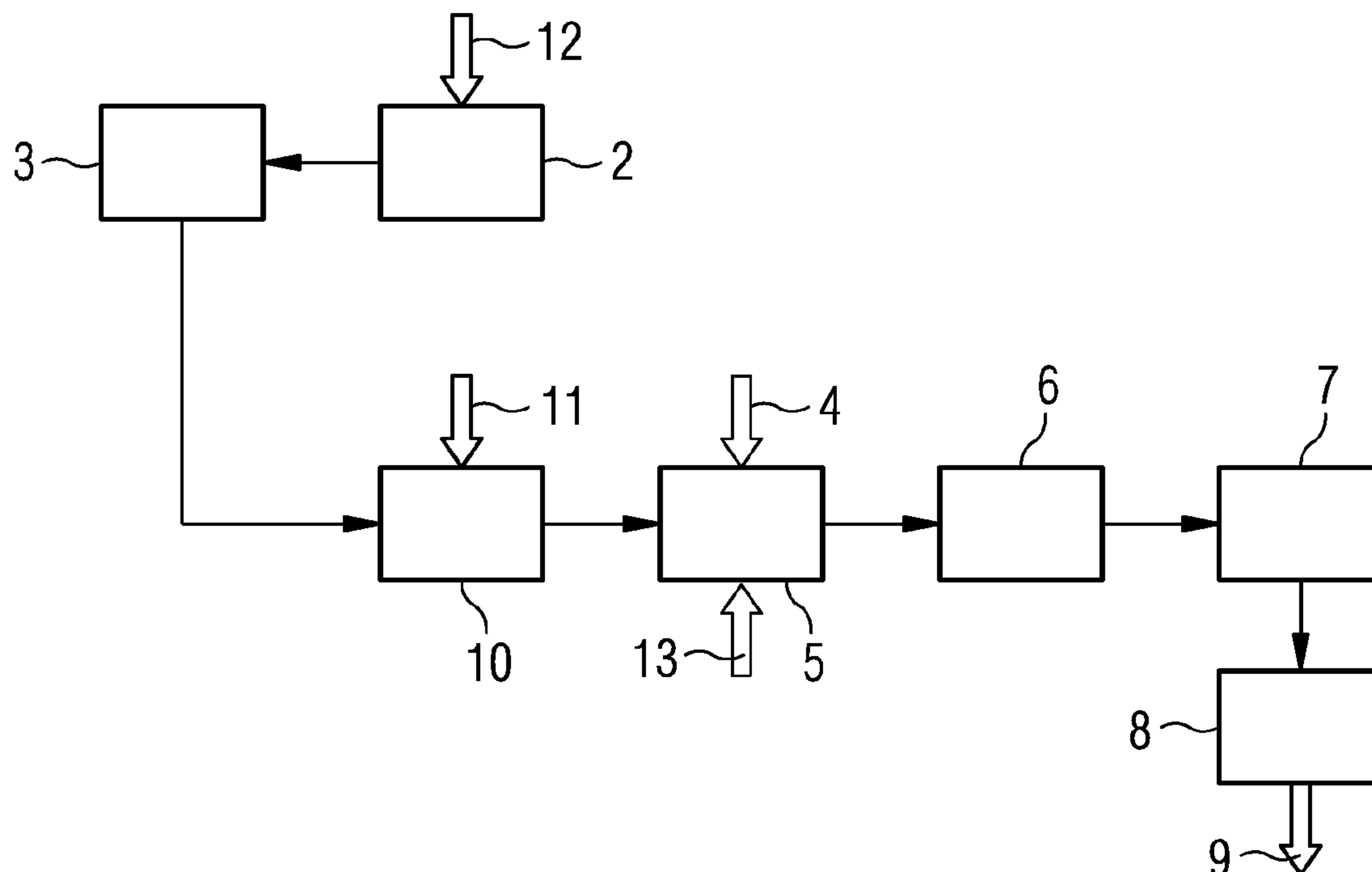
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(54) Titre : PROCEDE DE PRODUCTION DE PRODUITS PRESSES CONTENANT DES PARTICULES DE CARBONE  
(54) Title: METHOD FOR PRODUCING PRESSED ARTICLES CONTAINING COAL PARTICLES

**FIG 2**



(57) **Abrégé/Abstract:**

The invention relates to a method for producing pressed articles containing coal particles, to the pressed articles obtained in this way, and to the use of the pressed articles in methods for producing pig iron in a fixed bed or in methods for producing carbon carriers for methods for producing pig iron in a fixed bed. To this end, a partial amount of the coal particles to be processed into pressed articles is impregnated with a substance before the material to be processed into pressed articles is mixed with a binder system containing water and finally being processed into pressed articles.

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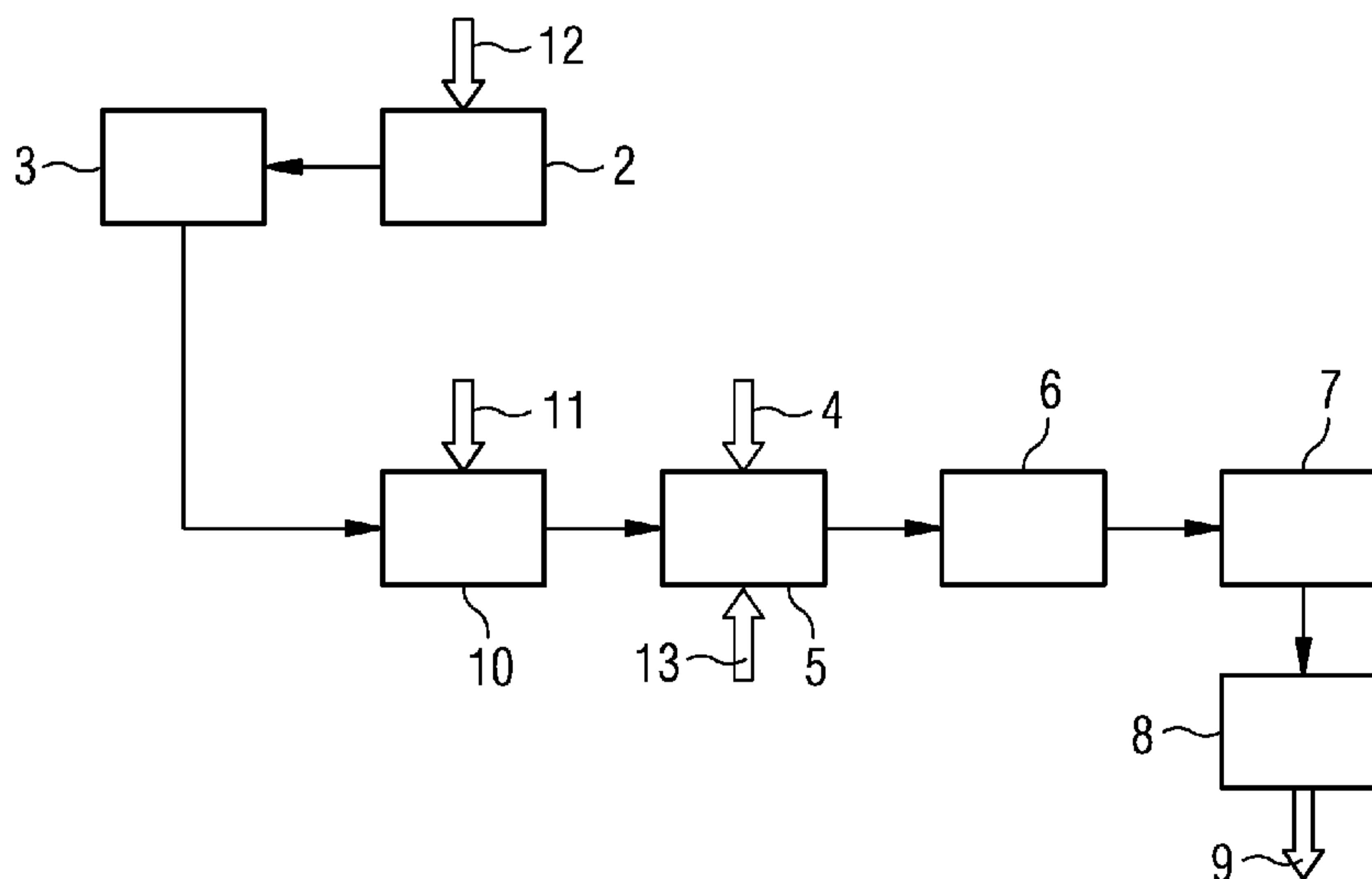
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[Fortsetzung auf der nächsten Seite]

(54) Title: METHOD FOR PRODUCING PRESSED ARTICLES CONTAINING COAL PARTICLES

(54) Bezeichnung : VERFAHREN ZUR HERSTELLUNG VON KOHLEPARTIKEL ENTHALTENDEN PRESSLINGEN

FIG 2



(57) Abstract: The invention relates to a method for producing pressed articles containing coal particles, to the pressed articles obtained in this way, and to the use of the pressed articles in methods for producing pig iron in a fixed bed or in methods for producing carbon carriers for methods for producing pig iron in a fixed bed. To this end, a partial amount of the coal particles to be processed into pressed articles is impregnated with a substance before the material to be processed into pressed articles is mixed with a binder system containing water and finally being processed into pressed articles.

(57) Zusammenfassung:

[Fortsetzung auf der nächsten Seite]

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— vor Ablauf der für Änderungen der Ansprüche geltenden Frist; Veröffentlichung wird wiederholt, falls Änderungen eingehen (Regel 48 Absatz 2 Buchstabe h)

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Die Erfindung betrifft ein Verfahren zur Herstellung von Kohlepartikel enthaltenden Presslingen, die dabei gewonnenen Presslinge sowie die Verwendung der Presslinge in Verfahren zur Roheisenerzeugung in einem Festbett oder in Verfahren zur Herstellung von Kohlenstoffträgern für Verfahren zur Roheisenerzeugung in einem Festbett. Dabei wird eine Teilmenge des zu Presslingen zu verarbeitenden Gutes Kohlepartikel mit einer Substanz imprägniert, bevor das zu Presslingen zu verarbeitende Gut mit einem Wasser enthaltenden Bindemittelsystem vermischt und schließlich zu Presslingen verarbeitet wird.

**Description****Description of the invention**

Method for producing pressed articles containing coal  
particles

**Technical field**

The invention relates to a method for producing pressed articles containing coal particles, to the pressed articles obtained thereby and to the use of the pressed articles in methods for producing pig iron in a fixed bed or in methods for producing carbon carriers for methods for producing pig iron in a fixed bed.

**Prior art**

Pressed articles containing coal particles, for example briquettes, used in methods for producing pig iron in a fixed bed, for example in melter gasifiers or in methods for producing carbon carriers for methods for producing pig iron in a fixed bed, for example coke production for blast furnaces, must have a certain shatter strength and compressive strength after discharge from the press. The shatter strength is required to ensure that the original size of the pressed articles is retained to the greatest extent possible during charging into a process irrespective of unavoidable falls, for example on transfer from one conveyor belt to another or on charging into a material bunker. The compressive strength is required to ensure that the original size of the pressed articles is retained after charging into a material bunker or a fixed-bed reactor despite a pressure exerted by superposed layers of material. These strength requirements are also subsumed under the term cold strength.

In addition to cold strength, the hot strength of pressed articles - particularly during use in thermal processes - is a criterion for their suitability for use. In the special case of the use of pressed articles containing fine-grained coal particles in methods for producing pig iron, such as for example in a melter gasifier or blast furnace, the term the hot strength refers a) to a strength of the semicoke or coke particles remaining after the pyrolysis of the pressed articles in a high-temperature zone and b) to a strength of these semicoke or coke particles following a chemical attack by a hot CO<sub>2</sub>-containing gas. A minimum level of hot strength makes it possible for the size of these particles to be largely retained following the conversion of the pressed articles by pyrolysis into semicoke or coke particles. In the case of methods for producing pig iron in a fixed bed, the development of undersized material from pressed articles or coke particles before charging into a fixed bed or within a fixed bed is undesirable because this causes a deterioration in the permeability of the fixed bed. In the special case of a method for producing pig iron, this concerns both the gas permeability and the drainage behavior of the fixed bed with respect to the liquid pig iron and the slag. If there is a deterioration in the permeability of the fixed bed, adverse effects are likely on its productivity, its specific energy requirement and its product quality.

It is known from WO 02/50219A1 to produce pressed articles with sufficient cold strength from fine-grained coal particles by means of a binder system comprising quicklime and molasses. This involves mixing fine-grained coal particles of fine coal and quicklime, leaving the mixture to rest for the purpose of progressing the slaking reaction with moisture from the coal particles, then mixing in molasses, kneading the mixture

obtained thereby and finally pressing pressed articles therefrom.

There are coals that exhibit an extremely high water-absorbing capacity, particularly characterized by a high inherent moisture content. However, for use in the production of pig iron, the moisture content of the pressed articles should not be too high, that is no more than 7% by weight. This is because, when using the pressed articles to produce pig iron or for producing carbon carriers for methods for producing pig iron, this moisture is a drain on energy, since the specific consumption of carbon carriers increases significantly with the moisture content of the pressed articles. Therefore, coals with a moisture content higher than this have to be dried before being processed into pressed articles. In addition to the unwetted pore volume that already exists in the undried coal, additional pore volume is produced by driving out water from cavities during the drying. The unwetted pore volume can absorb a corresponding amount of water or aqueous media. Obviously, the additional pore volume can also absorb water or an aqueous medium once again. Moreover, certain coals also have a tendency - particularly in the case of intensive drying - to generate additional pore volume as a result of grain damage. When drying a coal with a high water-absorbing capacity to an acceptable moisture content before the application of the method described in WO 02/50219A1 for producing pressed articles, a large additional pore volume is generated. Therefore, a dried coal particle sucks into its pores a considerable part of the molasses required for establishing bonding on the particle surface and which can be regarded as an aqueous solution. Therefore, it is not possible to obtain sufficient strength of the processed article for such coals with conventionally used additions of molasses of

$\leq 10\%$  by weight, based on the weight of the coal to be processed. In order nevertheless to be able to produce pressed articles with sufficient strength on the basis of molasses binders, it is necessary

- to dispense with the generation of unwetted pore volume by drying or
- to add as much additional molasses as taken up by the pore volume and which is, therefore, not available for bonding on the surface of the coal particles.

However, these measures are undesirable for reasons of process economy.

Even in the case of coals that are naturally less moist and which do not have to be dried to achieve a moisture content of the pressed articles of at most 7% by weight, a part of the molasses is sucked into pores of the coal particles. However, molasses contains components which act catalytically with regard to a reaction of carbon with hot  $\text{CO}_2$ -containing gas, whereby the extent of a reaction of solid carbon with  $\text{CO}_2$  according to the Boudouard reaction increases, particularly in the hot zones of a fixed bed used to produce pig iron at temperatures of  $> 800\text{--}1000^\circ\text{C}$ , depending on the pressure. Consequently, the hot strength of semicoke or coke particles obtained by pyrolysis from pressed articles treated with molasses decreases.

The use of bitumen as a binder proposed in W09901583A1 does not give rise to problems of this kind associated with molasses. However, the production of pressed articles with bitumen is associated with very high binder costs.

The use of an aqueous bitumen emulsion as a binder system proposed in AT005765U1 reduces bitumen consumption by up to

more than 50%. However, it has been found in practice that the charging coals must have moisture contents of significantly more than 5% by weight in order to ensure that stable pressed articles are produced when using bitumen emulsions of this kind. There is also the problem that pores present in the coal particles can absorb aqueous bitumen emulsion or extract water from the emulsion and hence destabilize it as a result of droplet coalescence before a largely uniform distribution of the emulsion within the material to be processed into pressed articles, and correspondingly uniform wetting of the particle surface by the emulsion, can take place. This reduces the effectiveness of the emulsion as a binder.

### **Summary of the invention**

#### **Technical object**

The object of the present invention is to provide a method for producing pressed articles with which these drawbacks of the prior art are overcome and pressed articles with sufficient with green and hot strength can be produced, even when using coal particles that have to be pre-dried, using a lower amount of a water-containing binder system compared to known methods.

#### **Technical solution**

This object is achieved by a method for producing a pressed article containing coal particles in which the coal particles are mixed with a water-containing binder system and the mixture obtained thereby is further processed into pressed articles by pressing, characterized in that, before mixing with the water-containing binder system, a partial amount of the coal particles is subjected to an impregnating step in which it is impregnated with a substance.

**Advantageous effect of the invention**

During the impregnation, either the substance penetrates the pores of the coal particles and, by filling the pore space, correspondingly prevents penetration of components of the aqueous binder system or the substance is deposited in the outlets of the pores on the coal particle surface, also known as pore necks, and, by this clogging of the pore necks, prevents penetration of components of the aqueous binder system into the pores. This avoids the case in which the aqueous binder system that is required on the coal particle surface for binding purposes is no longer able to fulfill these binding purposes after penetration into the pores. Correspondingly, the amount of aqueous binder system required is reduced compared to a method with which the aqueous binder system is able to penetrate the pores.

Preferably, before the impregnating step, the coal particles to be processed into pressed articles, or at least a partial amount of them, are subjected to a drying to a moisture content of less than 8% by weight, preferably to a moisture content of less than 7% by weight. A moisture content in a range of greater than/equal to 4% by weight to less than 8% by weight is particularly preferable, a moisture content in a range of greater than/equal to 5% by weight to less than 7% by weight is particularly preferable.

Apart from water, the aqueous binder system can contain one or more further components.

The impregnating step can consist of damping the coal particles with the substance, spraying the coal particles with the substance, incorporating the substance in a moving packed

bed of the coal particles or incorporating the substance in a fluidized bed of the coal particles.

The partial amount of the coal particles which is subjected to an impregnating step before the mixing with the water-containing binder system and the coal particles which are not subjected to an impregnating step can be the same material - with respect to the sort of coal and average particle size. According to another variant, the partial amount of the coal particles which is subjected to an impregnating step before mixing with the water-containing binder system can be of the same coal sort as the coal particles that are not subjected to an impregnating step, but have a different average particle size than the coal particles that are not subjected to an impregnating step.

It is not the total amount of the coal particles to be processed into pressed articles that is impregnated but only a partial amount.

According to another variant, the partial amount of the coal particles which is subjected to an impregnating step before mixing with the water-containing binder system can be another sort of coal than the coal particles which are not subjected to an impregnating step. In this case, the partial amount of the coal particles to be impregnated and the coal particles which are not to be impregnated can have the same or different average particle size.

If partial amounts of the coal particles from which pressed articles are to be produced differ in that they belong to different coal sorts and pressed articles with different values for cold strength or hot strength would be produced

from the different coal sorts, it is advantageous to impregnate the partial amount which would supply pressed articles with more unfavorable values for cold strength or hot strength.

If the coal particles from which pressed articles are to be produced belong to a single coal sort but differ in that they have different average particle sizes, it can be advantageous to impregnate a partial amount with the largest possible average particle size. Since the specific surface for coal particles with a larger average particle size is lower than for coal particles with a smaller average particle size, in this way, a given amount of impregnating agent can be used to impregnate a greater part in mass of the coal particles to be processed into pressed articles than is the case with the impregnation of coal particles with a smaller average particle size.

If the coal particles from which pressed articles are to be produced belong to a single coal sort but differ in that they have different average particle sizes, it can also be advantageous to impregnate a partial amount with the smallest possible average particle size. Since the specific surface for coal particles with larger average particle size is smaller than for coal particles with a smaller average particle size, with a given part of a mass to be impregnated, more surface is impregnated than when using a partial amount with a larger average particle size. This has the advantage that, for example, reactions with hot CO<sub>2</sub>-containing gas, which take place over the surface of the coal particles, are more extensively influenced by the impregnation since more surface is impregnated.

If a partial amount of the coal particles from which pressed articles are to be produced exerts a negative influence on the cold strength or hot strength of the pressed articles compared to pressed articles produced without this partial amount, it is advantageous to impregnate this partial amount. This enables its negative influence on the properties of the pressed articles to be reduced.

Following the performance of the impregnating step according to the invention of a partial amount of the coal particles, the impregnated partial amount of the coal particles is combined with the non-impregnated coal particles and the combined coal particles are further processed into pressed articles.

The combination of the impregnated partial amount of the coal particles with the non-impregnated coal particles can take place in a combination step in which only a combination and optionally mixing takes place. In this case, the further steps for producing the pressed articles, especially the mixing with a water-containing binder system, are performed with the product of the combination. The combination of the impregnated partial amount of the coal particles with the non-impregnated coal particles can also take place during the mixing with a water-containing binder system.

The substance used for the impregnation is preferably used as a liquid or by means of a liquid for impregnation. A liquid denotes, for example, substances which are liquid at the prevailing temperature during the impregnating step.

Impregnation by means of a liquid denotes, for example, impregnation with substances, which, although they are not actually liquid at the conditions prevailing during the impregnating step, are emulsified or suspended in a liquid.

Compared to the use of solid substances, this improves or even enables penetration into pores or the clogging of pore necks. In order to ensure that a substance used during an impregnating step remains liquid during the impregnating step, the coal particles to be impregnated are preferably heated to a temperature at which the substance is liquid.

According to one embodiment, the substance with which the partial amount of the coal particles is impregnated in the impregnating step is water.

Then, in the impregnating step, water is sucked into the pores, which as a result no longer show any tendency to absorb components of the aqueous binder system fed to the coal particles after the impregnating step. As a result, components which, in the case of previous methods, were sucked into pores and consequently became ineffective for the binding of the pressed articles can make a contribution to the binding of the pressed articles.

Limiting the proportion of pressed articles impregnated with water in a charging mixture for a pig iron production process in combination with carbon carriers which have a lower moisture content than these pressed articles enables the amount of water introduced into the pig iron production process to be limited to an acceptable amount.

According to another embodiment, the substance with which the partial amount of the coal particles is impregnated in the impregnating step is a water-insoluble and/or water-repellent substance.

If the pores are filled with a substance of this kind in the impregnating step and the pore walls are thereby coated with substances of this kind, the tendency of the pores to absorb

components of the aqueous binder system decreases. If the outlets of the pores on the coal particle surface are closed by substances of this kind, no components of the aqueous binder system are able to penetrate the pores. As a result, components which were previously sucked into pores and consequently became ineffective for the binding of the pressed articles can make a contribution to the binding of the pressed articles.

The water-insoluble and/or water-repellent substance preferably belongs to the group of substances comprising waxes, organic coking-plant or refinery products and plastics or plastics scrap. It can also be used oil. It can also be bitumen. These substances are usually available in large amounts at low cost.

In this case, the impregnating step is advantageously performed at a temperature at which the water-insoluble and/or water-repellent substance is liquid, particularly viscous. In this context, liquids are considered to be viscous if their viscosity is at least 1 Pas and at the most 100 Pas, for example 10 Pas. Under these conditions, the substance is dispersed on the surface of the coal particles and penetrates the outlets of the pores but hardly penetrates the interior of the pores at all. This keeps the consumption of the water-insoluble and/or water-repellent substance in the impregnating step low. Advantageously, the water-insoluble and/or water-repellent substance solidifies in the outlets of the pores on the coal particle surface on cooling.

According to another embodiment, the substance with which the partial amount of the coal particles is impregnated in the impregnating step is an aqueous solution of a substance or a

mixture of substances. For example, it is molasses, which is an aqueous solution of a mixture of carbohydrates and other natural substances.

In principle, dissolved substances of all kinds that improve the hot strength and cold strength of the pressed articles can be used, for example starch or lignin lyes from spent liquors from pulp production.

It is preferred to use solutions of substances or mixtures of substances which are converted to water-insoluble substances by heat treatment and/or chemical reaction. As a result, the effects induced by these substances or mixtures of substance are not lessened by their being dissolved in the water of the water-containing binder system and being washed out of the pores.

According to another embodiment, the substance with which the partial amount of the coal particles is impregnated in the impregnating step is an aqueous suspension of solid colloids, wherein the solid substance has water-repellent properties. Examples are suspensions of colloidal talc, graphite or waxes in water. If the solid substances are deposited in the pores or in the pore necks, it is more difficult for water-containing binder systems to enter due to the high surface tension of the water-repellent solid substances.

According to a further embodiment, the substance with which the partial amount of the coal particles is impregnated in the impregnating step is an emulsion containing water on the one hand and carbon-containing substances on the other, such as for example bitumens, crude tars obtained from hard coal, pitches, waxes or oils. When emulsions of this kind penetrate the pores, the carbon-containing substances are deposited in thin layers on the pore surface. During pyrolysis, carbon

layers are produced from these thin layers. These reduce the reactivity of the pressed articles with respect to hot CO<sub>2</sub>-containing gases compared to an embodiment in which no thin layers of the substances are deposited in the pores. An effect of this kind also occurs if the substance with which the partial amount of the coal particles is impregnated in the impregnating step is not an emulsion, for example if the substance is bitumen.

The occurrence of an effect of this kind is due to the fact that the carbon layers produced from the substances contain little or no substances that react catalytically with respect to the reaction with hot CO<sub>2</sub>-containing gases. By contrast, the coal particles or material that is to be processed into pressed articles contain catalytically acting compounds, for example iron or alkalis. Correspondingly, the reactivity of a pressed article of which the surface and pores are covered with a carbon layer created from the substances is less than that of a pressed article without a carbon layer of this kind.

When using coal particles which require pre-drying before being processed into pressed articles, it is of advantage for commercial reasons not to pursue the drying to a moisture content much below 5% by weight, that is to a moisture content of at most 4% by weight. As a result, the creation of additional pore volume as a consequence of the drying limited and correspondingly less substance is absorbed in the impregnating step. Correspondingly, less substance is used in the impregnating step. In addition, less expenditure in terms of equipment and energy is required for the drying.

The lower limit of the amount of substance added in the impregnating step, known as impregnating agent, is 0.3% by weight, preferably 0.5% by weight, particularly preferably 1%

by weight, the upper limit is 5% by weight, preferably 3% by weight, particularly preferably 2% by weight, based on the weight of the partial amount to be impregnated of the material to be processed into pressed articles, that is of the partial amount to be impregnated of the coal particles. The addition of more than 5% by weight of impregnating agent is not economically advisable. If less than 0.3% by weight of impregnating agent is added, impregnation is no longer effective.

According to one embodiment of the method according to the invention, the binder system contains molasses and quicklime or hydrated lime. It can also consist of these components. According to further embodiments, the binder system contains molasses in combination with strong inorganic acids, such as, for example phosphoric acid, sulfuric acid or nitric acid.

According to one embodiment of the method according to the invention, the binder system contains an emulsion of bitumen in water. It can also consist of an emulsion of this kind.

According to further embodiments, the binder system contains products from spent liquors of pulp production, starches, cellulose, beet chips, waste paper pulp, wood pulp or also long-chain polyelectrolytes such as, for example, carboxy methylcellulose.

Since binder systems containing quicklime or hydrated lime have the drawback that quicklime  $\text{CaO}$  and hydrated lime  $\text{Ca(OH)}_2$  increase the reactivity of the pressed articles with respect to hot  $\text{CO}_2$ -containing gases as a result of catalytic effectiveness, the embodiments without quicklime or hydrated

lime have the advantage of providing pressed articles with comparatively lower reactivity.

According to one embodiment of the method according to the invention, iron- or iron-oxide-containing particles are also processed in a mixture with the coal particles into pressed articles.

According to a special refinement of the method according to the invention, the pressed articles are subjected to heat treatment after the pressing. The heat treatment is performed at a higher temperature than the pressing. The heat treatment causes drying and/or hardening of the pressed articles. The heat treatment can be performed at temperatures of preferably  $\geq 250^{\circ}\text{C}$  and  $\leq 350^{\circ}\text{C}$  at which irreversible chemical processes can transform binder components. For example, water-soluble binder components can be transformed into water-insoluble compounds.

The compounds produced with such transformations can make a contribution to the strength of the pressed articles.

In the case of a binder system containing molasses, for example, transformation of molasses by caramelization takes place.

According to a special refinement of the method according to the invention, at least the partial amount of the coal particles which was subjected to an impregnating step is subjected to heat treatment after the impregnating step before mixing with the water-containing binder system.

The heat treatment can be performed by subjecting the impregnated partial amount to heat treatment separately and combining it with the non-impregnated coal particles after the

heat treatment or the impregnated partial amount can be combined with the non-impregnated coal particles before the heat treatment of the coal particles.

The heat treatment causes drying. If there are solutions or emulsions in the pores, the heat treatment also causes concentration of the solutions, suspensions or emulsions and correspondingly a coating of the pore walls with dissolved, suspended or emulsified components. In addition to the aqueous binder system to be subsequently added, these can make a contribution to increased hot strength and cold strength of the pressed articles. The heat treatment can also effect the transformation of the coating of the pore walls, initially produced as a result of the heat treatment, into water-insoluble compounds, or into compounds lowering the reactivity of the coal particles with respect to hot CO<sub>2</sub>-containing gases. The maximum temperature of the heat treatment is restricted by the pyrolysis of the coal particles and is 350°C. The lower limit for the temperature in this heat treatment is 150°C.

If the same water-containing emulsion is used for the impregnation as is used as the water-containing binder system, the amount added in the impregnating step is less than the amount of water-containing binder system added in the subsequent mixing. For example, when using a bitumen-in-water emulsion in the impregnating step and as the binder system, 2-3% by weight is added in the impregnating step, while 7-10% by weight is added later as the binder system.

The same applies if the same aqueous solution of a substance or of a mixture of substances is used for the impregnation as that used as the water-containing binder system. For example, when using molasses in the impregnating step and as the binder system, 3 to 5% by weight is added in the impregnating step, while 6 to 8% by weight are added later as the binder system.

In such cases, the limits of the specified ranges are also included. In these cases, heat treatment is necessary after the addition in the impregnating step in order to remove the carrier liquid, water, to the extent that the emulsified substances or the dissolved substances settle in the pores or the pore necks. As a result, the pores are occupied or the pore necks become clogged. Overall, therefore, less water-containing binder system is required to produce the pressed articles than in the case of production without an impregnating step.

Following mixing with a water-containing binder system, processing into pressed articles can be performed by known methods, for example as described in WO 02/50219A1 or AT005765U1 or by any method suitable for processing coal particles with a water-containing binder system into pressed articles.

The addition of water-containing binder systems which, according to the invention, is only performed after the impregnating step of a partial amount of the coal particles with a water-insoluble and/or water-repellent substance during the production of pressed articles reduces the costs of the method compared to conventional methods, such as, for example, according to WO02/50219A1. The avoidance of water absorption by the coal during the production of pressed articles with water-containing binder systems on the one hand reduces the specific coal consumption in pig iron production methods in which the pressed articles or coke obtained from them are used, since less water from the binder system is present in the pressed article and correspondingly less energy has to be expended for vaporizing said water. On the other hand, when the method according to the invention is used, it is possible

to dispense with the necessity to after-dry the pressed articles that occurs in conventional methods for producing pressed articles due to the water absorption from the binder system, or it is possible to reduce the degree of drying thus resulting in energy saving. Since it is correspondingly possible to dispense with the installation or operation of apparatus for after-drying or it is possible to reduce the dimensions of the apparatuses and the effort involved in their operation, this is synonymous with reduced operating costs and reduced investment costs.

Depending upon the type of substance used for the impregnation, lessening of the  $\text{CO}_2$  reactivity of the semicoke produced after the pyrolysis of the pressed articles in a melter gasifier or of the coke obtained from pressed articles may be obtained as an additional advantageous effect. Low  $\text{CO}_2$  reactivity is desirable during the operation of a melter gasifier in order that the semicoke in the fixed bed of the melter gasifier or the coke in the fixed bed of a blast furnace remains stable from the charging onto the bed surface to the achievement of the direct gasification zone in the region of the oxygen nozzles or the tuyeres and as a result promotes the permeability of the fixed bed with respect to the gas distribution and drainage of molten phases. The lessening of the  $\text{CO}_2$  reactivity of the semicoke or of the coke is achieved by the inner surface of the pores of the coal particles in the pressed article originating from the impregnated partial amount of the coal particles no longer being able to be coated by the impregnation by a binder containing reactivity-promoting substances. For example, the binder component molasses contains alkalis as reactivity-promoting substances. If coating of the inner surface of the pores with molasses is avoided by impregnation with substances

containing bitumens or waxes, for example, CO<sub>2</sub>-reactivity is therefore lowered compared to semicoke or coke obtained by means of a method without an impregnating step.

A lower proportion of undersized coke is often added to the charging coal in the COREX® or FINEX® methods for producing pig iron in a fixed bed of a melter gasifier in order to improve the permeability of the fixed bed. When using pressed articles produced according to the invention or coke produced therefrom, softening of the semicoke or coke particles is inhibited by hot CO<sub>2</sub> and hence disintegration of the particles is counteracted. Pressed articles produced according to the invention have namely been found to have improved thermomechanical stability of the semicoke compared to pressed articles produced in the conventional way. Thermomechanical stability relates, therefore, to the aspect of the hot strength, which concerns the strength of the semicoke or coke particles remaining after pyrolysis of the pressed articles in a high-temperature zone. Thermomechanical stability relates to a test method in which the pressed articles are exposed to a thermal shock procedure and the semicoke obtained thereby is subjected to tumbling. The improved thermomechanical stability is manifested by the fact that the proportion of large-size material in the tumbled semicoke is increased compared to conventionally produced pressed articles due to the impregnation according to the invention.

A packed fixed bed of pressed articles produced according to the invention from semicoke derived by pyrolysis enables much better gas permeability and better drainage behavior of the fixed bed than according to the prior art. The improvement in the reactivity properties of the semicoke therefore enables

the reduction or even avoidance of the addition of coke to the COREX® or FINEX® charging coal.

In the field of coking plant technology, it is known that increasing the bulk density of the charging coal improves the quality of the coke produced therefrom. The use of many charging coals for producing metallurgical coke is made possible in the first place by compacting the charging coal. Therefore, in addition to tamping coking plants, method variants for coking plants in loose-fill operation were developed providing briquetting or partial briquetting of the charging coals. However, from today's viewpoint, briquetting with bituminous binders is problematic for commercial reasons, hot briquetting or briquetting with binders originating from hard coal tar is problematic for health reasons and briquetting with molasses or comparable binders is problematic due to the introduction of undesired substances into the coke.

The method according to the invention for producing pressed articles makes it possible to reduce binder consumption or to mitigate the harmful effects of reactivity-promoting binder components, even when producing coke using articles pressed from charge materials.

The pressed articles can, for example, be briquettes or compressed strips from compacting.

The pressed articles contain up to 97% by weight of coal particles and up to 15% by weight of components of a binder system and, based on the weight of the coal particles to be processed into pressed articles, water-insoluble and/or water-repellent substances, or solid substances with water-repellent properties, in an amount of which the lower limit is 0.5% by

weight, preferably 1% by weight, and the upper limit is 5% by weight, preferably 3% by weight, particularly preferably 2% by weight.

Here, the 15% by weight of the components of a binder system should be understood as meaning water is not included as a component of the binder system - therefore, the 15% by weight relate to the non-aqueous components of the binder system.

According to one embodiment, the pressed article also contains iron- or iron-oxide-containing particles. Particles of this kind can, for example, originate from dusts or slurries occurring in the production of pig iron or steel.

#### **Description of embodiments**

Table 1 shows the evaluation of tests for the production of pressed articles with respect to the shatter strength (SS) and the point compressive strength (PCS) of the pressed articles in the context of a series of tests. For this, the pressed articles according to the method according to the invention were produced with impregnation of a partial amount of the coal particles.

The pressed articles according to the prior art were produced in such a way that all coal particles were impregnated with water - with the addition of 3% by weight of water over a period of one minute.

The pressed articles were briquettes.

The shatter strengths of green pressed articles and pressed articles produced according to the invention and green pressed articles and pressed articles produced according to the prior art - with the same charge materials in each case using 12% by

mass of molasses and under otherwise identical conditions - were in the same order of magnitude for both green pressed articles and for air-dried and thermally dried pressed articles.

The water-containing binder system used was a system consisting of molasses and quicklime. The molasses itself had a water content of 20% by mass. The following commercially available molasses was used in the binder system: sugarcane molasses from the company Tate & Lyle with a total sugar content of 51%. The quicklime in the binder system was white pulverized quicklime from the company Walhalla Kalk. For the impregnation, bitumen was used as the impregnating agent. The bitumen used was Mexphalte 55 from the company Shell.

The impregnating agent bitumen was mixed-in in a FM130D plough share mixer from the company Lödige; the other mixtures were produced in a R08 W batch mixer from the company Eirich.

The kneading machine from the company Koeppern used for the kneading processes consisted of a vertical cylindrical container through which a central rotating shaft with kneader blades is guided.

The green pressed articles were produced by means of a type 52/10 test roller press from the company Koeppern. The cushion-shaped format chosen for the green pressed articles had a nominal volume 20 cm<sup>3</sup>. The material to be pressed was charged by means of gravity feeders. The test roller press was used to produce strips comprising a plurality of green pressed articles. These strips contained green pressed articles in both the edge region of the strips and the central region of

the strips.

In order to obtain individual green pressed articles or individual pressed articles for the determination of the shatter strength or the point compressive strength, the strips were broken along dividing line between the individual green pressed articles. As a rule, the strips broke into individual green pressed articles on discharge from the test roller press.

Following the kneading process in the kneading machine, the kneaded mixtures as material to be pressed were subjected to pressing in the test roller press in order to produce green pressed articles.

The green pressed articles obtained thereby were still soft - which is indicated in technical jargon by the prefix "green" - and were subjected to hardening in order to obtain the finished pressed article. This hardening can for example be performed by at least partially drying by storage in the air and/or thermal treatment.

After the pressing, individual green pressed articles, in technical jargon green, were each immediately tested with respect to shatter strength (SS) and point compressive strength (PCS). The results of these tests are shown in the "immediate" columns for PCS and SS. The measurements of shatter strength and point compressive strength were each repeated after 1 h hardening in the air and after 24 h hardening in the air. The results of these tests are shown in the "1 h" and "24 h" columns.

In the drop-shatter test (with reference to ASTM D440) to determine the shatter strength, a specimen weighing 2 kg of green pressed articles or pressed articles which had been

hardened by drying in the air or by thermal drying was dropped four times through a downpipe from a height of 5 m into a collection receptacle with a base formed from a solid steel plate. The downpipe had a diameter of 200 mm and the collection receptacle a diameter of 260 mm. The steel plate was 12 mm thick. The evaluation of the drop-shatter test by screen analysis was performed after the second and fourth drop. The numerical values for shatter strength SS in Table 1 each indicate the proportion of grain size fraction  $> 20$  mm after four drops.

The point compressive strength was determined on a type 469 test machine from the company ERICHSEN. During this test procedure, individual green pressed articles or pressed articles which had been hardened by drying in the air or by thermal drying were clamped between two platens with the lower one being coupled to a force transducer and the upper one being continuously loaded by means of a spindle drive for applying a creeping swelling pressure load. The lower platen was formed from a round plate with an 80 mm diameter and the upper by a horizontal round iron bar with a 10 mm diameter. The rate of advance for the upper platen was 8 mm/min. The point compressive strength PCS was recorded as the maximum load-bearing capacity of a green or hardened pressed article before breaking - the entries in Table 1 show the average point compressive strength at break in newtons. In each case, six green pressed articles or pressed articles from the central region and six green pressed articles or pressed articles from the edge region of the strips produced in the test roller press were tested. The mean values of the data produced in these tests were calculated, wherein in each case the minimum and maximum values were ignored. The mean values are shown in Table 1.

Table 1

Test number	PCS [N] immediate	PCS [N] 1 h	PCS [N] 24 h	SS immediate	SS 1 h
1	46	84	149	70	78
2	104	166	252	73	67

In test 1 according to the prior art, a mixture of 70% by weight Ensham coal with an average particle size, d50, of 0.95 mm together with 30% by weight Blackwater coal with an average particle size d50 von 0.8 -1.0 mm was used as coal particles to be processed into pressed articles.

Blackwater coal originates from the company BHP Billiton in Queensland, Australia.

Ensham coal originates from the company Ensham Resources in Queensland, Australia.

This material to be processed into pressed articles was processed into pressed articles as shown below in Figure 1 for coal 1. The molasses in the water-containing binder system was used in an amount of 12% by weight, based on the weight of the material to be processed into pressed articles. The molasses used itself had a water content of 20% by weight water. In addition to molasses, the water-containing binder system also comprises 2.5% by weight quicklime, based on the weight of the material to be processed into pressed articles. The point compressive strength and shatter strength at different times are shown in the first data column in Table 1.

In test 2 according to the inventive method, the same material to be processed into pressed articles was used. However, the Ensham coal used was impregnated with bitumen. The bitumen used was Shell Special Bitumen A with a softening point of 85°C. The amount of bitumen used was 2.1% by weight, based on the weight of the material to be processed into pressed articles or 3% by weight based on the Ensham coal to be impregnated. The temperature of the coal before mixing in the bitumen was 108°C. After the impregnation, the impregnated Ensham coal was combined with the Blackwater coal. After combination, the processing was performed similarly to test 1 but the molasses in the water-containing binder system was used in an amount of 8% by weight, based on the weight of the material to be processed into pressed articles. The molasses used itself had a water content of 20% by weight. In addition to molasses, the water-containing binder system also comprised 2% by weight of quicklime, based on the weight of the material to be processed into pressed articles. After the addition of quicklime in each case a further 2% water, based on the weight of the coal particles to be processed into pressed articles, was mixed-in in order to endow the quicklime with the moisture required for its reaction.

It may be recognized that pressed articles produced according to the invention have a higher point compressive strength than pressed articles produced according to the prior art, while their shatter strength is comparable to the shatter strength of pressed articles produced according to the prior art.

The partial amount of the coal particles to be impregnated can also be subjected to two or more impregnating steps.

#### **Brief description of the drawings**

The following describes the method according to the invention with reference to the block diagrams shown in Figures 1 to 3.

Figure 1 shows a conventional method for producing pressed articles without an impregnating step.

Figure 2 shows a method according to the invention for producing pressed articles with an impregnating step, wherein two coal sorts are used.

Figure 3 shows a method according to the invention for producing pressed articles with the impregnating step, wherein only one coal sort is used.

According to Figure 1, the coal 1 to be processed into pressed articles, in this case briquettes, is subjected to drying 2 and then brought to a desired grain size by granulation 3. Then, a water-containing binder system 4, in this case molasses, is added to the coal particles obtained thereby, optionally with the addition of solid, fine-particle binder components such as hydrated lime or quicklime, while mixing 5, wherein the mixing 5 can be performed in one or more stages. The mixture obtained thereby is subjected to kneading 6 and pressing 7. The product 9 obtained after hardening 8 is the briquette.

The method according to the invention as shown in Figure 2 differs from the method shown in Figure 1 in that a partial amount A of the coal particles 12 used to produce the pressed articles are subjected to an impregnating step 10 in which they are impregnated with a substance 11, the impregnating agent. After this impregnating step 10, the mixing 5 with the water-containing binder system 4 and with a partial amount B

of the coal particles 13 used to produce the pressed articles and the further processing of the mixture obtained thereby takes place in a way corresponding to Figure 1. The coal particles used to produce the pressed articles therefore comprise both the partial amount A 12 and the partial amount B 13. Partial amount A 12 and partial amount B 13 belong to different coal sorts.

Unlike the case in Figure 2, in Figure 3 the partial amounts A 12 and B 13 of the coal particles used to produce the pressed articles belong to the same coal sort. A coal 1 to be processed is subjected to drying 2 and then brought to a desired grain size by granulation 3. The coal particles obtained thereby are subjected to screening 14. As partial amount A of the coal particles 12 used to produce the pressed articles, the coarse-grained fraction obtained thereby is subjected to an impregnating step 10 in which they are impregnated with a substance 11, the impregnating agent. After this impregnating step 10, the mixing 5 with the water-containing binder system 4 and with a partial amount B of the coal particles 13 used to produce the pressed articles and the further processing of the mixture obtained thereby takes place in a way corresponding to Figure 1. The partial amount B of the coal particles 13 used to produce the pressed articles is the fine-grained fraction obtained during the screening 14.

After the impregnating step 10, heat treatment 12 can be performed before the mixing with the water-containing binder system 4.

Generally, during the production of pressed articles according to the present invention, the addition of the water-containing binder system molasses/quicklime to material to be processed

into pressed articles can take place such that molasses and quicklime are added simultaneously or such that quicklime and molasses are added consecutively.

Here, with the use of the impregnating agent bitumen, it is preferable that first a partial amount of the molasses intended for the production of the pressed articles is added and then mixed and then quicklime is added. After the mixture obtained thereby has been allowed to rest, the remaining amount of the molasses intended for the production of the pressed articles is added. The partial amount and remaining amount together provide the molasses intended for the production of the pressed articles. The advantage of this procedure is that kneading of the quicklime into soft impregnating agent during the mixing of the material to be processed into pressed articles with the water-containing binder system is avoided or reduced.

The addition of molasses, which itself contains water, before the addition of quicklime, enables the quicklime also to use moisture from the molasses for its reactions.

Up to a half, preferably up to a third, of the molasses can be added before the quicklime.

**List of reference characters**

- 1 Coal
- 2 Drying
- 3 Granulation
- 4 Water-containing binder system
- 5 Mixing
- 6 Kneading
- 7 Pressing
- 8 Hardening
- 9 Product
- 10 Impregnating step
- 11 Substance (impregnating agent)
- 12 Partial amount A of the for producing the pressed articles
- 13 Partial amount B of the for producing the pressed articles
- 14 Screening

## **Citations**

### **Patent literature**

WO02/50219A1

WO9901583A1

AT005765U1

**Claims**

1) A method for producing a pressed article containing coal particles, in which the coal particles are mixed with a water-containing binder system and the mixture obtained thereby is further processed into pressed articles by pressing, characterized in that before mixing with the water-containing binder system a partial amount of the coal particles is subjected to an impregnating step in which it is impregnated with a substance.

wherein the lower limit of the amount of substance added in the impregnating step is 0.3% by weight based on the weight of the coal particles to be processed into pressed articles.

2) The method as claimed in claim 1, characterized in that the impregnating step consists of damping the coal particles with the substance, spraying the coal particles with the substance, incorporating the substance in a moving packed bed of the coal particles or incorporating the substance in a fluidized bed of the coal particles.

3) The method as claimed in any one of the preceding claims, characterized in that the substance with which the coal particles are impregnated in the impregnating step is water.

4) The method as claimed in any one of claims 1-2, characterized in that the substance with which the coal particles are impregnated in the impregnating step is a water-insoluble and/or water-repellent substance.

5) The method as claimed in any one of claims 1-2, characterized in that the substance with which the coal

particles are impregnated in the impregnating step is an aqueous solution of a substance or a mixture of substances.

6) The method as claimed in any one of claims 1-2, characterized in that the substance with which the coal particles are impregnated in the impregnating step is an aqueous suspension of solid colloids, wherein the solid substance has water-repellent properties.

7) The method as claimed in any one of claims 1-2, characterized in that the substance with which the coal particles are impregnated in the impregnating step is an emulsion containing water on the one hand and carbon-containing substances on the other.

8) The method as claimed in any one of the preceding claims, characterized in that the upper limit of the amount of substance added in the impregnating step is 5% by weight, preferably 3% by weight, particularly preferably 2% by weight, based on the weight of the coal particles to be processed into pressed articles.

9) The method as claimed in any one of the preceding claims, characterized in that the binder system contains molasses and quicklime or hydrated lime.

10) The method as claimed in any one of the preceding claims, characterized in that the binder system contains an emulsion of bitumen in water.

11) The method as claimed in any one of the preceding claims, characterized in that iron- or iron-oxide-containing-

containing particles are also processed in a mixture with the coal particles.

12) The method as claimed in any one of the preceding claims, characterized in that the pressed article is subjected to heat treatment after the pressing.

13) The method as claimed in any one of the preceding claims, characterized in that at least the partial amount of the coal particles that was subjected to an impregnating step is subjected to heat treatment after the impregnating step before mixing with the water-containing binder system.

14) A pressed article, containing up to 97% by weight of coal particles and up to 15% by weight of components of a binder system,

characterized in that, based on the weight of the coal particles to be processed into pressed articles, it contains water-insoluble and/or water-repellent substances, or solid substances with water-repellent properties, in an amount of which the lower limit is 0.3% by weight, preferably 0.5% by weight, particularly preferably 1% by weight, and the upper limit is 5% by weight, preferably 3% by weight, particularly preferably 2% by weight.

15) The pressed article as claimed in claim 14, characterized in that the water-insoluble and/or water-repellent substance belongs to the group of substances consisting of waxes, organic coking-plant or refinery products and plastics or plastics scrap and used oil.

16) The pressed article as claimed in any one of claims 14 and 15, characterized in that the pressed article also contains iron- or iron-oxide-containing particles.

17) The use of a pressed article as claimed in any one of claims 14 to 16 as a carbon carrier in a process for producing pig iron in a fixed bed or in a process for producing carbon carriers for a process for producing pig iron in a fixed bed.

FIG 1

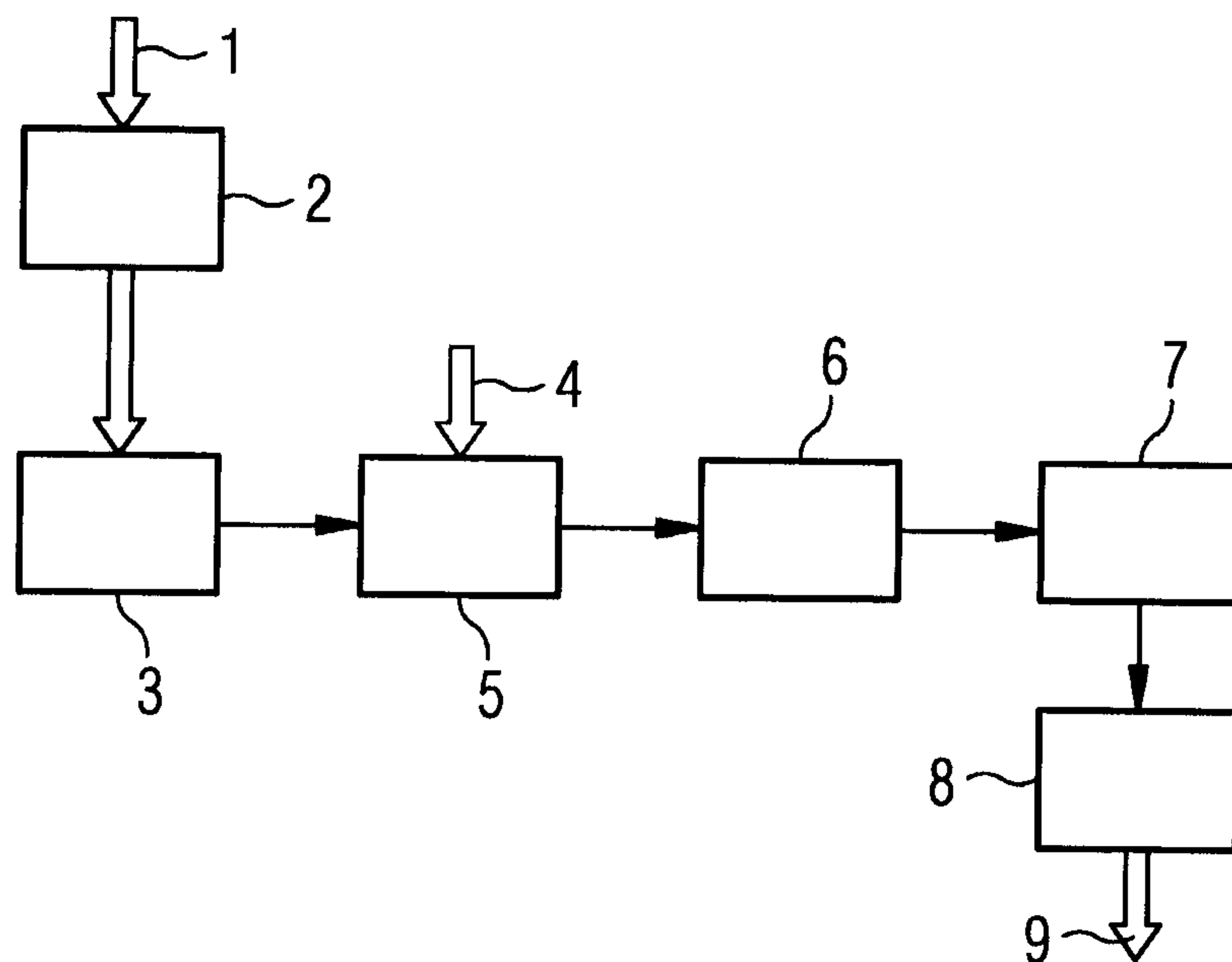


FIG 2

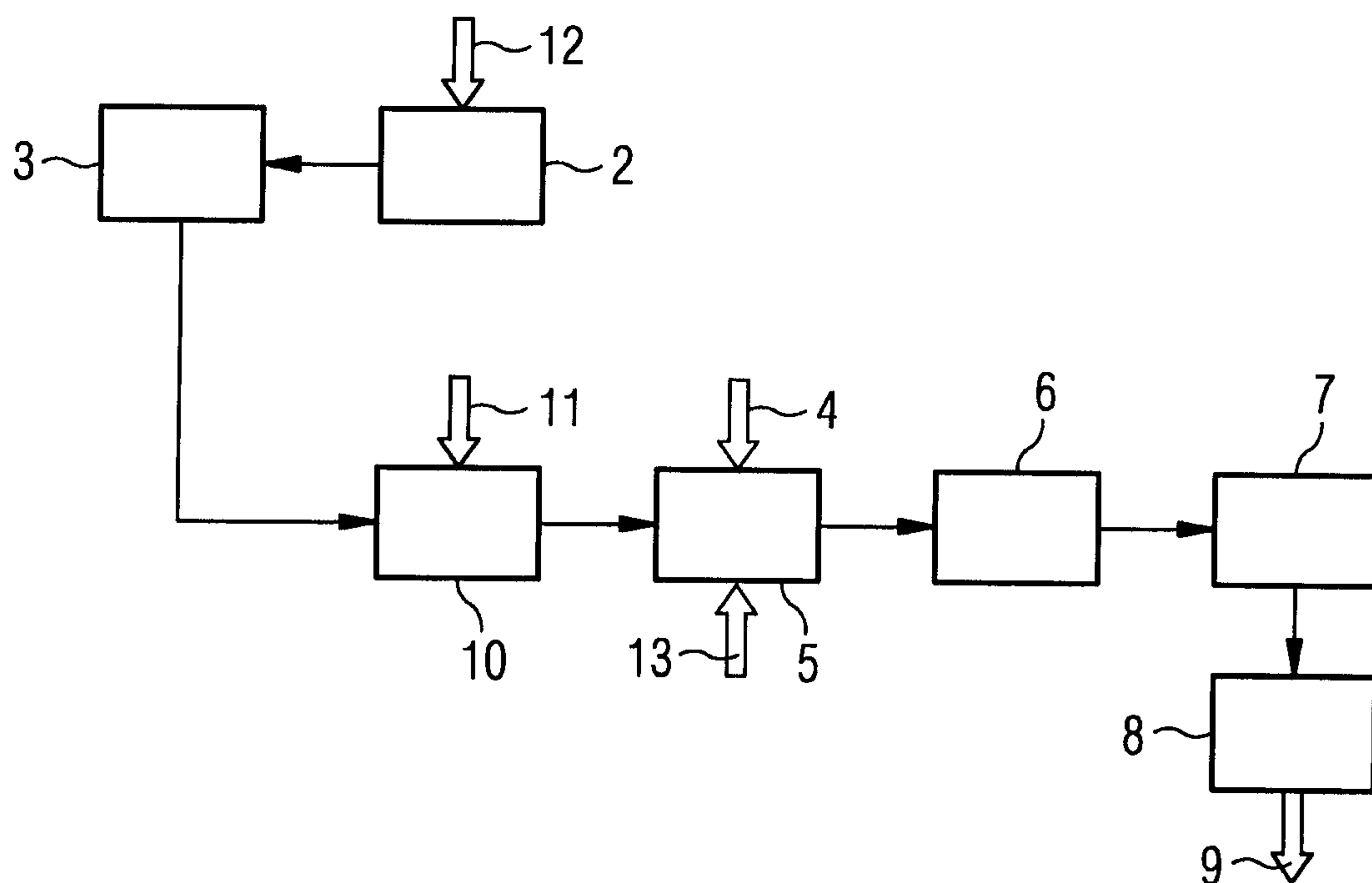


FIG 3

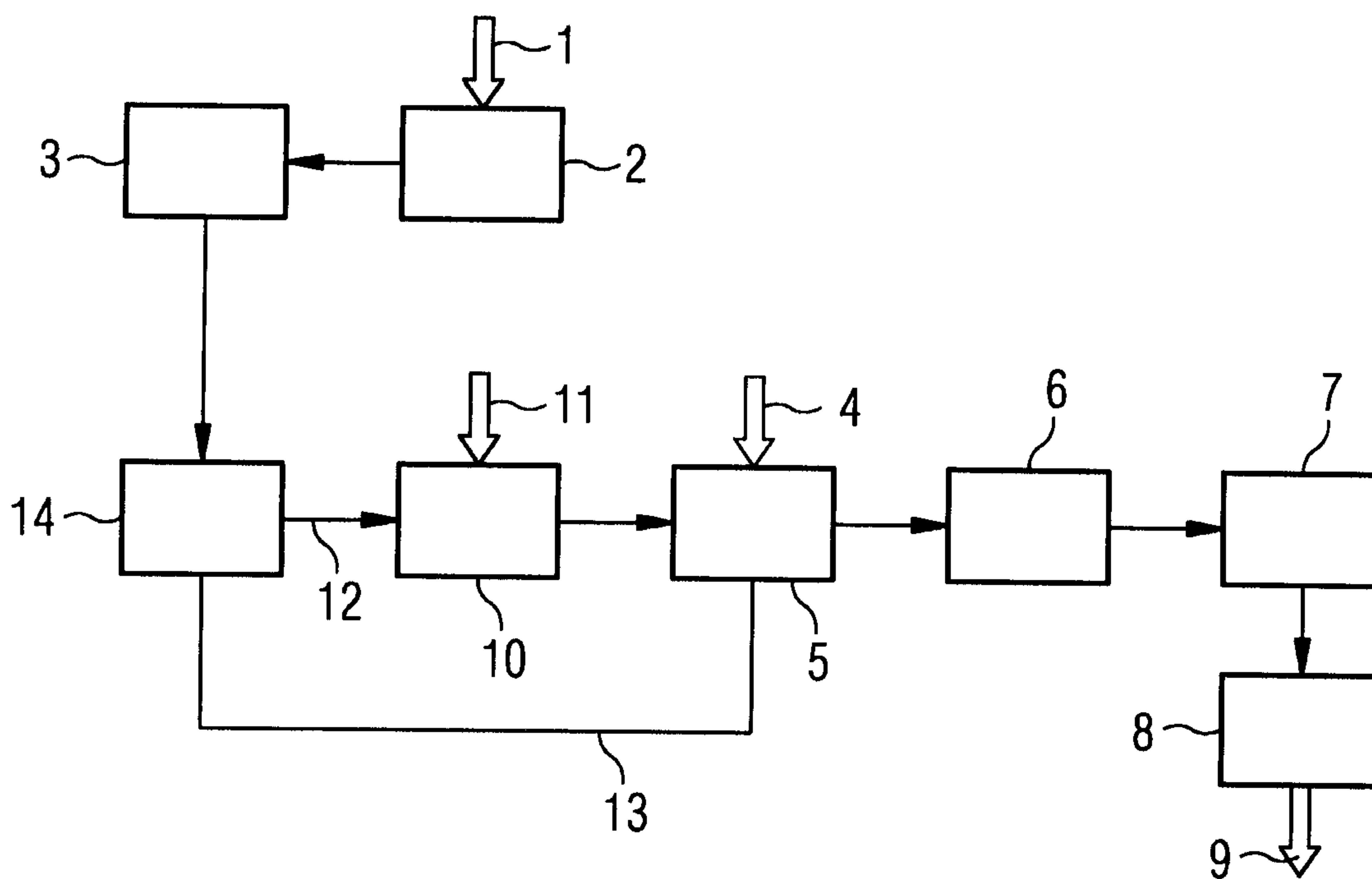


FIG 2

