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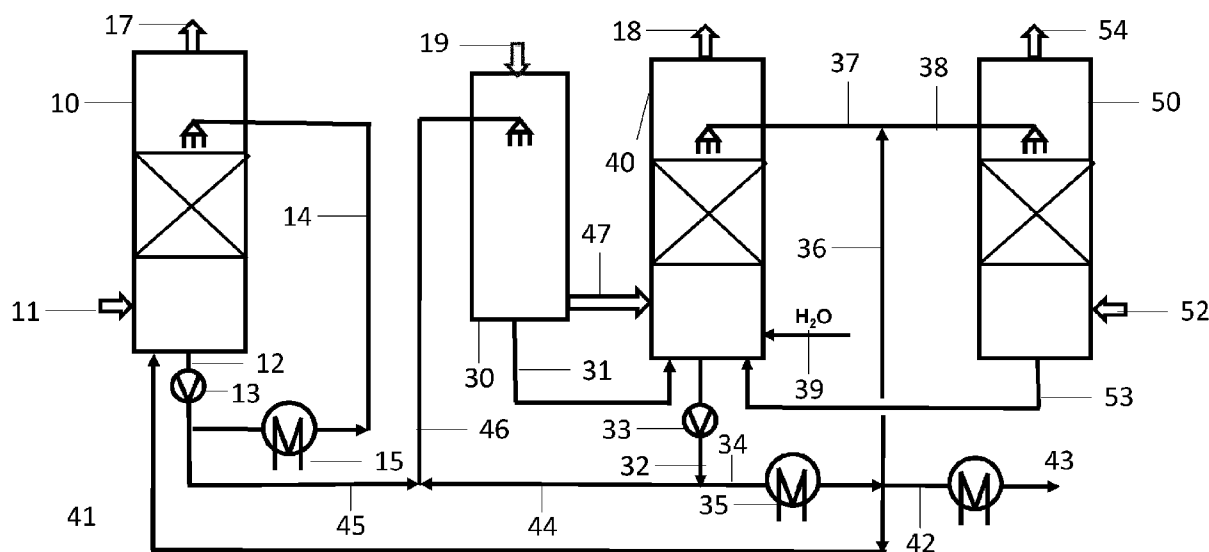


Fig. 2

(57) Abstract: A process and its relating plant for producing sulfuric acid by catalytic oxidation of SO<sub>2</sub> to SO<sub>3</sub> and subsequent absorption of the SO<sub>3</sub> in concentrated sulfuric acid in an intermediate and a final absorber stage. Therein, the intermediate absorber stage features a pre-absorber and a post-absorber. These two absorbers arranged such that the not-absorbed SO<sub>3</sub> leaving the pre-absorber is supplied to a post-absorber and such that the sulfuric acid leaving the post-absorber is split into two streams of which the first stream is recirculated back in the pre-absorber and the second stream is at least partly passed into the post-absorber. The first stream is uncooled directly injected into the pre-absorber with a temperature between 80 and 200 °C, preferably between 100 and 150 °C and the second stream passes a cooling and is at least partly fed into the post-absorber with a temperature between 60 and 90 °C.



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### Process and plant for the production of sulfuric acid

The invention deals with a process and its relating plant for producing sulfuric acid  
5 by catalytic oxidation of  $\text{SO}_2$  to  $\text{SO}_3$  and subsequent absorption of the  $\text{SO}_3$  in  
concentrated sulfuric acid in an intermediate and a final absorber stage, wherein  
the intermediate absorber stage features a pre-absorber and a post-absorber,  
which are arranged such that the not-absorbed  $\text{SO}_3$  leaving the pre-absorber is  
10 supplied to a post-absorber and such that the sulfuric acid leaving the post-ab-  
sorber is split into two streams of which the first stream is recirculated back in the  
pre-absorber and the second stream is at least partly passed into the post-ab-  
sorber.

Typically, sulfur dioxide ( $\text{SO}_2$ ) is formed in a combustion of elemental sulfur. Al-  
15 ternatively, sulfur dioxide can be obtained from off gases out of metallurgical pro-  
cesses, e.g. from the pyrometallurgical production of non-ferrous metals, from the  
roasting of sulfide ores, from the thermal decomposition of metal sulfates or alkali  
sulfates or from the processing of contaminated waste sulfuric acid by thermal  
20 decomposition. First, impurities are removed from this off gas, which impair or  
could impair the quality of the sulfuric acid or the catalytic conversion to sulfur  
trioxide. In a downward second cleaning step, the purified exhaust gas is dried in  
a drying tower with concentrated sulfuric acid of e.g. 94 - 96 wt.-%  $\text{H}_2\text{SO}_4$ , i.e.  
quantitatively freed from water moisture, whereby this sulfuric acid is diluted ac-  
cordingly by the water absorption.

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Independent from the source of the sulfur dioxide, sulfuric acid is usually produced  
using the so-called double absorption process. Thereby, the  $\text{SO}_2$  is converted into  
sulfur trioxide ( $\text{SO}_3$ ) in a multistage converter with the aid of a solid catalyst, e.g.  
with vanadium pentoxide as an active component. The obtained  $\text{SO}_3$  is drawn off  
30 after the first contact stages of the converter and fed to an intermediate absorber.

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SO<sub>3</sub> being drawn off after the last contact stage of the converter is fed to a final absorber. In each absorber, the SO<sub>3</sub> containing gas is traditionally fed counter currently to concentrated sulfuric acid and absorbed in at least one of these two absorbers.

5

In both absorbers, the absorption takes place in concentrated sulfuric acid with a feed concentration of e.g. 98.5 wt.-% H<sub>2</sub>SO<sub>4</sub>, whereby its concentration increases. The water required to form sulfuric acid from SO<sub>3</sub> and H<sub>2</sub>O and to dilute it to approx. 98.5 wt.-% H<sub>2</sub>SO<sub>4</sub> comes partly from the gas/air humidity absorbed in the drying tower. The remaining water is supplied as process water to the intermediate absorber and/or the final absorber. The concentration of sulfuric acid is kept constant by suitable control.

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From WO 2009/065485 it is known to use acid withdrawn from the drying tower in the intermediate absorber. Figure 1 illustrates this well-known process.

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Air or SO<sub>2</sub>-containing gas containing water moisture is fed via line 11 into a drying tower 10. Dried gas is withdrawn by line 17, while acid is withdrawn via line 12 and pump 13. Parts of the acid are recirculated via line 14 and its related heat-exchanger 15. The recirculated acid is used as drying medium in said drying tower 10.

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The other part of the acid is added via line 16 into line 37, combining in line 38. Line 38 is the supply line for the absorbent. This absorbent is used in the intermediate absorber 20 to absorb sulfur trioxide, whereby sulfur trioxide is fed into the intermediate absorber 20 via line 31. To adjust the appropriate concentration, process water is also fed into the intermediate absorber 20 via line 39. The use of sulfuric acid coming from the drying tower via line 16 as an absorbent in the intermediate absorber is a so-called crossflow. Concentration of this acid is

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typically 94 – 96 wt.-%. After mixing of this acid with the recirculation flow line 37, this results in a concentration at line 38 between 97.8 and 98.2% H<sub>2</sub>SO<sub>4</sub>.

5 The SO<sub>3</sub> enriched acid is discharged via line 32 and transferred to the heat exchanger 35 via pump 33. From there, the acid is fed in a first partial flow into line 34. The flow in line 34 is divided again and one of the resulting partial flows is fed into line 37. This flows into the already discussed line 16. The total flow formed from these two partial flows forms the absorbent, which is introduced into the intermediate absorber 20 via line 38.

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The second partial flow from line 32 is returned to drying tower 10 via line 41. The third part is discharged via line 42 and a further heat exchanger 43.

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The second partial flow of line 35 is introduced via line 36 as an absorbent into the final absorber 50, which is fed with SO<sub>3</sub> containing gas via line 52. Sulfuric acid from this absorber 50 is recirculated back into the intermediate absorber 20 via line 53.

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Due to the high solubility of sulfur dioxide in the circulating sulfuric acid of the drying tower, this circuit is particularly suitable for off gases from pyrometallurgical processes containing sulfur dioxide. The concentration of the SO<sub>2</sub> dissolved in the above-mentioned crossflow line 16 is lowered in the acid circulation of the intermediate absorber 20. The partially stripping within this intermediate absorber 20 is favoured by the increased temperature of the acid. The resulting stripped gaseous SO<sub>2</sub> is leaving the intermediate absorber 20 via line 18 and fed with the residual gas to the not-shown subsequent catalytic stage and converted into SO<sub>3</sub>, where it is finally recovered as sulfuric acid in the final absorber 50. As a consequence, the crossflow acid from the drying tower 10 does not enter the acid circulation of the final absorber 50 and thus cannot be stripped there from the

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dissolved SO<sub>2</sub> either, which would ultimately lead to the SO<sub>2</sub> leaving the plant in the chimney gas via line 54 and thus to inadmissible emissions.

5 A disadvantage of this arrangement, however, is that the acid concentration on top of the intermediate absorber is below the optimum of 98.5 wt.-% H<sub>2</sub>SO<sub>4</sub>. This optimal value results from the fact that the minimum acid vapour pressure (azeotrope) is at ~98.5 wt.-% H<sub>2</sub>SO<sub>4</sub>, which, therefore, enables a quantitative absorption of the sulfur trioxide at said concentration. The increase in concentration due to the absorption of SO<sub>3</sub> within the packing in the absorber is limited to a typical  
10 acid outlet concentration of 99.3 to 99.4 wt.-% H<sub>2</sub>SO<sub>4</sub>. The acid outlet concentration must not significantly exceed 99.3-99.4 wt.-% H<sub>2</sub>SO<sub>4</sub>, since the SO<sub>3</sub> partial pressure increases noticeably and would impede complete absorption.

15 However, the discussed lower acid concentration during the entry into the intermediate absorber caused by the addition of the crossflow acid from the drying tower according to the above-mentioned process known from WO 2009/065485 is no longer correlated to the explained azeotropic concentration. Consequently, the process is also no longer has the lowest total vapour pressure and at acid concentrations below 98.5 wt.-% H<sub>2</sub>SO<sub>4</sub>, both the H<sub>2</sub>SO<sub>4</sub> vapour pressure and the  
20 SO<sub>3</sub> partial pressure decrease, while the H<sub>2</sub>O partial pressure increases. As a result, the lower acid concentration does not lead to a reduction in SO<sub>3</sub> absorption, but leads to increased mist formation, which in turn requires the installation of highly efficient mist filters (candle filters) in the head of the intermediate absorber.

25 Moreover, excessive acid mist would also form within the hot SO<sub>3</sub>-containing gas entering the tower. This formed mist has the disadvantage that it could not be separated in the subsequent packing and would require additional extensive measures to remove it in order to protect the downstream apparatus from corrosion.

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The concentration difference of typical (outlet - inlet)  $99.3 - 98.5 = 0.8$  wt.-%  $H_2SO_4$  together with the amount of sulfur trioxide to be absorbed determines the required acid circulation quantity at the intermediate absorber. With increasing  $SO_2$  content of the input gas and thus a higher  $SO_3$  content of the gas entering the intermediate absorber, the acid circulation quantity must be increased practically proportionally. As a result, the flooding limit of conventional ceramic packing would be exceeded. In such cases, it is therefore necessary to increase the diameter of the tower considerably for hydraulic reasons, even if the mass transfer does not require this.

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A splitting of the intermediate absorption stage in at least two devices is also known from US 7,837,970 B2. This ensures the possibility of cheap device, like a venturi as a first pre-absorber and smaller packed bed absorber as a post absorber. As a result, investment costs can slightly be reduced. However, it does not have any impact on mist formation.

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Therefore, it is the subject of the current invention, to provide a process and a correlating plant with improved stripping of dissolved  $SO_2$  and, therefore preventing stripped  $SO_2$  vanishing the plant towards the stack, and also a reduction of acid mist formation without a hydraulic over-dimensioning of the intermediate absorber.

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This aim is achieved with a process according with the features of claim 1.

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Such a process for producing sulfuric acid by catalytic oxidation of  $SO_2$  to  $SO_3$  and subsequent absorption of the  $SO_3$  in concentrated sulfuric acid requires intermediate and a final absorber, wherein the absorption takes place. In both absorbers, the absorption takes place in concentrated sulfuric acid with a feed concentration of e.g. 98.5 wt.-%  $H_2SO_4$ , whereby its concentration increases.

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The intermediate absorber features a pre-absorber and a post-absorber. Both absorbers, which build together the intermediate absorber, are arranged such that the not-absorbed SO<sub>3</sub> leaving the pre-absorber is supplied to the post-absorber. The sulfuric acid withdrawn from the post-absorber is split into two streams of which the first stream is recirculated back in the pre-absorber and the second stream is at least partly passed into the post-absorber.

It is the basic idea underlying the invention that the first stream is uncooled directly injected into the pre-absorber with a temperature between 80 and 200 °C. Preferably, the temperature of the sulfuric acid being injected into the pre-absorber is between 100 and 150 °C. Uncooled in the sense of the invention means that it does not pass any heat exchanger, so heat losses via line losses are the only heat losses. It is possible to admix the first stream with sulfuric acid from another source, like the drying tower.

The second stream passes a cooling and is at least partly fed into the post-absorber with a temperature between 60 and 90 °C. Further streams for the post-absorber or a complete withdrawing can be branched off.

This design has the technical effect that mist formation in the pre-absorber can be reduced significantly or completely suppressed. This due to the low temperature difference between the entering gas and acid fed to the pre-absorber.

Moreover, the heat exchanger is smaller which leads to lower investment costs.

In a preferred embodiment, the pre-absorber is designed as an empty tube or venturi, which leads to very low investment costs due to the simple design. Also, controlling and maintenance can be done economically and easily.



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Additionally or alternatively, the post-absorber is designed as a packed-bed absorber. This ensures a maximum efficiency of the absorption. Due to the additional pre-absorber, the packed bed can be smaller without a risk of hydraulic flooding, which further lowers investment costs.

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Moreover, the first stream features a flow rate between 20 and 80 %, preferably between 25 and 40 % of the sulfuric acid withdrawn from the post-absorber. This leads to the most efficient heat distribution. Particular the preferred range leads to significantly reduced dimensions of and, therefore, costs for the post-absorber

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While the pre-absorber is operated with a feed acid concentration below 98.5% H<sub>2</sub>SO<sub>4</sub> (because of the addition of “weaker” acid from drying tower crossflow), the post-absorber is operated with conventional 98.5% acid at the inlet and thus operates under ideal absorption conditions.

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It is also preferred that the first acid stream entering the pre-absorber features a temperature difference to the entering SO<sub>3</sub> containing gas of less than 50 °C, preferably less than 30 °C. Thus, the temperature of both streams are nearly equal which increases absorption rates. In detail, the elevated temperature of this acid thus favors the stripping of the SO<sub>2</sub>. As a result, the inventive process significantly reduces mist formation, which means less efficient mist filters are required and thus lower gas pressure loss therein due to the lower temperature difference between the incoming gas and the feed liquid / acid at the pre-absorber.

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As a further advantage of the invention, the SO<sub>2</sub> containing feed gas to the plant has a concentration of more than 9.0 vol.-% prior to the conversion, preferably more than 11.5 vol.-%. These are relatively high concentrations, which are difficult to handle in formally known processes due to the flooding limit of conventional ceramic packing.

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It has to be understood that the higher the concentration of the SO<sub>2</sub> content in the input gas, the more advantageous the process according to the invention. According to the invention, the input gas into the converter for the conversion of the sulfur dioxide has 6.5 to 30 vol.% SO<sub>2</sub>, preferably > 11 vol.% SO<sub>2</sub>. The process is therefore particularly suitable for use in conjunction with a process for the catalytic conversion of gases with a high SO<sub>2</sub> content, as described in DE 102 49 782 A1.

Furthermore, air or SO<sub>2</sub> containing gas is dried in at least one drying tower by means of sulfuric acid for removing water moisture.

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It is preferred to admix the sulfuric acid supplied into the pre-absorber with sulfuric acid from another source. In this context, it is particularly preferred that the sulfuric acid from the other source is sulfuric acid from the drying tower. Consequently, a closed loop of sulfuric acid as absorbent is possible. Preferably, the temperature of the acid from the other source, particularly the drying tower is between 80 and 180 °C, preferably above 90 °C.

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It is also preferred that the second stream is mixed with sulfuric acid from another source upstream to the cooling. This reduces the number of acid circuits.

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It has further been recognized that the second stream can be further split in at least two streams downstream of the cooling. So, this third stream can be fed in the final absorber after the cooling. As a result, an additional cooling device for the final absorber is omitted.

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In addition, it is necessary to add process water to the sulfuric acid for adjusting the acid concentration. In the invention, it is preferred to feed this into the lower vestibule of the pre-absorber and/or the lower vestibule of the post-absorber and/or the lower vestibule of the final absorber. So, no separate devices are necessary.

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However, it is also preferred that process water is added to the sulfuric acid second stream as discharged from the acid cooler, at a separate device. As a result, concentration of sulfuric acid is higher when passing the cooler, which avoids or  
5 substantially reduces corrosion. It is also possible that process water is added to the sulfuric acid first stream entering the pre-absorber, at a separate device.

The invention further belongs to a plant according to claim 12, particular a plant  
10 for performing a process according to claims 1 to 11.

Such a plant for the production of sulfuric acid for producing sulfuric acid by catalytic oxidation of  $\text{SO}_2$  to  $\text{SO}_3$  and subsequent absorption of the  $\text{SO}_3$  in concentrated sulfuric acid in an intermediate and a final absorber. The intermediate absorber features a pre-absorber and a post-absorber which are arranged such that  
15 the not-absorbed  $\text{SO}_3$  leaving the pre-absorber is supplied to a post-absorber. A discharge conduit for sulfuric acid from the post-absorber is split into a recirculation conduit feeding sulfuric acid back into the pre-absorber and a supply conduit feeding sulfuric acid into the post-absorber.

20 It is essential for the invention that a cooler is only foreseen in the supply conduit to the post-absorber. The plant does not feature a cooling of the acid directed to the pre-absorber, particular no heat exchanger is placed in the acid feeding line of the pre-absorber.

25 This design has the benefit that only part of the conventionally required amount of cooled circulating acid is transferred to the said post-absorber, hydraulic flooding is prevented. Simultaneously, size and cost of this post absorption tower are reduced. This is due to the fact that the total amount of acid is determined by increasing the concentration of circulating acid by typically 99.3 to 98.5 wt,-%  
30  $\text{H}_2\text{SO}_4$  based on the total  $\text{SO}_3$  supplied to the intermediate absorber, while the

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amount of acid supplied to the packed bed post-absorber is approximately 20-90 wt.-%, preferably 50-70 wt.-% of the conventionally calculated total amount of acid.

5 The inventive design splits the intermediate absorption tower into two separate vessels. Preferably, the pre-absorber is an empty tube or venturi type and/or the post-absorber is designed as a packed bed counter-current tower. While the post-absorber is fed with cooled acid, the pre-absorber is fed with non-cooled acid. So, only one cooler is foreseen, whereby the term "cooler" also include the possibility  
10 of at least two cooling devices.

In a preferred embodiment, an air-cooled heat exchanger is used as the cooling device for the post-absorber, which is of particular importance. The increasing scarcity of suitable cooling water is widespread and increasingly noticeable, especially in areas with limited / non-existent availability of cooling water, as well as  
15 e.g. with regulated limited withdrawal possibilities from rivers or oceans. For this reason, fin-fan air coolers have already been used in the past to dissipate excess energy from acid cooling. The available materials for this air coolers were e.g. 316L/304 with acid temperatures up to 80 °C or anodic protected air coolers from  
20 304 up to 110 °C, whereby the latter have very limited proven record in practice.

The use of air-cooled heat exchangers for cooling the circulating acid is advantageous for the invention-based process with acid temperatures of > 80 °C, better >100 °C, much better >120 °C, and the use of suitable material, such as Si-alloyed  
25 stainless-steel material e.g. SX<sup>TM</sup>, both in terms of investment and operating costs, as well as space required. The higher temperature level and the lower volume of circulating acid result in a considerably smaller cooler (dimensions, weights, cost) and lower power consumption for the cooling air fans. In addition, in the case of air coolers, the risk potential due to leakage of acid in cooling water  
30 (and vice versa) is eliminated, so that, for example, the possibility of the formation

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of hydrogen as a reaction product of corrosion (and the associated risk of explosion) can be largely prevented.

Moreover, it is preferred that the pre-absorber is made of carbon steel with internal lining and the post-absorber is made of stainless steel or carbon steel with an internal brick lining. Conventional absorbers are usually made of carbon steel with an internal acid-resistant brick lining. Although the materials used are relatively inexpensive, the installation is costly due to the manual work involved in the lining. Alternatively, absorption towers are built from stainless steel, resulting in reduced costs mostly caused by faster installation.

However, these stainless steels are susceptible to corrosion if the operating conditions deviate from the specifications. Preferably such towers are made of Si-alloyed stainless-steel material, e.g. SX<sup>TM</sup>, or austenitic alloys, e.g. 310S, whereas Si-alloyed stainless steel offers lower resistant to oleum (with free SO<sub>3</sub> = > 100 wt.-% H<sub>2</sub>SO<sub>4</sub>). Due to the high SO<sub>3</sub> concentration of the inlet gas, oleum can form locally and lead to significant corrosion. Therefore, the acceptance of this design in industry is somewhat sceptical. The inventive process eases this situation in the post-absorber, wherein the incoming SO<sub>3</sub> is already partly absorbed in the pre-absorber and consequently the gas at the inlet to the post-absorber has a much lower SO<sub>3</sub> concentration compared to the conventional process and hence a lower risk of oleum formation. According to the inventive process, the pre-absorber is made of carbon steel with internal lining, and the post-absorber offers the opportunity of comfortable design as "brick-lined as well as stainless", which can, therefore, be used for cost optimization. This leads to reduced investment costs.

In addition, a drying tower for drying a gas containing air or SO<sub>2</sub> containing gas with sulfuric acid is foreseen. The drying tower features preferably a pre-drying tower and a post-drying tower, which are arranged such that moisture containing

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air or SO<sub>2</sub> containing gas is supplied to a pre-drying tower, wherein a discharge conduit for acid from the post-drying tower is split into a recirculation conduit feeding sulfuric acid back into the pre-drying tower and a supply conduit feeding sulfuric acid into the post-drying tower. This enables an improved drying similar as  
5 the splitting of the intermediate absorber into two devices.

Further developments, advantages and possible applications can also be taken from the following description of exemplary embodiments and the drawings. All features described and/or illustrated from the subject matter of the invention per  
10 se or in any combination, independent of their inclusion in the claims or their back reference.

In the drawings:

- 15 Fig. 1 shows schematically a process known from the state of the art
- Fig. 2 shows schematically a first process according to the invention whereby the post-absorber and the final absorber feature a common acid supply conduit,  
20
- Fig. 3 shows schematically a second process according to the invention whereby the post-absorber and the final absorber feature separate acid circuit,
- 25 Fig. 4 shows schematically a third process according to the invention whereby drying tower is separated in a pre-dryer and a post-dryer,
- Fig. 5 shows schematically a fourth process according to the invention whereby the dryer, the post-absorber and the final absorber feature  
30 a common acid supply conduit,

Fig. 6 shows schematically a fifth process according to the invention whereby only one source for process water is used in a common acid circuit and

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Fig. 7 shows schematically a sixth process according to the invention whereby only one source for process water is used in the pre-absorber.

10 Figure 1 has already been discussed in the introduction discussing the state of the art in detail.

Figure 2 shows an installation according to the invention method for processing. A sulfur dioxide containing gas, e.g. a pyrometallurgical off gas containing SO<sub>2</sub>,  
15 is introduced via conduit 19 into pre-absorber 30, which represents together with post-absorber 40 the intermediate absorber. Preferably, the pre-absorber is designed as an empty tower or as a venturi. Sulfuric acid is added via conduit 46 is absorption agent.

20 Liquid and gaseous parts are withdrawn via conduit 31 and 47 and introduced in the post-absorber 40, which is preferably designed as a packed-bed absorber. Therein, remaining SO<sub>3</sub> is absorbed, preferably counter-current, with liquid acid fed in via conduit 37 and withdrawn via conduit 32 and pump 33. Parts of the withdrawn acid stream are recycled via conduit 44 to mix it with fresh sulfuric acid  
25 from conduit 45 and recirculated via conduit 46 into the pre-absorber 30.

Water is added into the sump of the post-absorber 40 via conduit 39. Alternatively, the process water can be added to the sump of the pre-absorber 30 or to a not-shown pump tank.

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The other part of the sulfuric acid stream is passed from conduit 32 in conduit 34 and cooler 35. Parts of the cooled acid are fed via conduit 36 and 37 back into the post-absorber 40. So, the absorption agent of the post-absorber 40 is cooled.

5 Talking about the sulfuric acid stream of conduit 36, it is further possible to send a part of this stream via conduit 38 into a final absorber 50, which is also preferably designed as a packed bed. Therein, sulphur trioxide containing gas is fed in via conduit 52. Gas is blown out via conduit 54 while acid from the sump of the final absorber 50 is recirculated back via line 53 into the sump of post-absorber  
10 40.

Another part of the stream from conduit 34 is withdrawn via conduit 42 and optionally an additional cooler 43. The last part is recycled via conduit 41 into a drying tower 10.

15

Sulfuric acid is fed via line 14 into this drying tower 10. Dried acid is withdrawn via line 12 and pump 13. Parts of the acid are recirculated via line 14 and its related heat-exchanger 15. The recirculated acid is used as drying medium in said drying tower 10. Parts of the sulfuric acid are withdrawn from conduit 12 and fed  
20 into the pre-absorber 30 via conduits 45 and 46.

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This embodiment of the invention shows a common acid circulation with the post-absorber 40 and the final absorber 50. It features the common acid cooler 35. The distribution of the circulating acid required for the intermediate absorption typically amounts to typically 65% cooled for the post-absorber 40 via lines 36 and 37 and the residual 35% uncooled via line 44 to the pre-absorber 30 but can vary depending on the SO<sub>2</sub> content of the gases to be processed.

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Figure 3 shows another embodiment which differs from figure 2 in the acid circuits. The cooled sulfuric acid from conduit 34 and cooler 35 is split into a stream



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recirculated via conduit 41 in the drying tower 10 and a cooled stream fed into the post-absorber 30 via conduit 37. The remaining part is passed into the sump of the final absorber 50 via conduit 42.

5 SO<sub>3</sub> containing gas is injected into this final absorber via conduit 52. An independent acid circuit feeds acid into the final absorber 50 via conduit 58. The acid is withdrawn via conduit 53, pump 51 and a separate cooler 55. Parts of the acid are withdrawn via conduit 56 and cooler 57 while the other part is recirculated via conduit 58. Water is added separately via conduit 59.

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Figure 4 shows an embodiment with an arrangement of the absorbers as it is known from figure 2. The difference is the arrangement of the drying towers, where the drying tower 10 is split into a pre-drying tower 1 and a post-drying tower 6. Gas is inserted via conduit 11 into the pre-drying tower 1 and dried with acid from conduit 9. Acid from the common acid circuit of post-absorber and final absorber is recycled in the sump of pre-drying tower 1 via conduit 41. Naturally, all other shown possibilities of separated acid circuits and water adding are also possible.

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20 Acid and gas are withdrawn from the sump of pre-drying tower 1 via conduits 2 and 3 and fed into post-drying tower 6, preferably counter-current. Acid from its sump is withdrawn via conduit 4 and pump 5. A first stream from conduit 4 is recirculated via conduit 9 into the pre-drying tower 1.

25 Another stream from conduit 4 is recirculated in the post-drying tower 6 via line 14 and cooler 15 while the last part is transported to the pre-absorber via conduits 45 and 46. A third stream is passed via line 9 back into the pre-drying tower 1.

Figure 5 shows another alternative of the current invention, using only one drying tower 10. In contrast to figure 2, figure 5 shows a common acid circuit of the

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absorbers 30, 40 and 50 as well as the drying tower 10. This leads to a further reduction of the coolers since only the two coolers 35 and 43 remain in the process.

5 In detail, the drying tower 10 does no longer feature a separate acid circuit, but its absorbent is fed in via conduit 41, which is branched off conduit 34 after passing cooler 35. So, it is derived from acid from the sump of post-absorber 40.

10 All acid from the sump of the drying tower 10 is withdrawn via conduit 12 and 49 and mixed with acid from the sump of the pre-absorber 30.

15 Figure 6 shows another embodiment of the invention directed to an additional admixing of process water. Therein, the common acid circuit of post-absorber 40 and final absorber 50 features an additional water diluter 60. Therein, water is admixed via conduit 61 to the stream in conduit 34, which is used as absorbent in both absorber 40 and 50. Alternatively, said diluter 60 can be located in-line 34 upstream the common acid cooler 35.

20 Figure 7 depicts another possibility for adding process water to regulate and control the acid concentration. A diluter 62 is positioned in the supplying conduit 46 for the acid used as absorbent in the pre-absorber 30.

**Reference numbers**

	1	pre-drying tower
5	2 - 4	conduit
	5	pump
	6	post-drying tower
	7 - 9	conduit
	10	drying tower
10	11, 12	conduit
	13	pump
	14	conduit
	15	cooler
	16 - 19	conduit
15	20	intermediate absorber
	30	pre-absorber
	31, 32	conduit
	33	pump
	34	cooler
20	35 - 39	conduit
	40	post-absorber
	41, 42	conduit
	43	cooler
	44 - 49	conduit
25	50	final absorber
	51 - 59	conduit
	60	diluter

61 conduit

62 diluter

63 conduit

**Claims:**

1. A process for producing sulfuric acid by catalytic oxidation of  $\text{SO}_2$  to  $\text{SO}_3$  and subsequent absorption of the  $\text{SO}_3$  in concentrated sulfuric acid in an intermediate and a final absorber stage, wherein the intermediate absorber stage features a pre-absorber and a post-absorber, which are arranged such that the not-absorbed  $\text{SO}_3$  leaving the pre-absorber is supplied to a post-absorber and such that the sulfuric acid leaving the post-absorber is split into two streams of which the first stream is recirculated back in the pre-absorber and the second stream is at least partly passed into the post-absorber, **characterized in** that the first stream is uncooled directly injected into the pre-absorber with a temperature between 80 and 200 °C, preferably between 100 and 150°C and the second stream passes a cooling and is at least partly fed into the post-absorber with a temperature between 60 and 90 °C.
2. A process according to claim 1, **characterized in** that the pre-absorber is designed as an empty tube or venturi and the post-absorber is designed as a packed-bed absorber.
3. A process according to claim 1 or 2, **characterized in** that the first stream features a flow rate between 20 and 80 %, preferably between 25 and 40 % of the sulfuric acid withdrawn from the post-absorber.
4. A process according any of the previous claims, **characterized in** that the first stream features a temperature difference to the entering  $\text{SO}_3$  containing gas of less than 50 °C, preferably less than 30 °C.

5. A process according any of the previous claims, **characterized in** that the SO<sub>2</sub> containing feed gas to the plant has a concentration of more than 9,0 vol.-% prior to the conversion, preferably more than 11.5 vol.-%.
- 5 6. A process according to any of the previous claims, **characterized in** that air or SO<sub>2</sub> containing gas is dried in at least one drying tower by means of sulfuric acid and that the sulfuric acid discharged from the drying tower is supplied to the pre-absorber.
- 10 7. A process according to any of the previous claims, **characterized in** that the sulfuric acid from another source is admixed with the first stream and/or the second stream.
- 15 8. A process according any of the previous claims, **characterized in** that the second stream is further split in at least two streams downstream of the cooling.
9. A process according any of the previous claims, **characterized in** that process water is added to the sulfuric acid.
- 20 10. A process according any of the previous claims, **characterized in** that process water is added to the sulfuric acid second stream as discharged from the acid cooler, at a separate device
- 25 11. A process according any of the previous claims, **characterized in** that process water is added to the sulfuric acid first stream entering the pre-absorber, at a separate device.
- 30 12. A plant for the production of sulfuric acid for producing sulfuric acid by catalytic oxidation of SO<sub>2</sub> to SO<sub>3</sub> and subsequent absorption of the SO<sub>3</sub> in concentrated sulfuric acid in an intermediate and a final absorber stage, wherein the intermediate absorber stage features a pre-absorber (30) and a post-absorber (40),

which are arranged such that the not-absorbed SO<sub>3</sub> leaving the pre-absorber (30) is supplied to a post-absorber (40), wherein a conduit (32) for withdrawing sulfuric acid from the post-absorber (40) is split into a conduit (46) recirculating sulfuric acid back into the pre-absorber (30) and a conduit (34) supplying sulfuric acid at least partly into the post-absorber, **characterized in** that a cooler (35) is foreseen only in any of the conduits (34,36) supplying sulfuric acid to the post-absorber (40).

13. A plant according to claim 12, **characterized in** that the cooler (35) is an air-cooled heat exchanger.

14. A plant according to claim 12 or 13, **characterized in** that the pre-absorber (30) is made of carbon steel with internal lining and/or the post-absorber (40) is made of stainless steel or carbon steel with an internal brick lining.

15. A plant according to any of the previous claims 12 to 14, **characterized in** that air or SO<sub>2</sub> containing gas is dried in a drying tower with sulfuric acid, wherein the drying tower features a pre-drying tower (1) and a post-drying tower (6), which are arranged such that air or SO<sub>2</sub> containing gas is supplied to the pre-drying tower (1), wherein a conduit (4) for acid from the post-drying tower (6) is split into a conduit (9) recirculating sulfuric acid back into the pre-drying tower (1) and a conduit (14) supplying sulfuric acid into the post-drying tower (6), and that a cooler (15) is only foreseen in the conduit (14) supplying sulfuric acid to the post-drying tower (6).

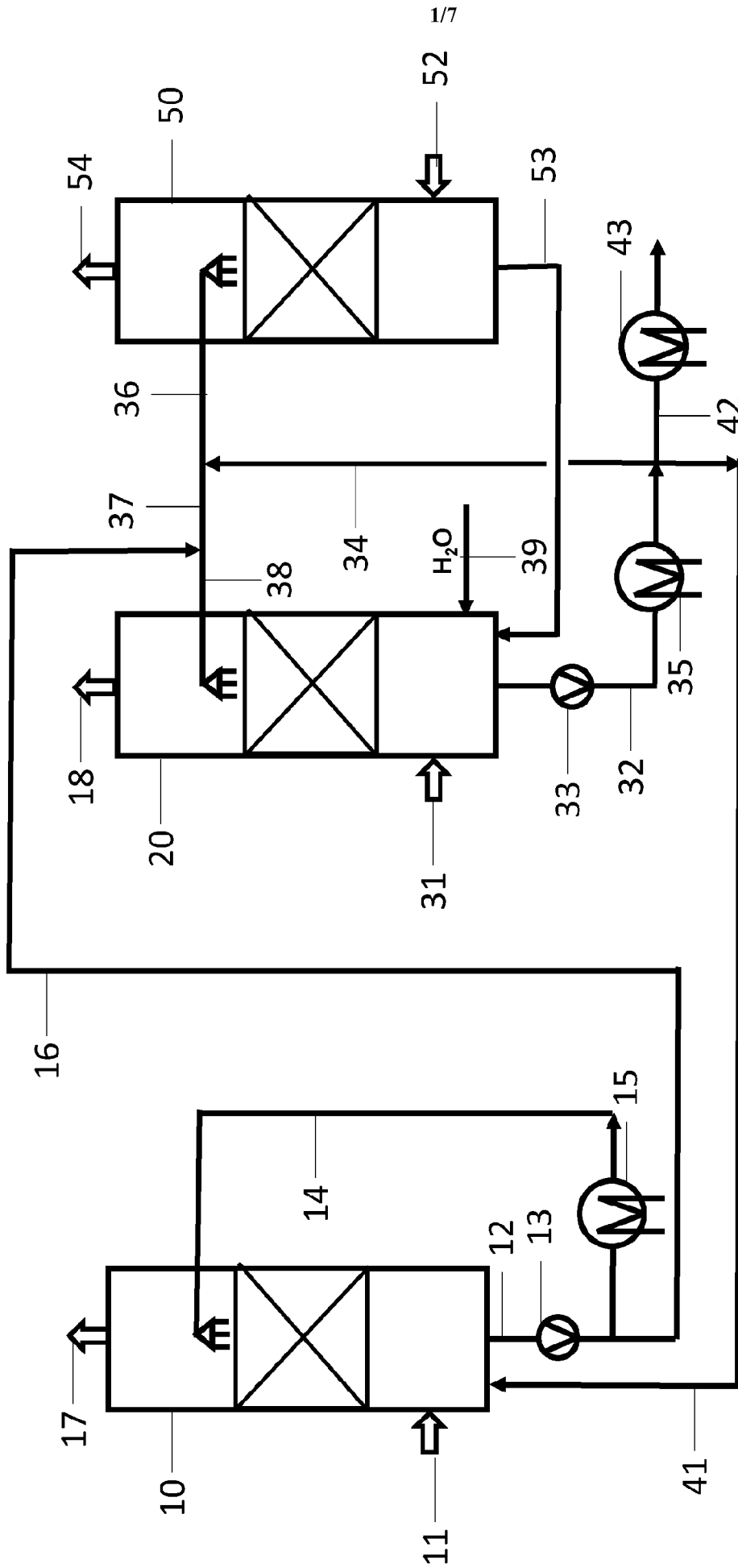


Fig. 1



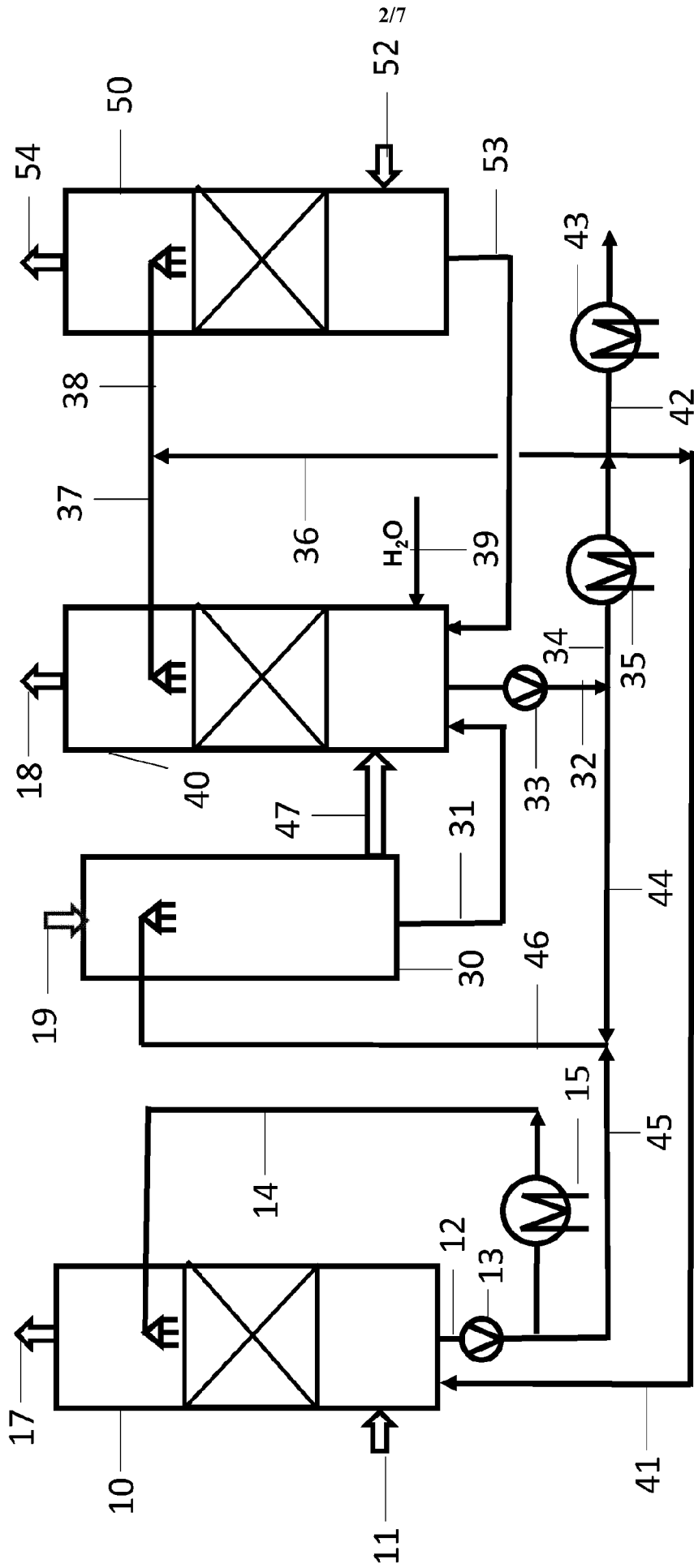


Fig. 2

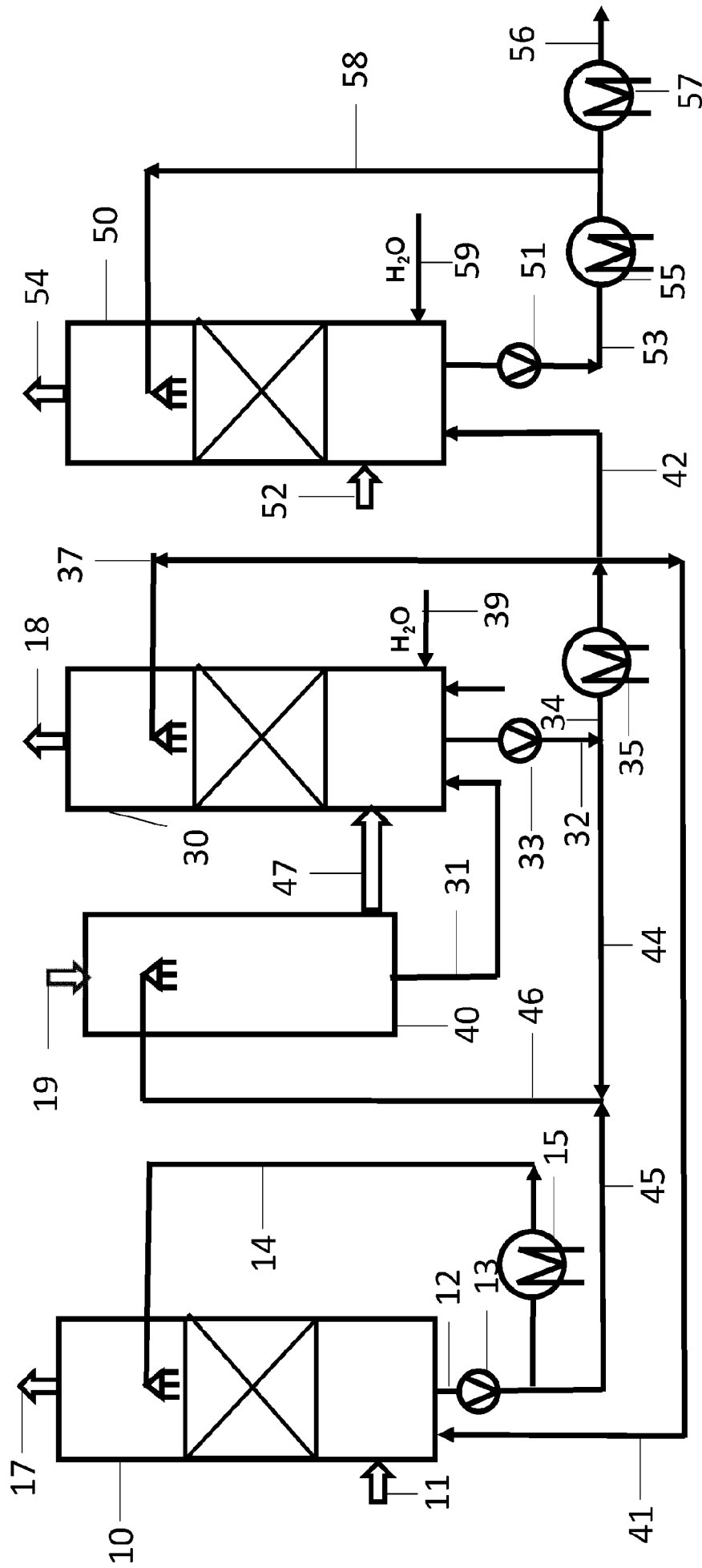


Fig. 3

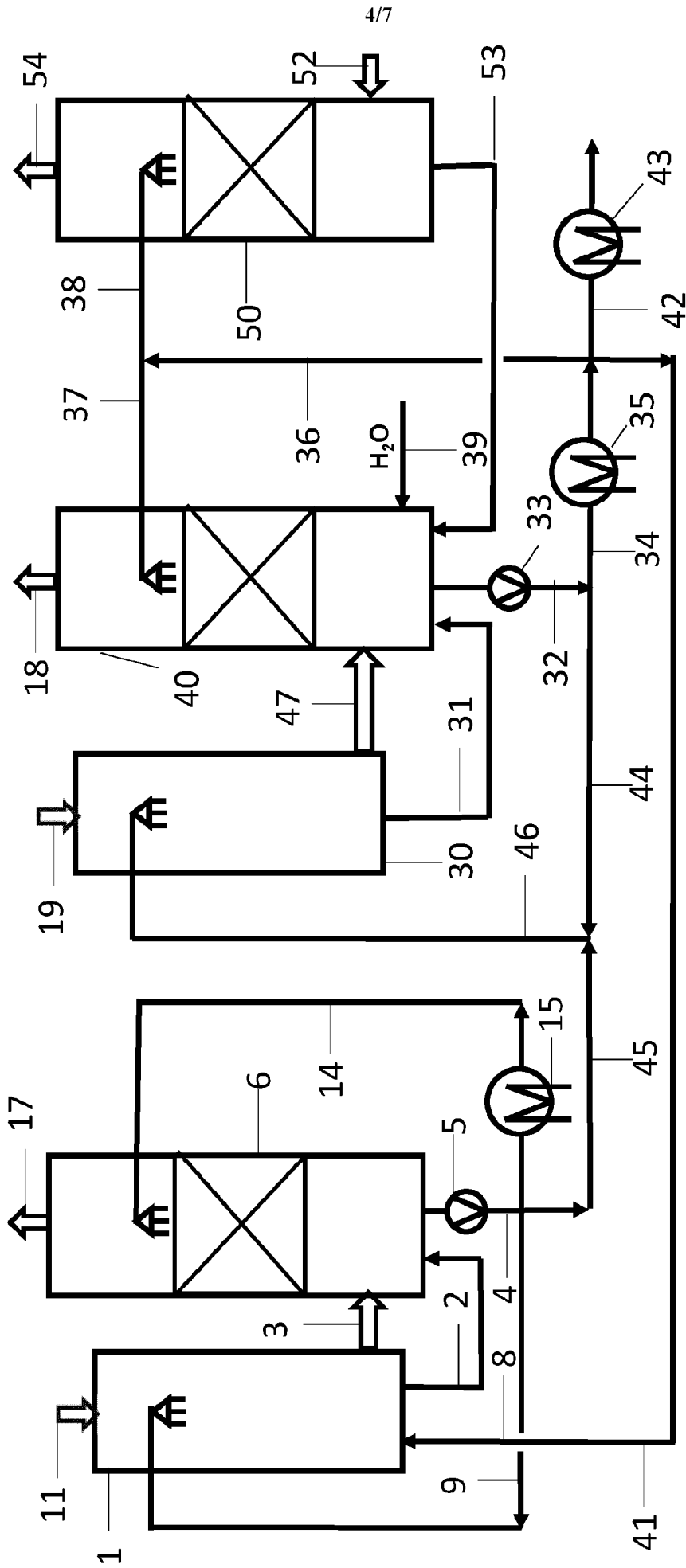


Fig. 4

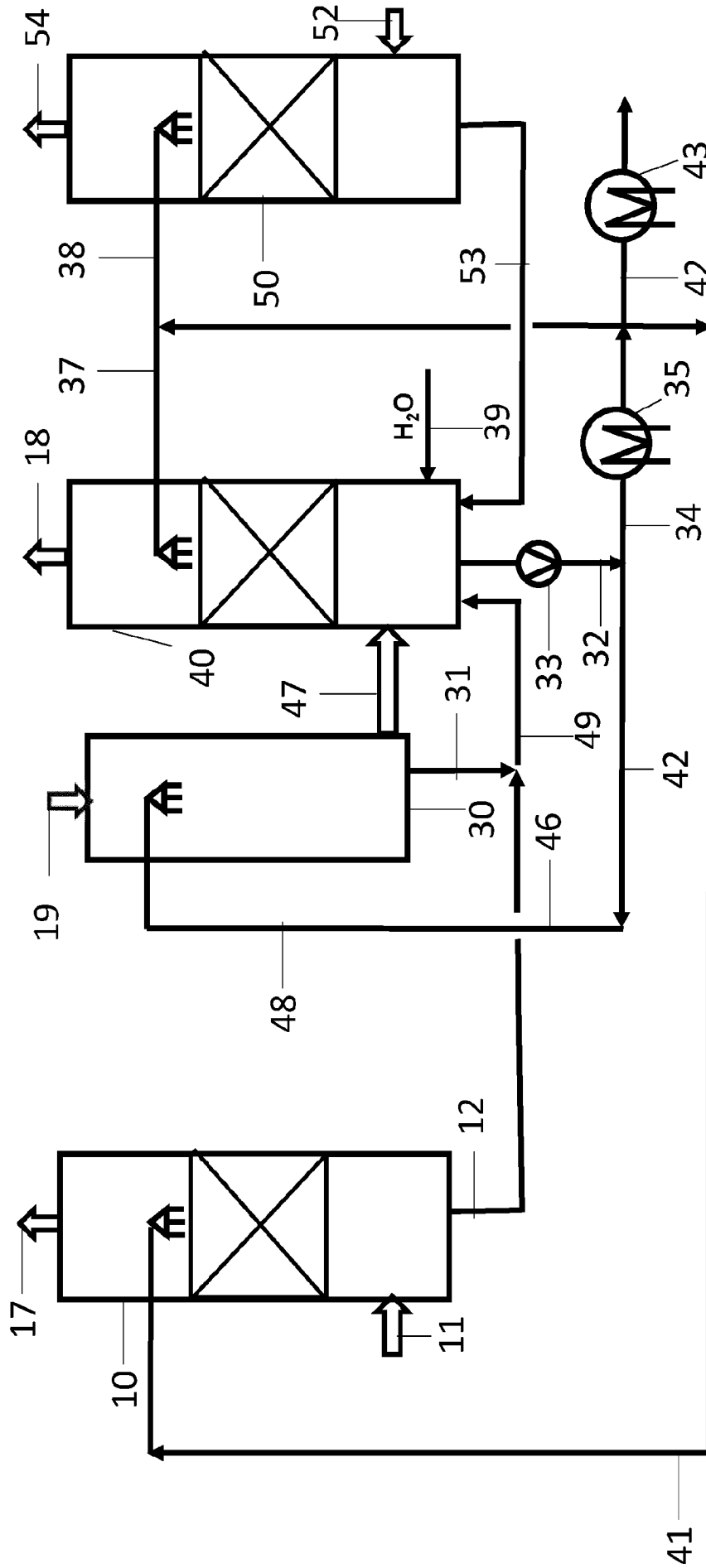


Fig. 5

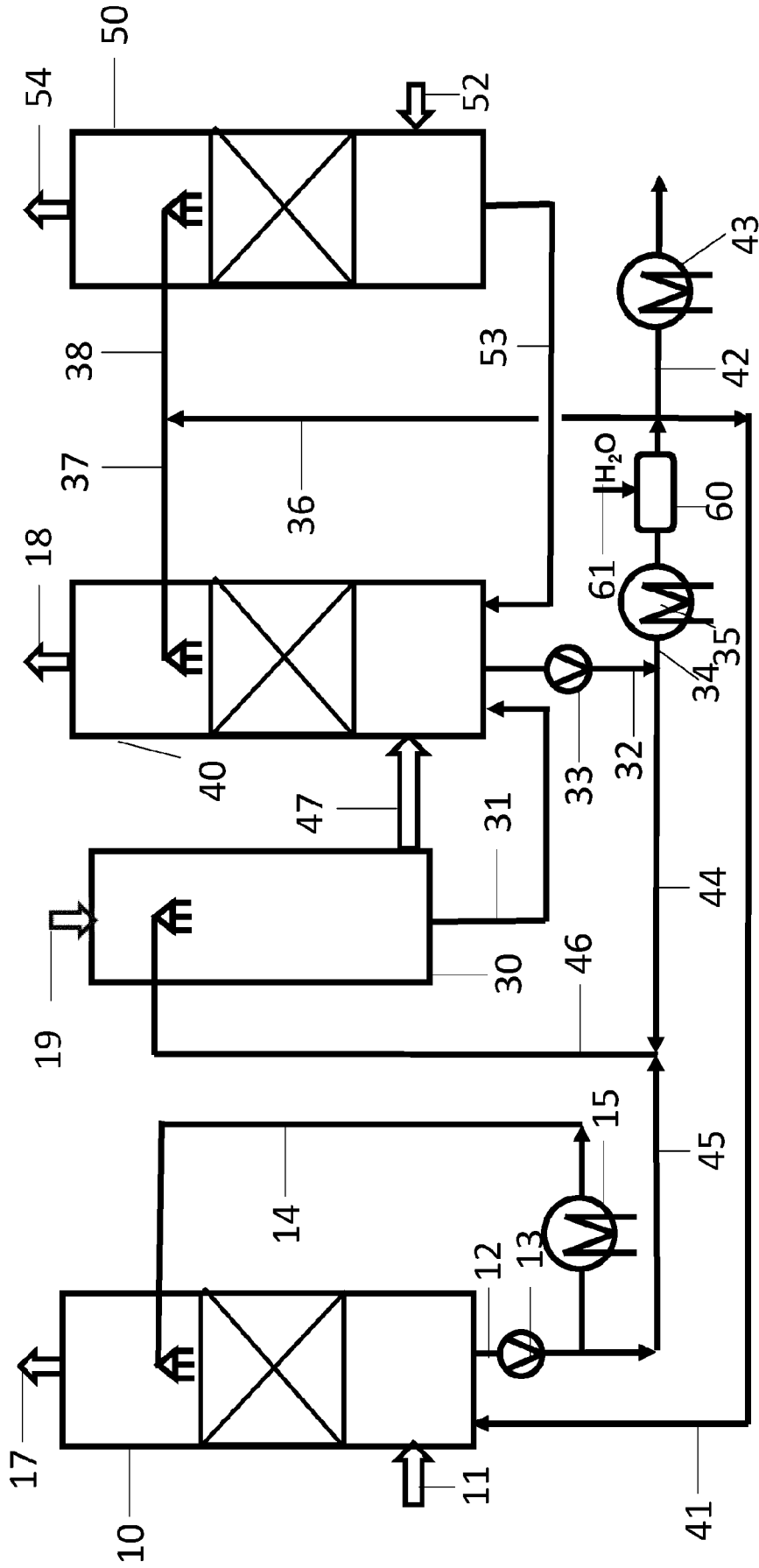


Fig. 6

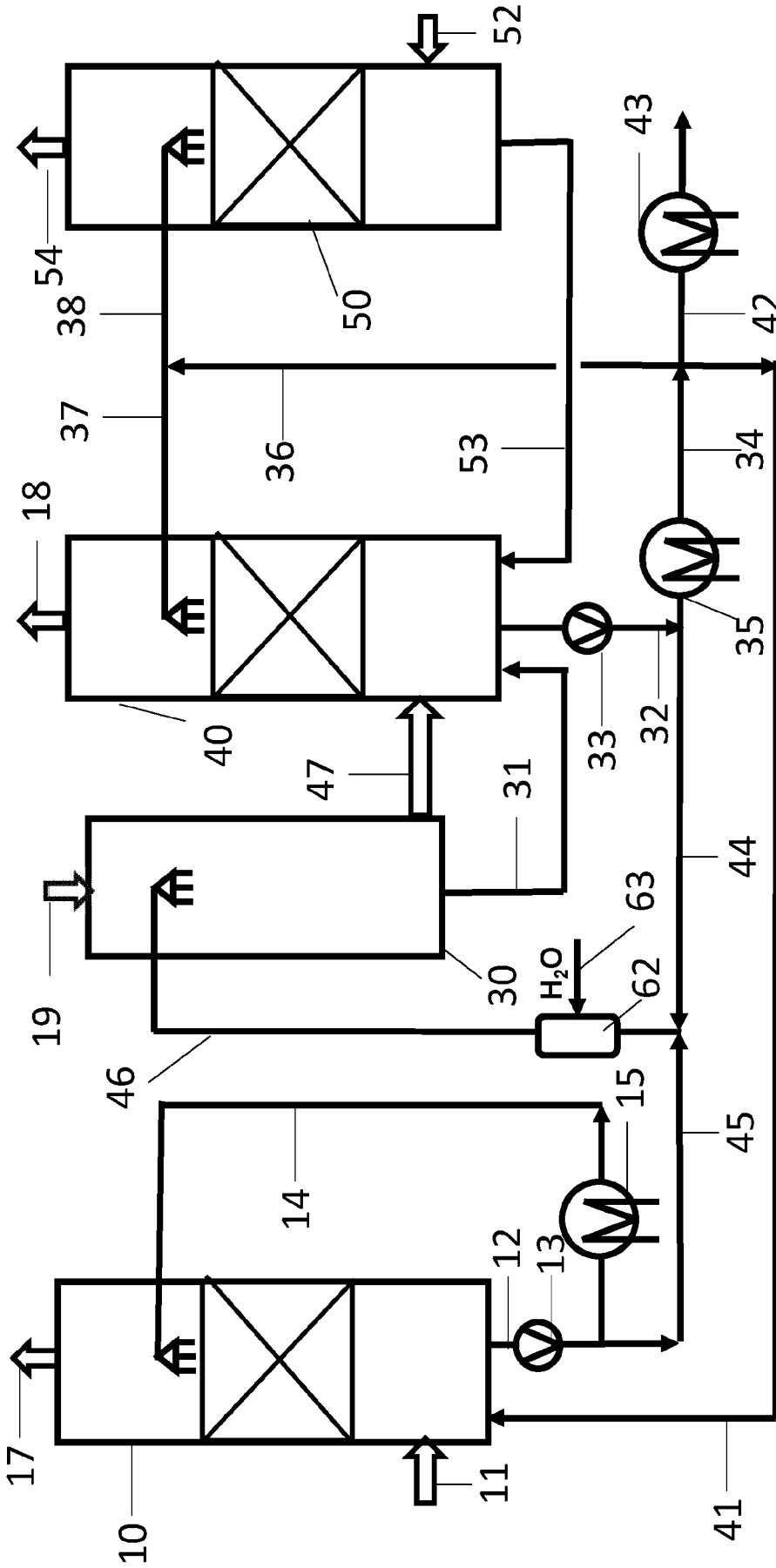


Fig. 7

INTERNATIONAL SEARCH REPORT

International application No  
PCT/EP2020/066982

A. CLASSIFICATION OF SUBJECT MATTER  
INV. C01B17/765 C01B17/80  
ADD.  
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED  
Minimum documentation searched (classification system followed by classification symbols)  
C01B  
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)  
EPO-Internal, CHEM ABS Data, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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A	WO 2009/065485 A2 (OUTOTEC OYJ [FI]; DAUM KARL-HEINZ [DE] ET AL.) 28 May 2009 (2009-05-28) cited in the application claims 1-19; figures 1-5	1-15
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Further documents are listed in the continuation of Box C.

See patent family annex.

\* Special categories of cited documents :

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- "&" document member of the same patent family

Date of the actual completion of the international search <b>11 March 2021</b>	Date of mailing of the international search report <b>23/03/2021</b>
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Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Authorized officer <b>Siebel, Eric</b>
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## INTERNATIONAL SEARCH REPORT

International application No  
PCT/EP2020/066982

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
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A	US 4 591 494 A (CAMERON GORDON M [CA] ET AL) 27 May 1986 (1986-05-27) claims 1-7; figure 1 -----	1-15



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International application No

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