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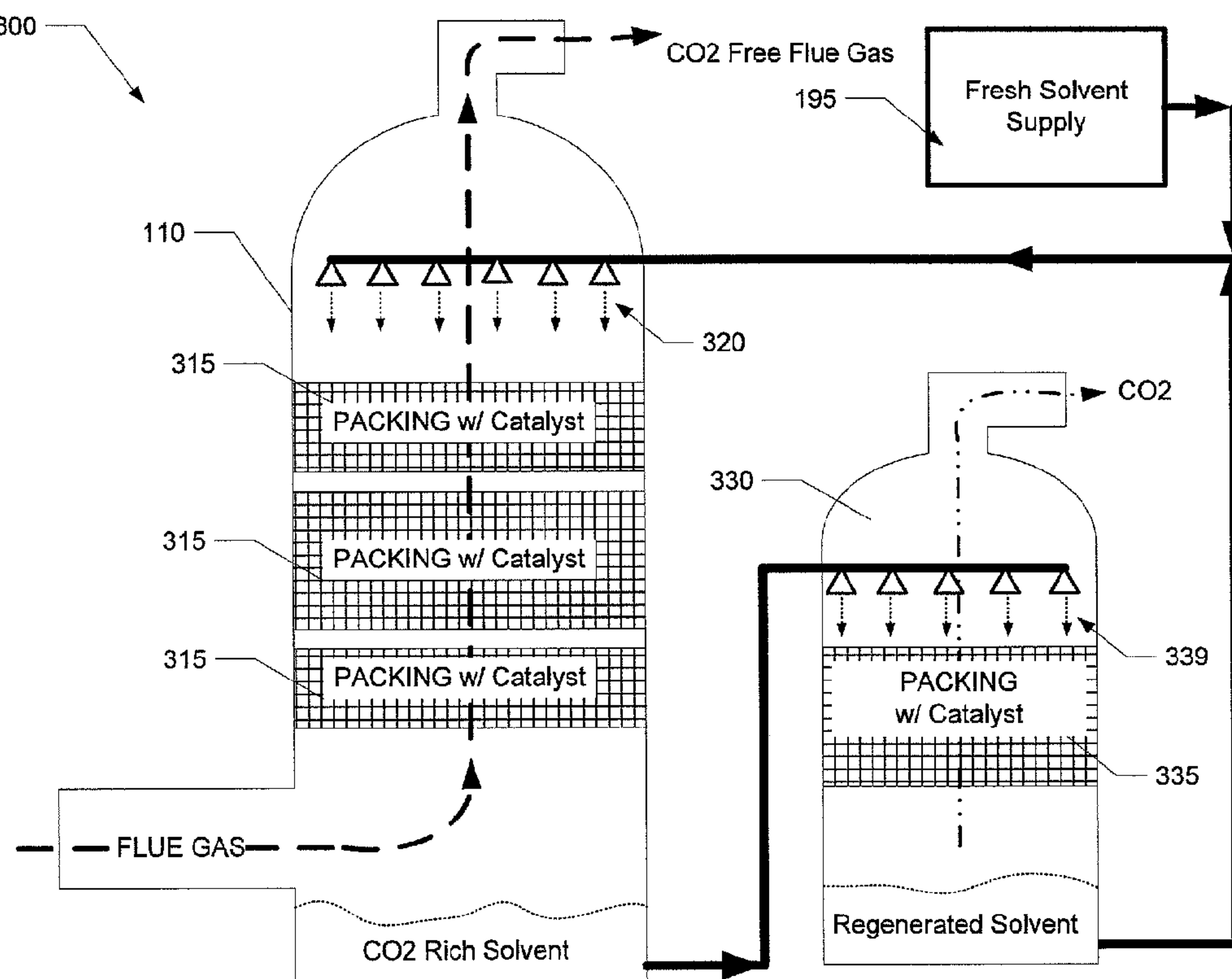
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GAZEUX MIXTE AU MOYEN D'UN CATALYSEUR

(54) Title: A SYSTEM AND METHOD FOR ENHANCED REMOVAL OF CO<sub>2</sub> FROM A MIXED GAS STREAM VIA USE  
OF A CATALYST



(57) Abrégé/Abstract:

The proposed invention is directed to a solvent based flue gas processing system for removing CO<sub>2</sub> from a flue gas stream. A catalyst is provided to increase the efficiency of the solvent in capturing CO<sub>2</sub> from the flue gas stream or in regenerating the solvent.

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12/406,289      18 March 2009 (18.03.2009)      US(71) Applicant (for all designated States except US): **ALSTOM TECHNOLOGY LTD** [CH/CH]; Brown Boveri Strasse 7, CH-5400 Baden (CH).(72) Inventor: **LIU, Zheng**; 11615 Laneborough Way #608, Knoxville, TN 37934 (US).

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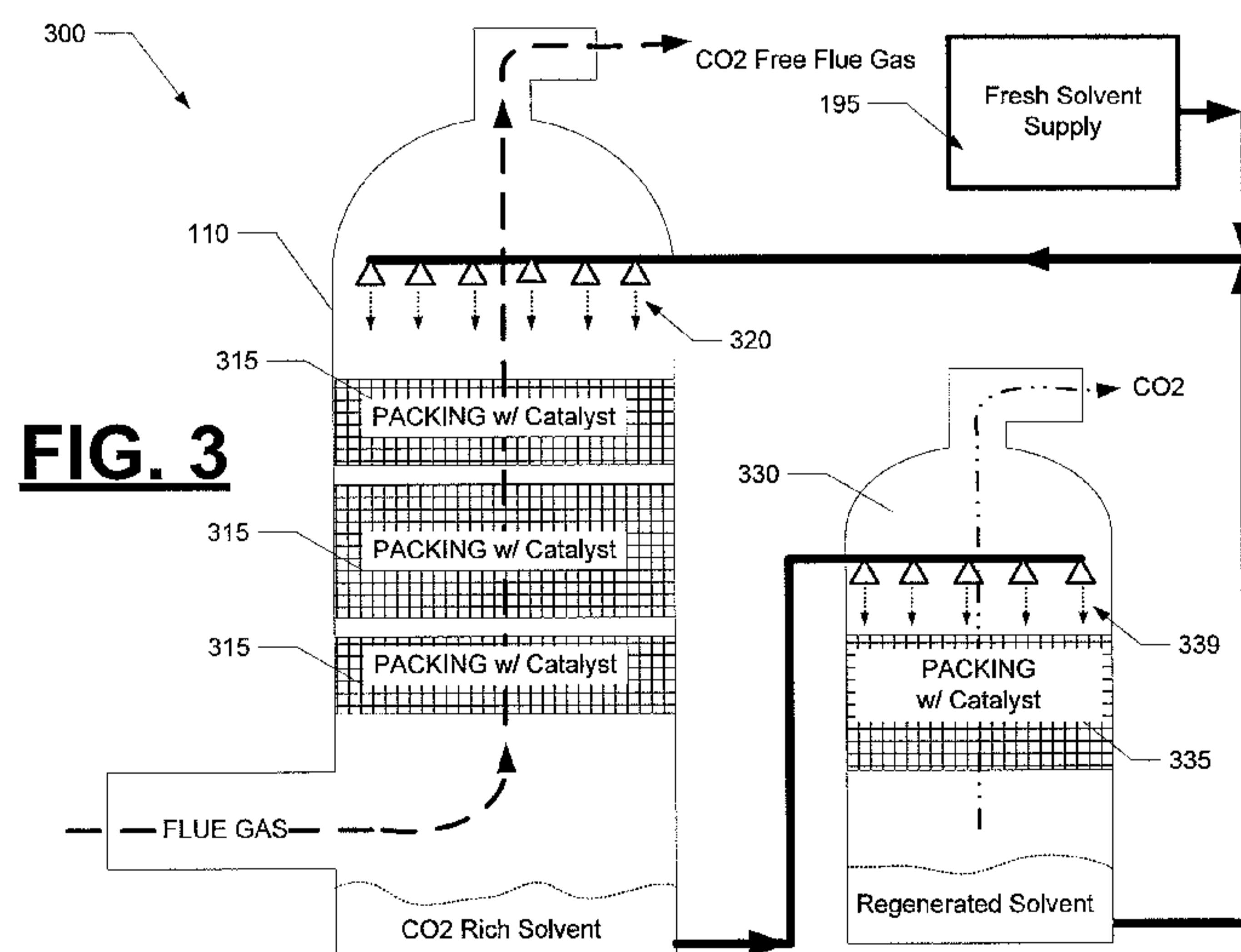
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(54) Title: A SYSTEM AND METHOD FOR ENHANCED REMOVAL OF CO<sub>2</sub> FROM A MIXED GAS STREAM VIA USE OF A CATALYST(57) Abstract: The proposed invention is directed to a solvent based flue gas processing system for removing CO<sub>2</sub> from a flue gas stream. A catalyst is provided to increase the efficiency of the solvent in capturing CO<sub>2</sub> from the flue gas stream or in regenerating the solvent.

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**A SYSTEM AND METHOD FOR ENHANCED REMOVAL OF CO<sub>2</sub>  
FROM A MIXED GAS STREAM VIA USE OF A CATALYST**

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**Field of the Invention**

The proposed invention relates to a system and method for removing carbon dioxide (CO<sub>2</sub>) from a process gas stream containing carbon dioxide and sulphur dioxide. More particularly, the proposed invention is directed to a solvent 10 based flue gas processing system for removing CO<sub>2</sub> from a flue gas stream in which a catalyst is provided to increase the efficiency of the solvent in capturing CO<sub>2</sub> from the flue gas stream or in regenerating the solvent.

**Background**

15 In the combustion of a fuel, such as coal, oil, peat, waste, etc., in a combustion plant, such as those associated with boiler systems for providing steam to a power plant, a hot process gas (or flue gas) is generated. Such a flue gas will often contain, among other things, carbon dioxide (CO<sub>2</sub>). The negative environmental effects of releasing carbon dioxide to the atmosphere have been widely recognised, and have resulted in the development of processes adapted 20 for removing carbon dioxide from the hot process gas generated in the combustion of the above mentioned fuels. One such system and process has previously been disclosed and is directed to a single-stage *Chilled Ammonia* based system and method for removal of carbon dioxide (CO<sub>2</sub>) from a post- 25 combustion flue gas stream.

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Known solvent based CO<sub>2</sub> capture systems, such as ammonia based systems and processes (CAP) provide a relatively low cost means for capturing/removing CO<sub>2</sub> from a gas stream, such as, for example, a post combustion flue gas stream. An example of such a system and process has 5 previously been disclosed in pending patent application PCT/US2005/012794 (International Publication Number: WO 2006/022885 / Inventor: Eli Gal), filed on 12 April 2005 and titled *Ultra Cleaning of Combustion Gas Including the Removal of CO<sub>2</sub>*. In this process the absorption of CO<sub>2</sub> from a flue gas stream is achieved by contacting a chilled ammonia ionic ammonia solution (or slurry) with a flue gas 10 stream that contains CO<sub>2</sub>.

FIG. 1A is a diagram generally depicting a flue gas processing system 15 for use in removing various pollutants from a flue gas stream FG emitted by the combustion chamber of a boiler system 26 used in a steam generator system of, for example, a power generation plant. This system includes a CO<sub>2</sub> removal 15 system 70 that is configured to remove CO<sub>2</sub> from the flue gas stream FG before emitting the cleaned flue gas stream to an exhaust stack 90 (or alternatively additional processing). It is also configured to output CO<sub>2</sub> removed from the flue gas stream FG. Details of CO<sub>2</sub> removal system 70 are generally depicted in FIG. 1B.

20 With reference to FIG. 1B, CO<sub>2</sub> removal System 70 includes a capture system 72 for capturing/removing CO<sub>2</sub> from a flue gas stream FG and a regeneration system 74 for regenerating ionic ammonia solution used to remove CO<sub>2</sub> from the flue gas stream FG. Details of capture system 72 are generally depicted in FIG. 1C.

25 With reference to FIG. 1C, a capture system 72 of a CO<sub>2</sub> capture system 70 (FIG. 1A) is generally depicted. In this system, the capture system 72 is a solvent based CO<sub>2</sub> capture system. More particularly, in this example, the solvent used is chilled ammonia. In a *chilled ammonia (CAP)* based system/method for CO<sub>2</sub> removal, an absorber vessel is provided in which 30 an absorbent ionic ammonia solution (ionic ammonia solution) is contacted with a flue gas stream (FG) containing CO<sub>2</sub>. The ionic ammonia solution is typically aqueous and may be composed of, for example, water and ammonium ions, bicarbonate ions, carbonate ions, and/or carbamate ions. An example of a

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known CAP CO<sub>2</sub> removal system is generally depicted in the diagrams of FIG. 1C.

With reference to FIG. 1C, an absorber vessel 110 is configured to receive a flue gas stream (FG) originating from, for example, the combustion chamber of a fossil fuel fired boiler 26 (see FIG. 1A). It is also configured to receive a lean ionic ammonia solution supply from regeneration system 74 (see FIG. 1B). The lean ionic ammonia solution is introduced into the vessel 110 via a liquid distribution system 121 while the flue gas stream FG is also received by the absorber vessel 110 via flue gas inlet 76.

The ionic ammonia solution is put into contact with the flue gas stream via a gas-liquid contacting device (hereinafter, mass transfer device, MTD) 111 used for contacting the flue gas stream with solvent and located in the absorber vessel 110 and within the path that the flue gas stream travels from its entrance via inlet 76 to the vessel exit 77. The gas-liquid contacting device 111 may be, for example, one or more commonly known structured or random packing materials, or a combination thereof.

Once contacted with the flue gas stream, the ionic ammonia solution acts to absorb CO<sub>2</sub> from the flue gas stream, thus making the ionic ammonia solution "rich" with CO<sub>2</sub> (rich solution). The rich ionic ammonia solution continues to flow downward through the mass transfer device and is then collected in the bottom of the absorber vessel 110. The rich ionic ammonia solution is then regenerated via regenerator system 74 (see FIG. 1B) to release the CO<sub>2</sub> absorbed by the ionic ammonia solution from the flue gas stream. The CO<sub>2</sub> released from the ionic ammonia solution may then be output to storage or other predetermined uses/purposes. Once the CO<sub>2</sub> is released from the ionic ammonia solution, the ionic ammonia solution is said to be "lean". The lean ionic ammonia solution is then again ready to absorb CO<sub>2</sub> from a flue gas stream and may be directed back to the liquid distribution system 121 whereby it is again introduced into the absorber vessel 110. Details of regenerating system 74 are shown in FIG. 1C. System 74 includes a regenerator vessel 130. Regenerator vessel 130 is configured to receive a rich solution feed from the capture system 72 and to return a lean solution feed to the capture system 72 once CO<sub>2</sub> has been separated from the rich solution.

During the regeneration process, the rich ionic ammonia solution is heated so that CO<sub>2</sub> contained in the solution separates from the chilled ammonia solution. Once separated from the CO<sub>2</sub>, ammonia (ammonia slip) is returned to the capture system for use in capturing further CO<sub>2</sub> from a gas stream.

These currently known solvent based CO<sub>2</sub> capture technologies typically consume approximately 20-30% of the power generated by the power generation system in order for the CO<sub>2</sub> capture process to work effectively. In addition, these technologies often require a large portion of thermal energy generated by boiler/re-boiler functions (reboiler duty) in order to regenerate amine solution for re-use in capturing CO<sub>2</sub> from a flue gas stream. In short, while there are known technologies for capturing CO<sub>2</sub> from a flue gas stream, they require immense amounts of energy in order to function well. Further, in order to maximize/optimize the amount of time that flue gas is in contact with amine, the physical size of the absorber and/or re-generator tanks in a typical system must be very large. The cost to design and implement these towers of such large scale is very high. Additionally, the physical space that is required on-site to accommodate these vessels is significant. Where on-site space is limited, additional steps must be taken to implement the vessels/system in the limited space, if possible.

20

### Summary Of the Invention

Embodiments of the present invention provide for flue gas processing system for use with a fossil fuel fired boiler. Briefly described, in architecture, one embodiment of the system, among others, can be implemented as a flue gas processing system that includes an absorber tower configured to receive a mixed gas stream containing carbon dioxide and to contact it with a solvent; and the absorber tower comprises packing materials that are coated with a catalyst.

Embodiments of the present invention can also be viewed as providing methods for processing a mixed gas stream wherein the method includes the steps of: receiving a flue gas stream from the combustion chamber of a boiler; contacting the flue gas stream with a solvent and contacting the solvent with a catalyst.

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Other systems, methods, features, and advantages of the present invention will be or become apparent to one with skill in the art upon examination of the following drawings and detailed description. It is intended that all such additional systems, methods, features, and advantages be included within this description, be 5 within the scope of the present invention, and be protected by the accompanying claims.

According to one aspect of the present invention, there is provided a gas processing system comprising: an absorber tower configured to receive a mixed gas stream containing carbon dioxide and to receive a solvent and a catalyst for 10 contact between said mixed gas stream and said solvent in the presence of the catalyst, whereby the catalyst promotes capture of carbon dioxide from said mixed gas stream by said solvent to produce a carbon dioxide rich solvent.

According to another aspect of the present invention, there is provided a process for removing carbon dioxide from a mixed gas comprising: passing a mixed 15 gas containing carbon dioxide through an absorber tower configured to receive a solvent and a catalyst for contact of the solvent with the mixed gas in the presence of the catalyst, whereby the catalyst promotes carbon dioxide capture from the mixed gas by the solvent to produce a carbon dioxide rich solvent.

#### Brief Description of the Drawings

20 The present invention may be better understood and its numerous objects and advantages will become apparent to those skilled in the art by reference to the accompanying drawings in which:

FIG. 1A - FIG. 1C are diagrams generally depicting a typical flue gas processing system 15 with provisions for CO<sub>2</sub> removal.

25 FIG. 2A - FIG. 2C are diagrams generally illustrating an example of packing materials 315 on which a catalyst has been immobilized on the wall/surfaces of the packing materials.

5 FIG. 3 is a diagram generally depicting relevant portions of an amine or ammonia based CO<sub>2</sub> capture system 70 in which absorber 110 and regenerator 330 are provided with packing materials 315 and 335, respectively that have been coated with a catalyst.

10 FIG. 4A is a diagram generally depicting relevant portions of an amine or ammonia based CO<sub>2</sub> capture system 70 in which a catalyst 425 is added to the solvent supply 195 (FIG. 4A) or alternatively it may be added directly into the solvent supply feed. In this example, the solvent is amine or ammonia.

15 FIG. 4B is a diagram generally depicting relevant portions of an amine or ammonia based CO<sub>2</sub> capture system 70 in which a catalyst 425 is added directly into the solvent supply feed.

### Discussion

15 The proposed invention is directed to increasing CO<sub>2</sub> capture efficiency in a solvent based capture system for use in processing, for example, a flue gas stream. The proposed invention is also directed to efficiently regenerating a solvent used to capture CO<sub>2</sub>.

In one embodiment of the proposed invention a catalyst 425 that is coated (or immobilized) on one or more surfaces of the packing materials 315 located in the absorber tower 110 of an amine or ammonia based CO<sub>2</sub> capture system 70. FIG. 2A - FIG. 2C are diagrams generally depicting packing materials 315 that have been coated with a catalyst 425. FIG. 2B and 2C shows that the packing materials 315 may be composed of, for example, a series of corrugated support structures 320 arranged in close proximity to each other so as to form a series of channels 334 through which flue gas entering the absorber vessel 110 flow. The catalyst 425 is coated on one or more surfaces of each corrugated support structures 320. The corrugated troughs 334 form pathways for the flue gas to pass through the packing materials 315 thereby contacting the catalyst layer 425.

The catalyst 425 may be either a homogeneous and/or a heterogeneous catalyst. Homogeneous type catalyst may include, for example, organo-metallic complex composed of transition metal and inorganic/organic ligands (such as bi-pyridine, aromatics, halogen, etc). Formation of the transition metal complex is one of the most powerful and universal ways of activating inert molecules. Coordination results in a change of reactivity of the ligands and creates favourable steric conditions for ligand interaction. This activation via coordination enables one to carry out numerous catalytic reactions under the influence of transition metal compounds. Other examples of catalyst that may be used include, but are not limited to, for example, organo-metallic complex catalysts, such as, for example, halogen-type complex, Ni(bpy)<sub>3</sub>Cl<sub>2</sub> [bpy: bipyridine].

Heterogeneous type catalyst may include, for example, metals or their compounds such as oxides (e.g. MgO), chloride MgCl<sub>2</sub>, etc. (Oxide preferably will not include sulphide and chloride. They are independent of each other) that are dispersed, preferably evenly, on solid materials with a high BET surface area, such as, for example, polymer, metal oxides, SiO<sub>2</sub>, molecular sieves, base- and/or acid- modified clay, etc.

The catalyst 425 is provided to promote the reaction of CO<sub>2</sub> with a solvent such as amine or chilled ammonia, thus resulting in the solvent being able to capture more CO<sub>2</sub> per given period of time (i.e. enhanced amine-based CO<sub>2</sub> capture efficiency).

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Catalyst on Packing Materials in Regeneration Tower - In one embodiment of the proposed invention, a catalyst is coated/immobilized on one or more surfaces of the packing materials in the regeneration tower of an amine or ammonia based CO<sub>2</sub> capture system. The catalyst may be either a 5 homogeneous and/or a heterogeneous catalyst. The catalyst is provided to promote the regeneration of solvent (ex: amine or ammonia) after it has reacted/interacted with CO<sub>2</sub>. The catalyst is selected and provided to promote the decomposition of products formed from the interaction/reaction between CO<sub>2</sub> and amine, thus resulting in the amine being able to regenerate more efficiently 10 from the rich amine solution per given period of time (i.e. enhanced amine-based CO<sub>2</sub> capture efficiency). This embodiment is generally depicted in FIG. 3.

In a further embodiment, a catalyst is added to a solvent, such as, for example, amine or ammonia, that is used to capture CO<sub>2</sub> from a flue gas stream. The catalyst may also be used to promote the regeneration of solvent after it has 15 reacted with CO<sub>2</sub>. In this embodiment, a homogenous catalyst may be used. This embodiment is generally depicted in FIG. 4A and FIG. 4B. FIG. 4A illustrates how the catalyst 425 can be fed directly to the solvent supply tank 195. FIG. 4B illustrates how the catalyst 425 can be combined with the solvent supply that is feeding the liquid distribution system 121.

20 In a flue gas processing system 15 wherein a catalyst 425 is coated/immobilized/layered on packing materials 315 in both the absorber tower 110 and the regeneration tower 330, the catalyst 425 used in both the absorber tower 110 and the regeneration tower 330 may be the same catalyst or different catalysts. It is not necessary that the same catalyst be used in connection with 25 both capture and regeneration.

The catalyst 425 may be dispersed/layered/coated onto the solid material 320 with a high BET surface area, such as for example, 100-1000 m<sup>2</sup>/g, square meter per gram solid materials, via, for example, a wet impregnation, followed by being coated/immobilized onto the packing materials 315 located in, for example, 30 the Absorber tower 110 and/or regeneration tower/stripper 330. This may be accomplished via, for example, known wash-coating techniques, such as those employed in, for example, industrial catalyst preparation processes. These solid materials may be used to make up the packing materials or parts thereof.

Alternately, catalysts (organo-metallic complex, transition metal as well as its salt) may be directly coated onto the packing materials 315 (see FIG. 2A – FIG. 2C) to form a catalyst film (or layer). This may be accomplished via, for example, known coating technologies such as those employed in, for example, 5 industrial semiconductor preparation or self-assembly process or electrochemical coating processes. Some examples of catalysts that may be used include, but are not limited to, for example, organo-metallic complex catalysts, such as, for example, halogen-type complex,  $\text{Ni}(\text{bpy})_3\text{Cl}_2$  [bpy: bipyridine] and transition metal-based inorganic catalysts.

10 Any homogeneous and/or heterogeneous catalysts can be used in  $\text{CO}_2$  capture operations to activate  $\text{CO}_2$  adsorbed onto the surface of the catalyst 425 and catalyse carbonation/bi-carbonation and carbamation of amine with  $\text{CO}_2$  via homogeneous and/or heterogeneous processes. Also these catalysts can be used in solvent (example: amine or ammonia) regeneration to catalyse the 15 decomposition of products formed from the carbonation/bi-carbonation and carbamation of amine with  $\text{CO}_2$ .

While the invention has been described with reference to a number of preferred embodiments, it will be understood by those skilled in the art that various changes may be made and equivalents may be substituted for elements 20 thereof without departing from the scope of the invention. In addition, many modifications may be made to adapt a particular situation or material to the teachings of the invention without departing from the essential scope thereof. Therefore, it is intended that the invention not be limited to the particular 25 embodiments disclosed as the best mode contemplated for carrying out this invention, but that the invention will include all embodiments falling within the scope of the appended claims. Moreover, the use of the terms first, second, etc. do not denote any order or importance, but rather the terms first, second, etc. are used to distinguish one element from another.

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CLAIMS:

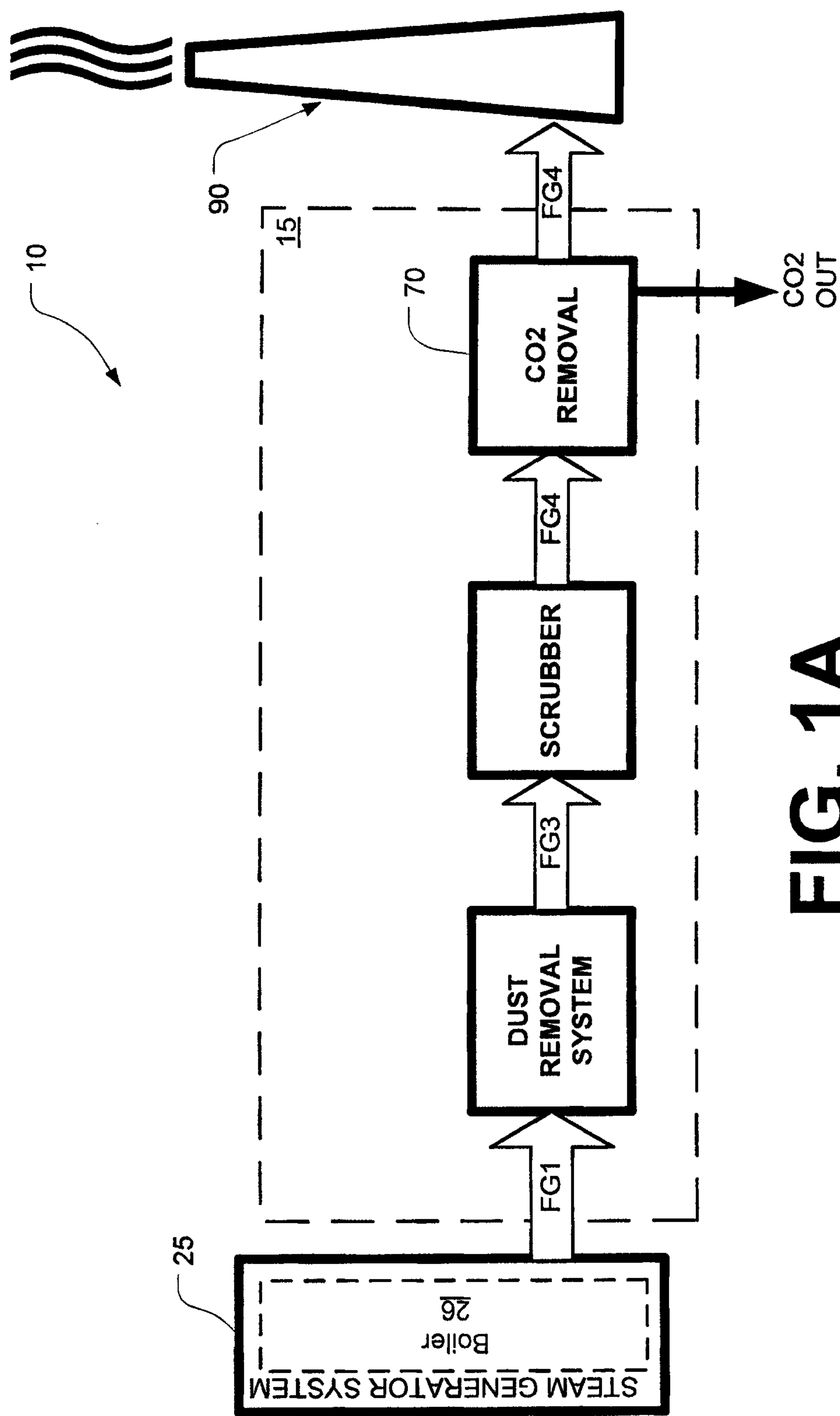
1. A gas processing system comprising:
  - an absorber tower configured to receive a mixed gas stream containing carbon dioxide and to receive a solvent and a catalyst for contact between said mixed gas stream and said solvent in the presence of the catalyst, whereby the catalyst promotes capture of carbon dioxide from said mixed gas stream by said solvent to produce a carbon dioxide rich solvent.
2. The system of claim 1, wherein the catalyst is a homogeneous catalyst.
3. The system of claim 1, wherein the catalyst is a heterogeneous catalyst.
4. The system of claim 1, wherein the system further includes a liquid distribution system that provides a solvent and catalyst mixture to the absorber tower.
5. The system of claim 1, wherein said solvent is amine based.
6. The system of claim 1, wherein said solvent is ammonia based.
7. The system of claim 1, wherein the catalyst is provided to a tank containing the supply of solvent.
8. The system of claim 1, wherein the catalyst is combined with the solvent flowing through a solvent distribution system.
9. The system of claim 1, wherein said solvent is a carbon dioxide lean solvent cooled prior to contact with said mixed gas stream.
10. The system of claim 1, further comprising a regeneration tower configured to receive said carbon dioxide rich solvent for contact with a catalyst to promote carbamation and to produce a carbon dioxide lean solvent.
11. The system of claim 10, wherein the carbon dioxide rich solvent is heated prior to contact with the regeneration tower catalyst.

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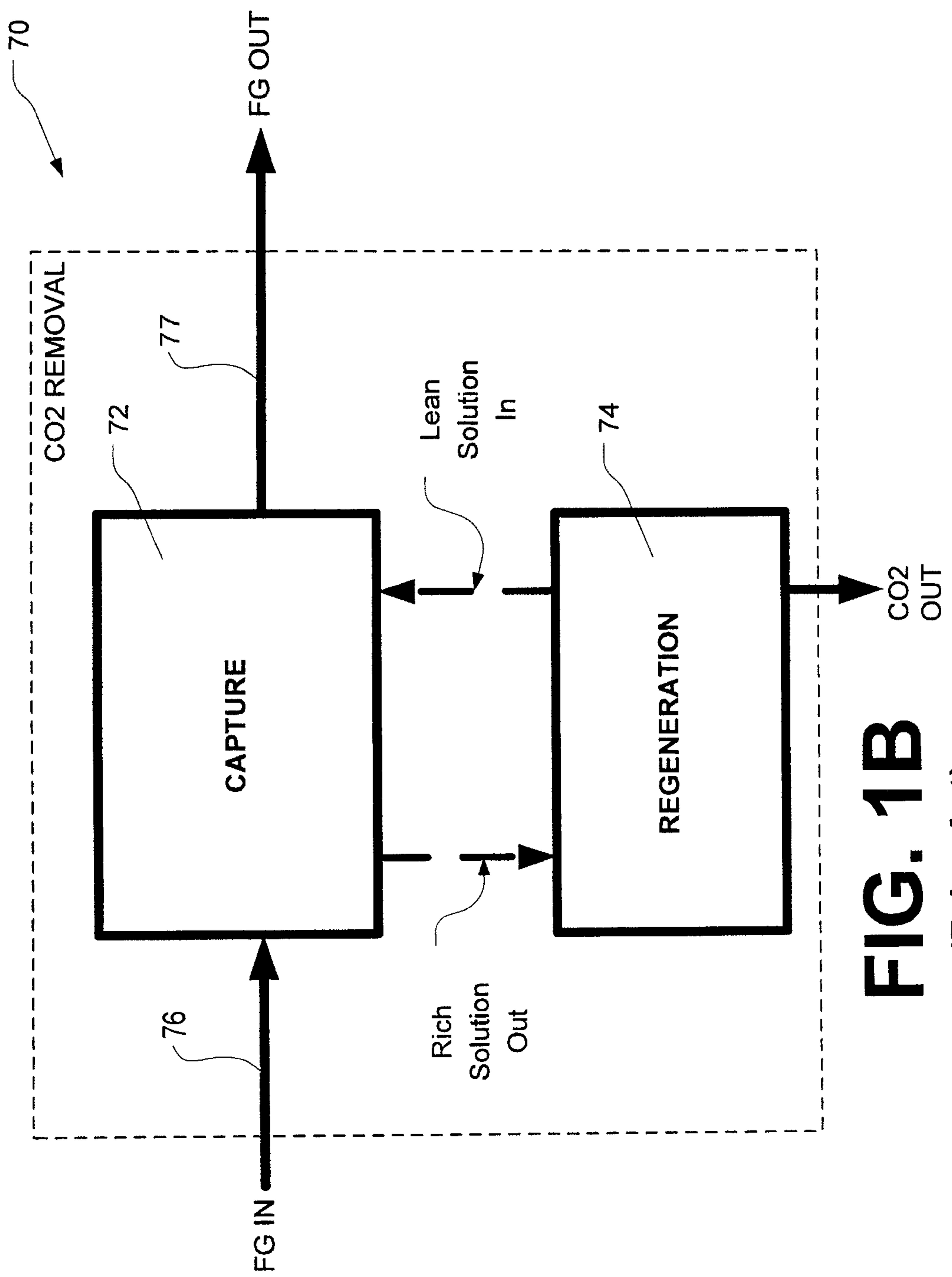
12. The system of claim 10, wherein the absorber tower catalyst and the regeneration tower catalyst may be the same or different catalysts.
13. The system of claim 1, wherein the mixed gas stream is a flue gas stream.
- 5 14. A process for removing carbon dioxide from a mixed gas comprising:  
passing a mixed gas containing carbon dioxide through an absorber tower configured to receive a solvent and a catalyst for contact of the solvent with the mixed gas in the presence of the catalyst, whereby the catalyst promotes carbon dioxide capture from the mixed gas by the solvent to produce a carbon dioxide rich solvent.
- 10 15. The process of claim 14, wherein the catalyst is a homogeneous catalyst.
16. The process of claim 14, wherein the catalyst is a heterogeneous catalyst.
- 15 17. The process of claim 14, wherein the process further includes providing a mixture of the solvent and the catalyst to a liquid distribution system to provide the mixture to the absorber tower.
18. The process of claim 14, wherein the solvent is amine based.
19. The process of claim 14, wherein the solvent is ammonia based.
- 20 20. The process of claim 14, further including providing the catalyst to a tank containing the solvent.
21. The process of claim 14, further including combining the catalyst with the solvent flowing through a solvent distribution system.

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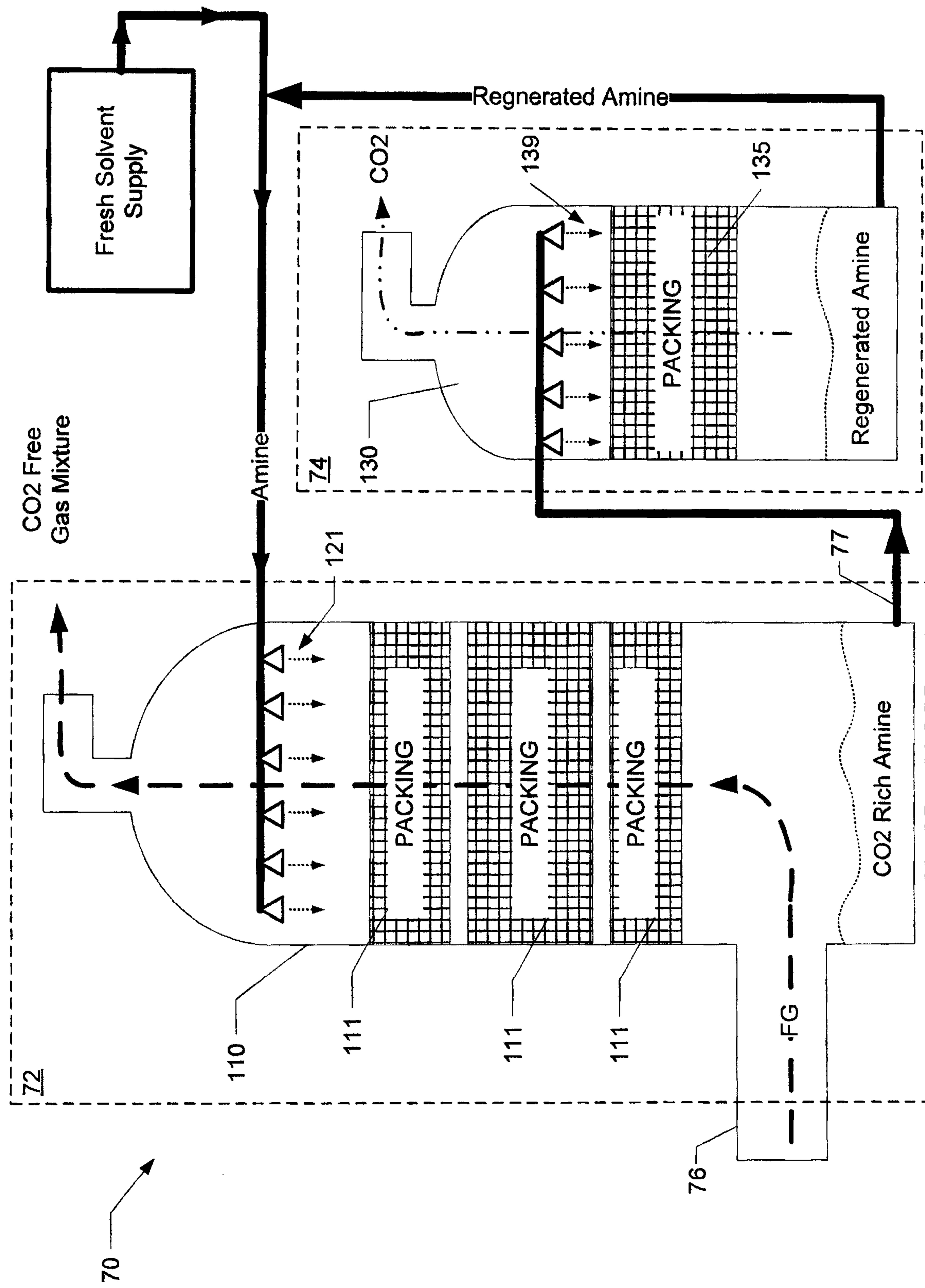
22. The process of claim 14, wherein the absorber tower is configured to prolong contact between the mixed gas and the solvent.
23. The process of claim 14, wherein the solvent is cooled prior contact with the mixed gas.
- 5 24. The process of claim 14, wherein the solvent is a carbon dioxide lean solvent.
25. The process of claim 14, further comprising a regeneration tower configured to receive the carbon dioxide rich solvent for contact with a catalyst to promote carbamation of the carbon dioxide rich solvent to produce a carbon dioxide 10 lean solvent.
26. The process of claim 25, wherein the carbon dioxide rich solvent is heated prior to or within said regeneration tower.
27. The process of claim 25, wherein said carbon dioxide lean solvent may be circulated to the absorber tower.
- 15 28. The process of claim 25, wherein said regeneration tower catalyst may be the same or different from the absorber tower catalyst.
29. The process of claim 24, wherein the mixed gas stream is a flue gas stream.



**FIG. 1A**  
(Prior Art)

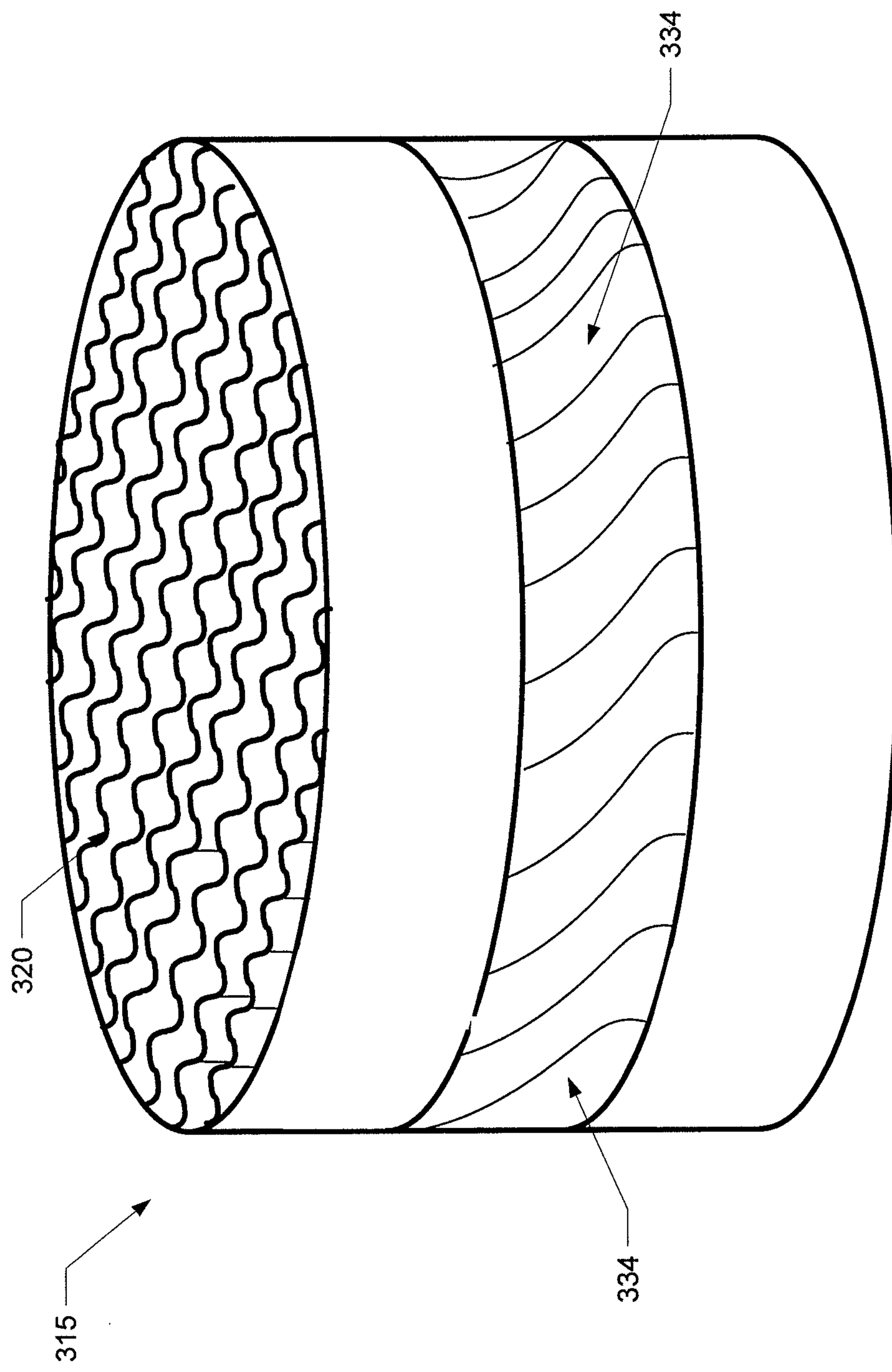


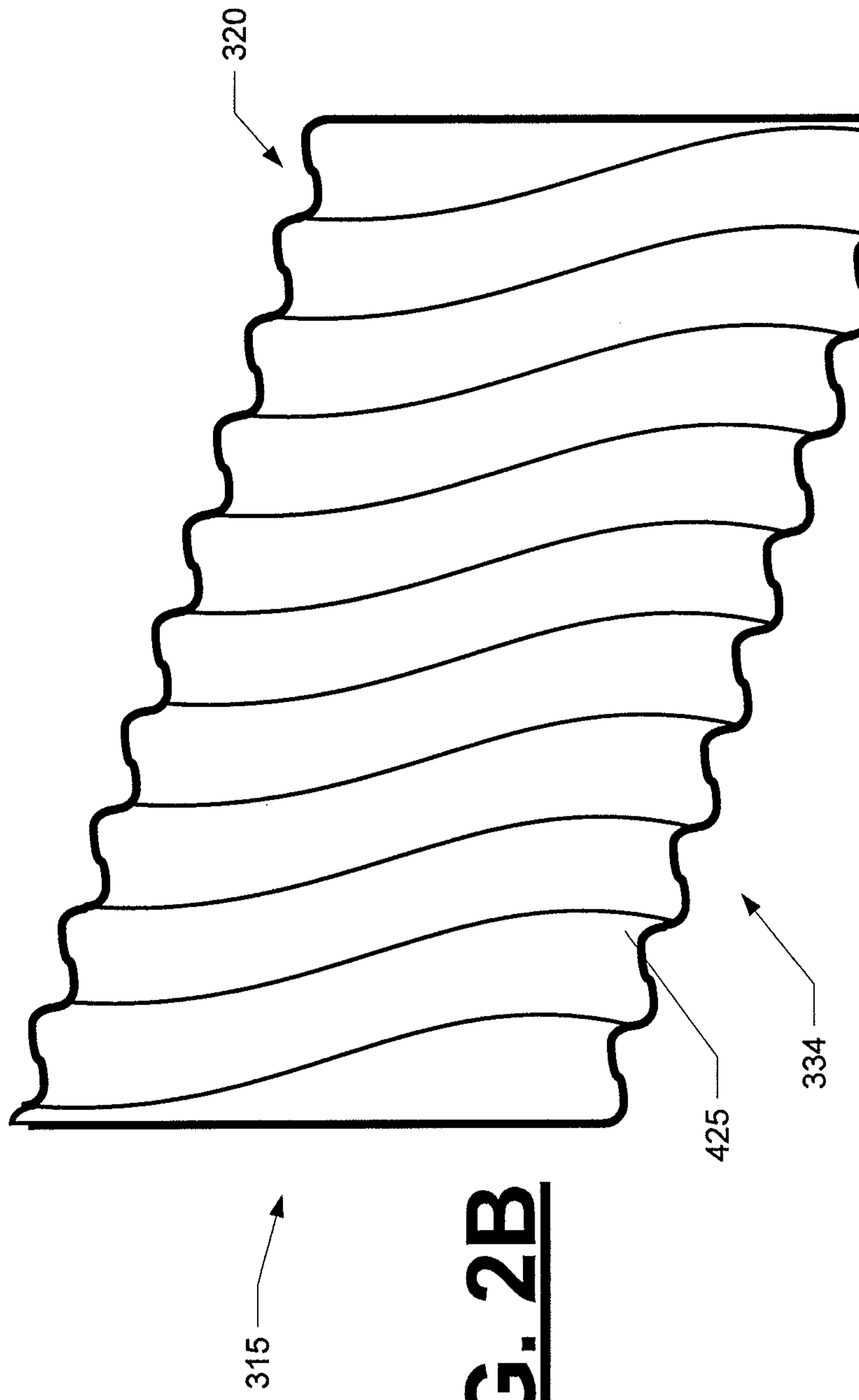
**FIG. 1B**  
(Prior Art)



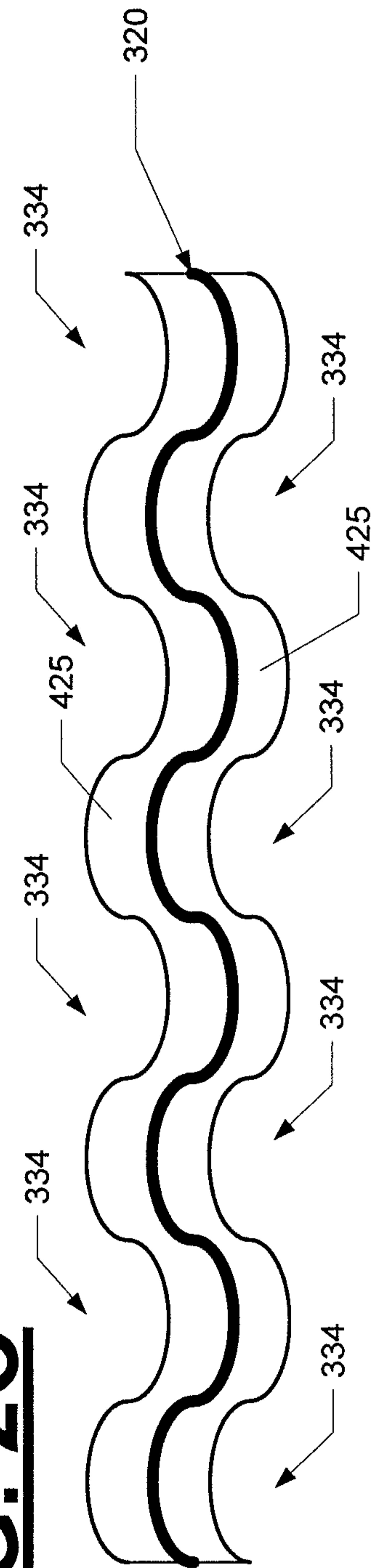
**FIG. 1C**  
(Prior Art)

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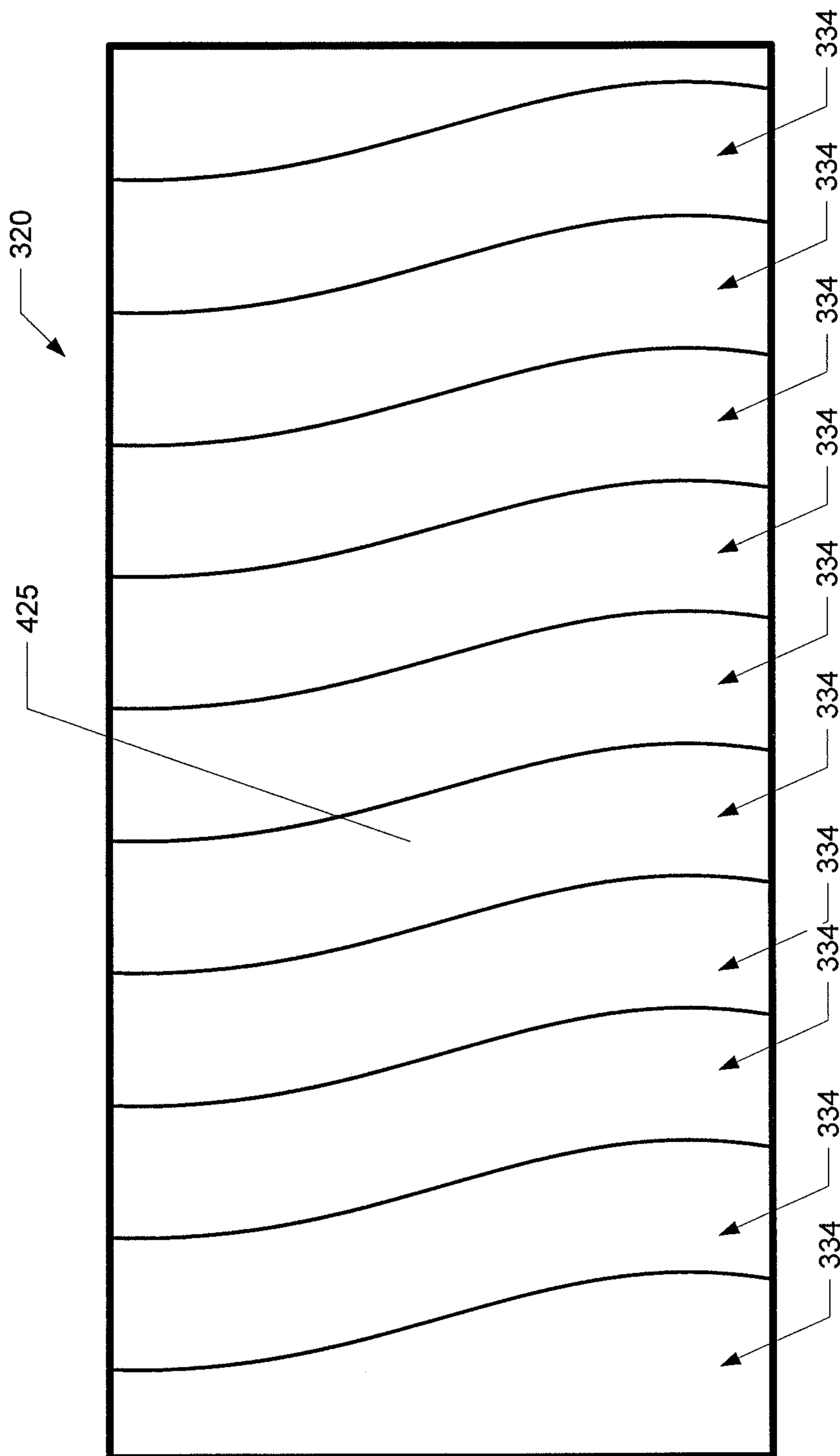
**FIG. 2A**



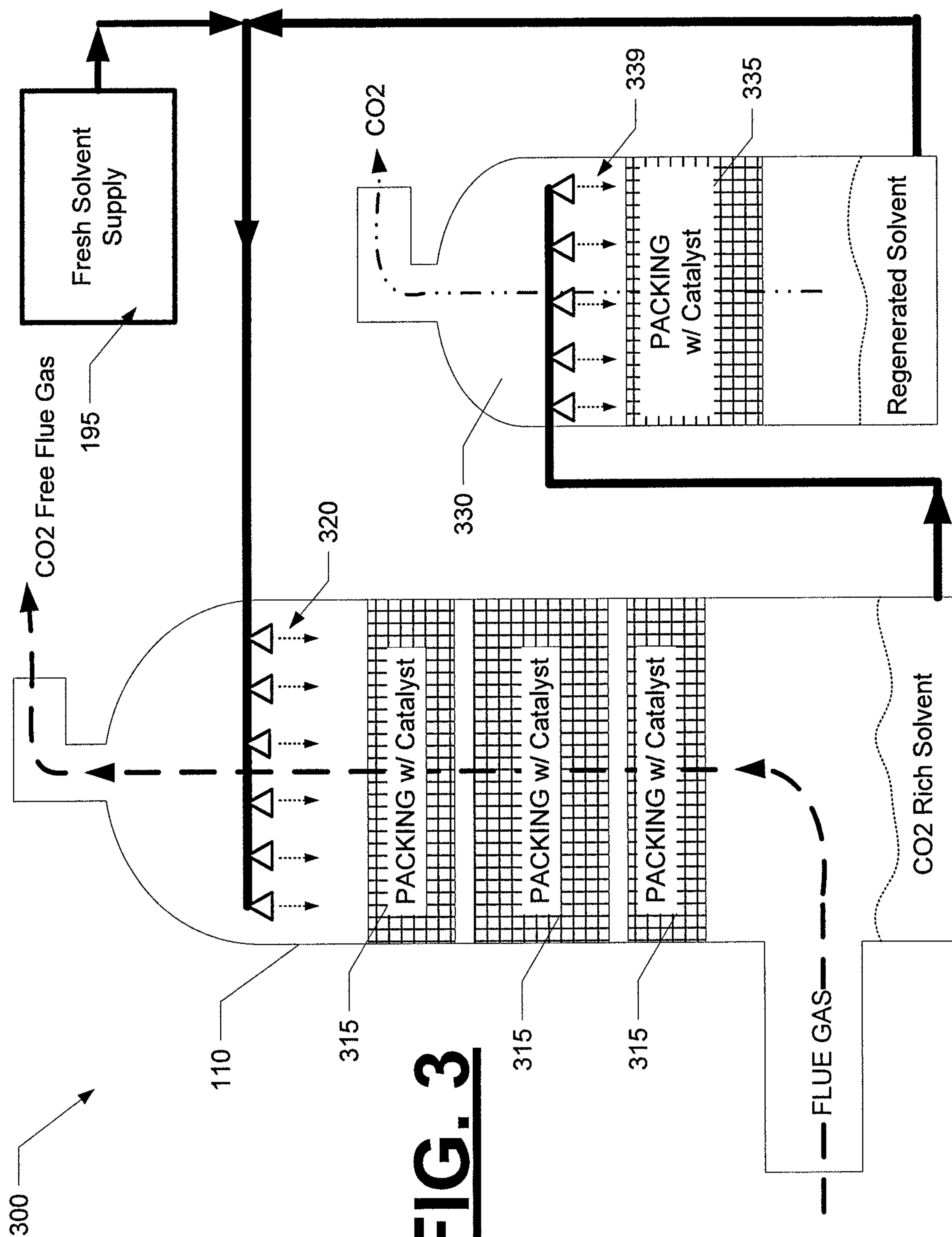
**FIG. 2B**

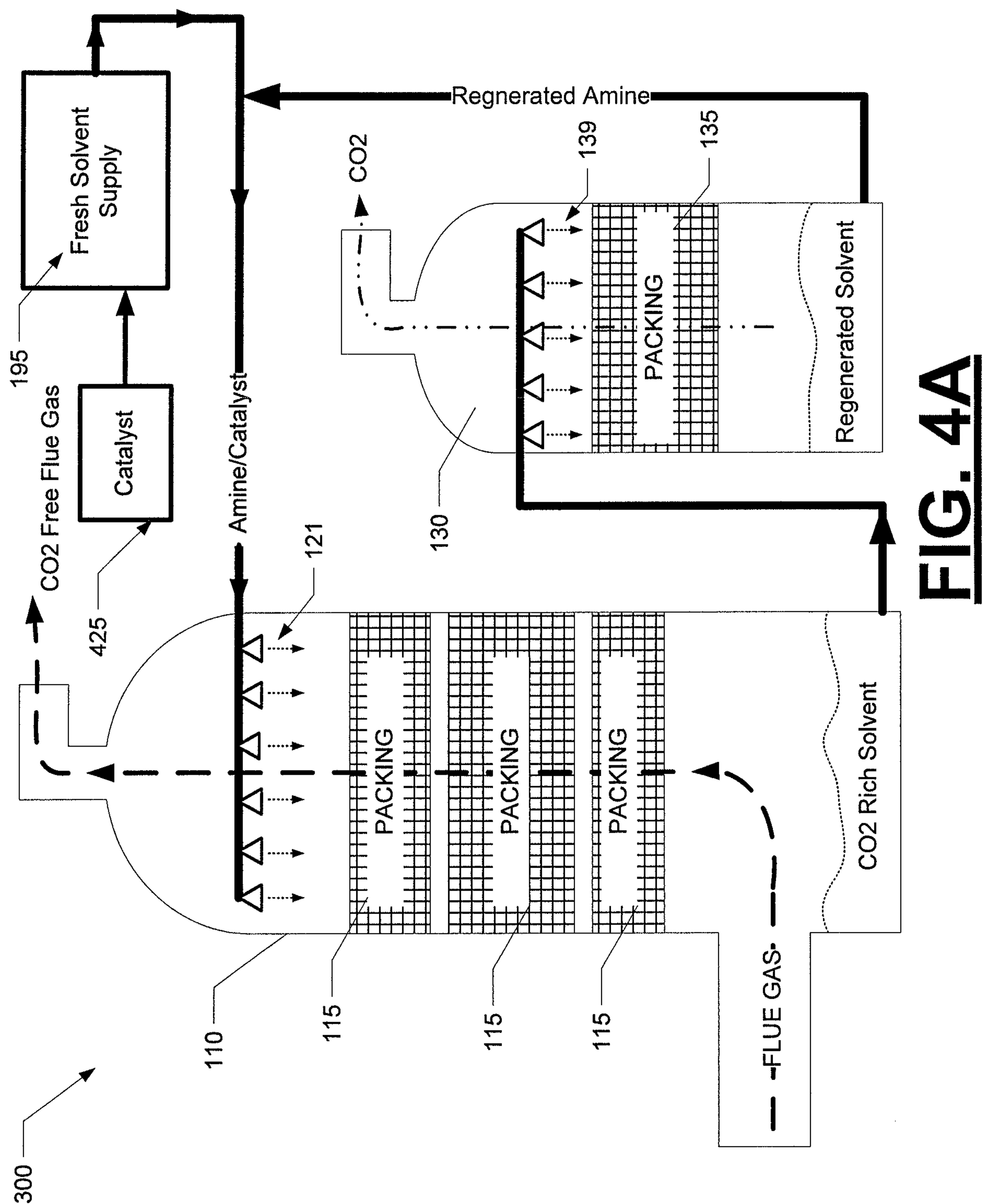


**FIG. 2C**

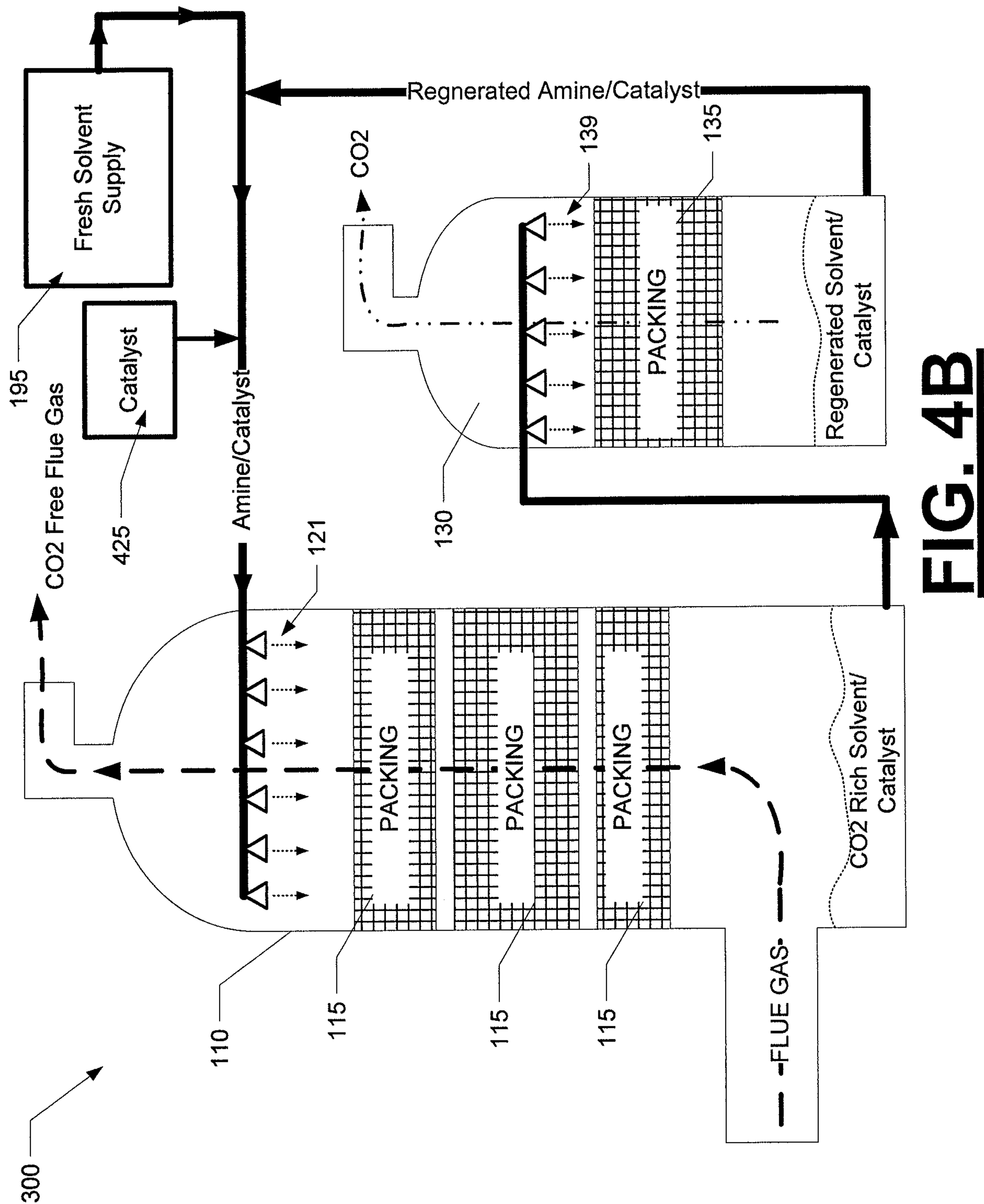


**FIG. 2D**





# 44. G. H.

**FIG. 4B**

