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### (54) THERMOCHEMICAL ENERGY STORAGE SYSTEM

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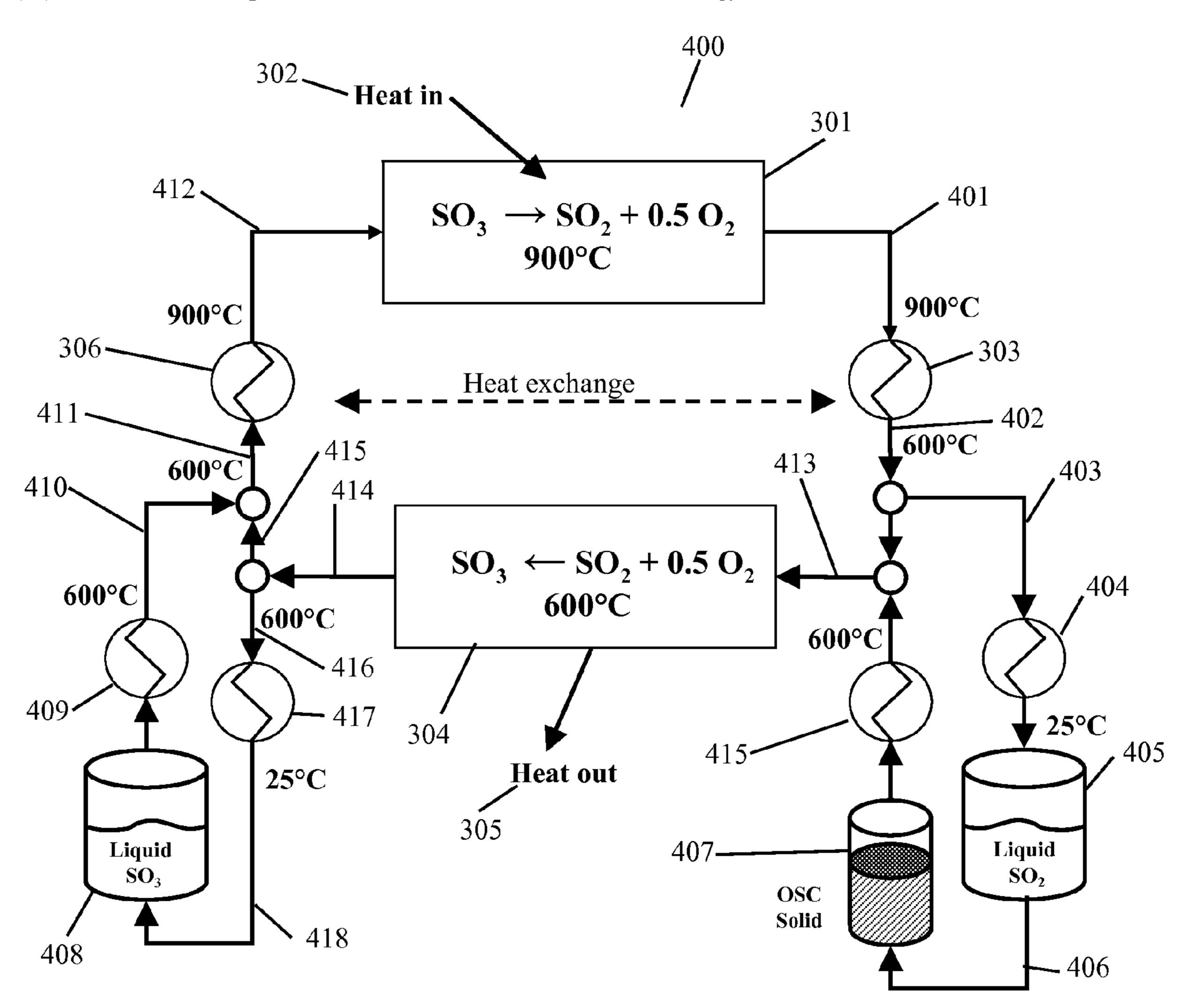
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#### **Publication Classification**

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- (57) ABSTRACT

A system is described for the thermochemical capture of heat energy and the transfer of this energy to a point of use using a cycle decomposing SO<sub>3</sub> to SO<sub>2</sub> and O<sub>2</sub> and the subsequent oxidation of SO<sub>2</sub> and O<sub>2</sub> to SO<sub>3</sub>. This system can store this energy in the form of chemical energy by storing liquid SO<sub>2</sub>. Embodiments are described wherein oxygen is stored by a solid oxygen storage material or removed and added to the process by selective membranes or by electrochemical pumping. In addition, an alternative embodiment uses an electrochemical generator for the direct conversion of SO<sub>2</sub> to electrical energy.



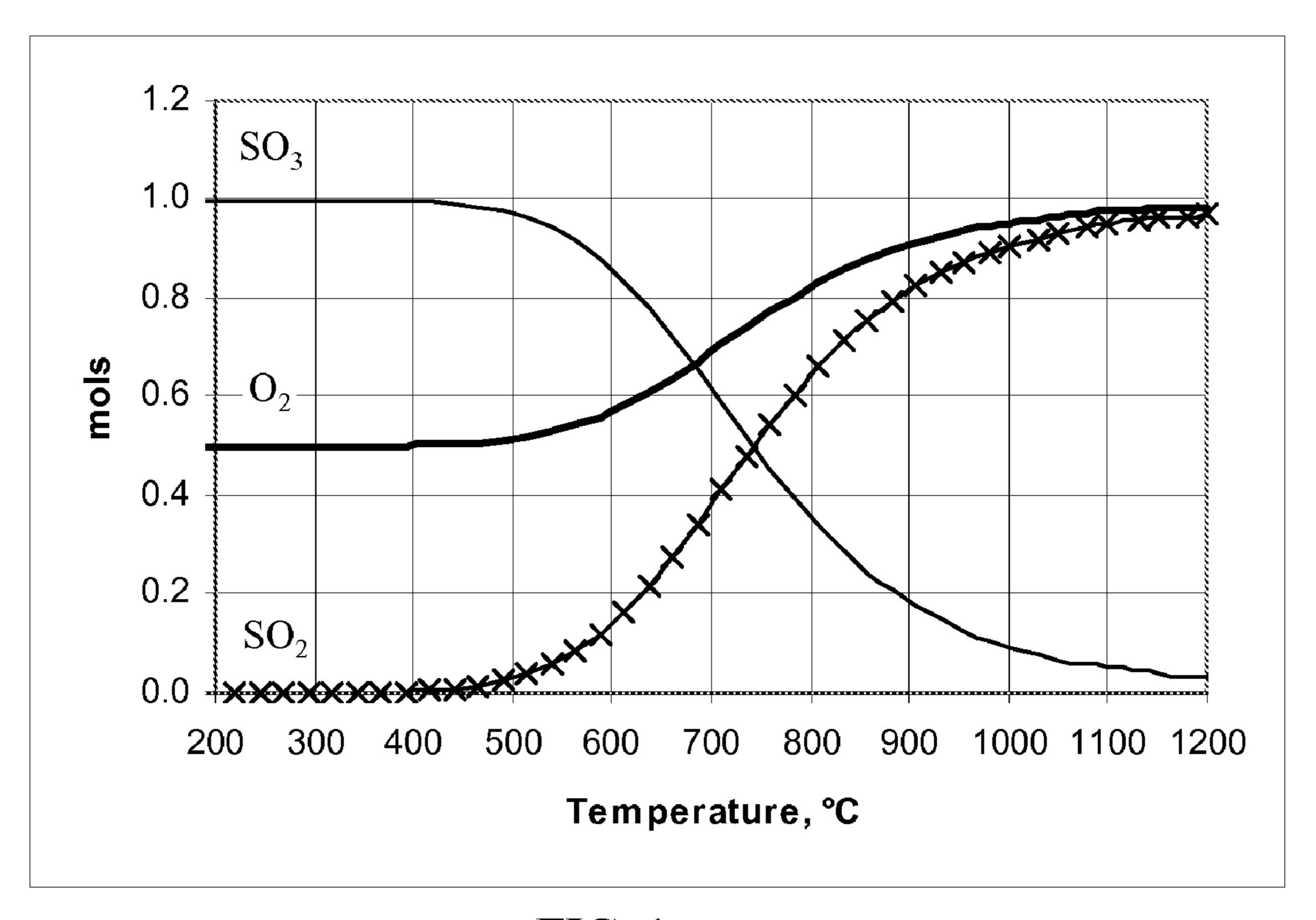


FIG. 1

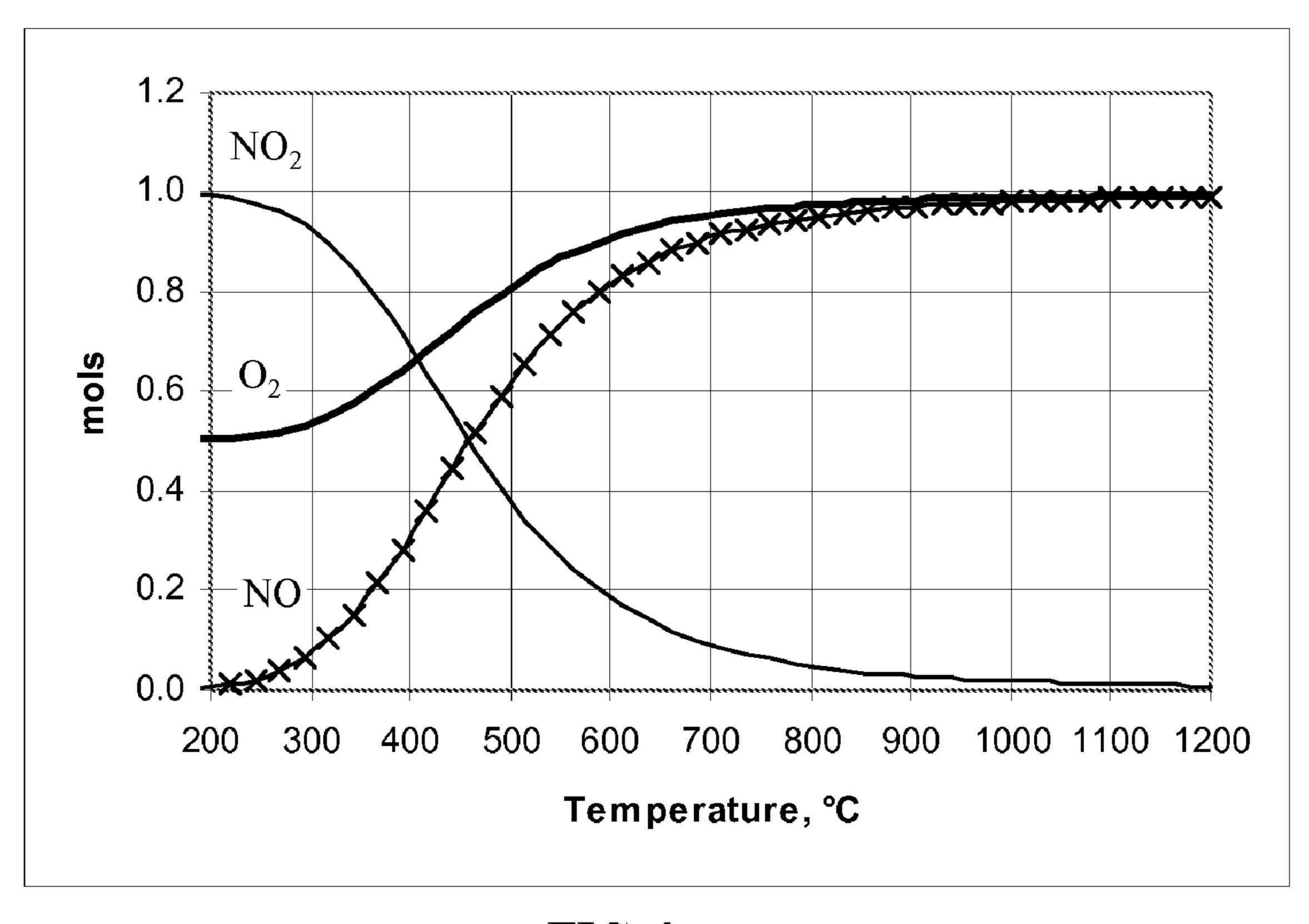


FIG. 2

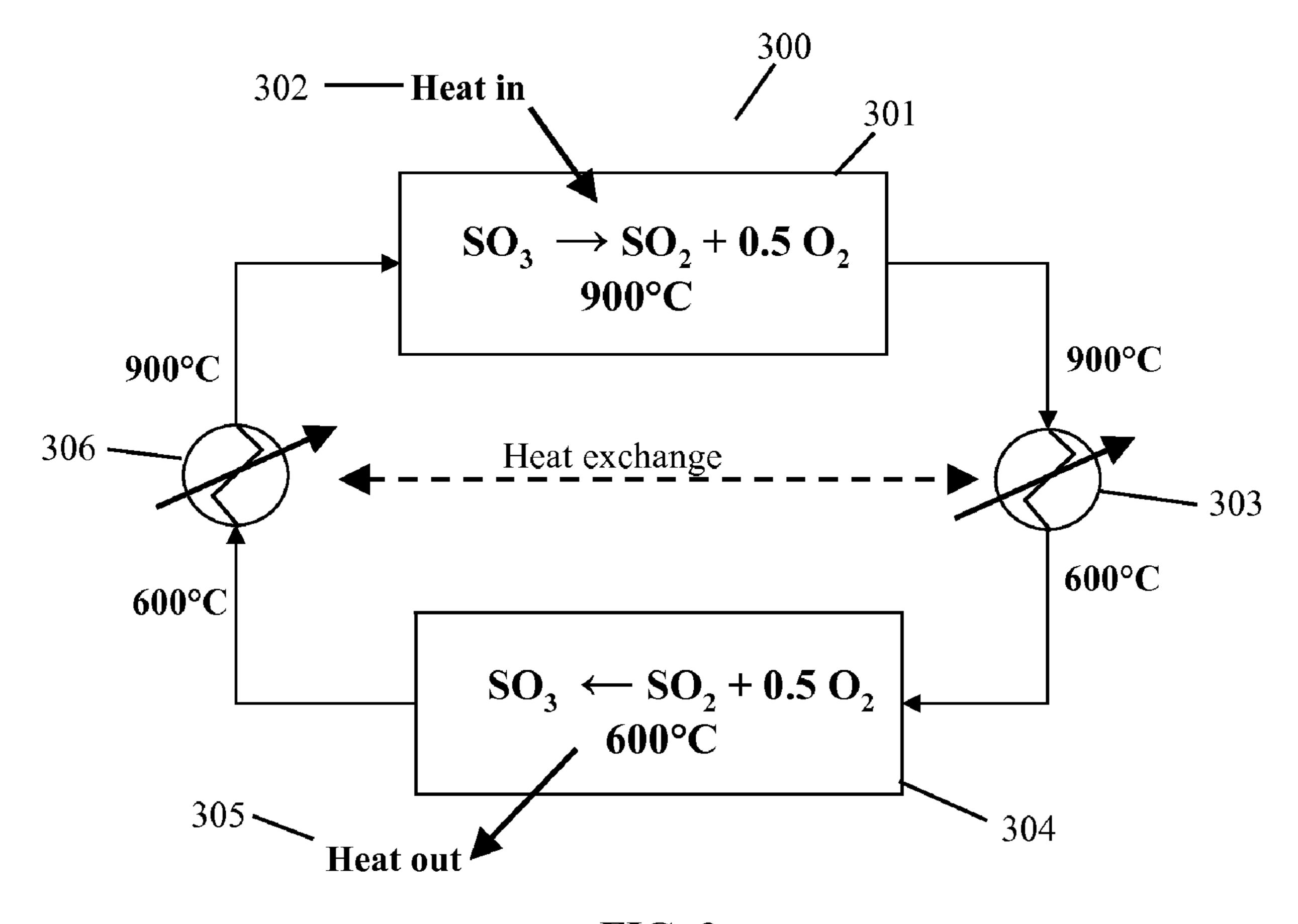
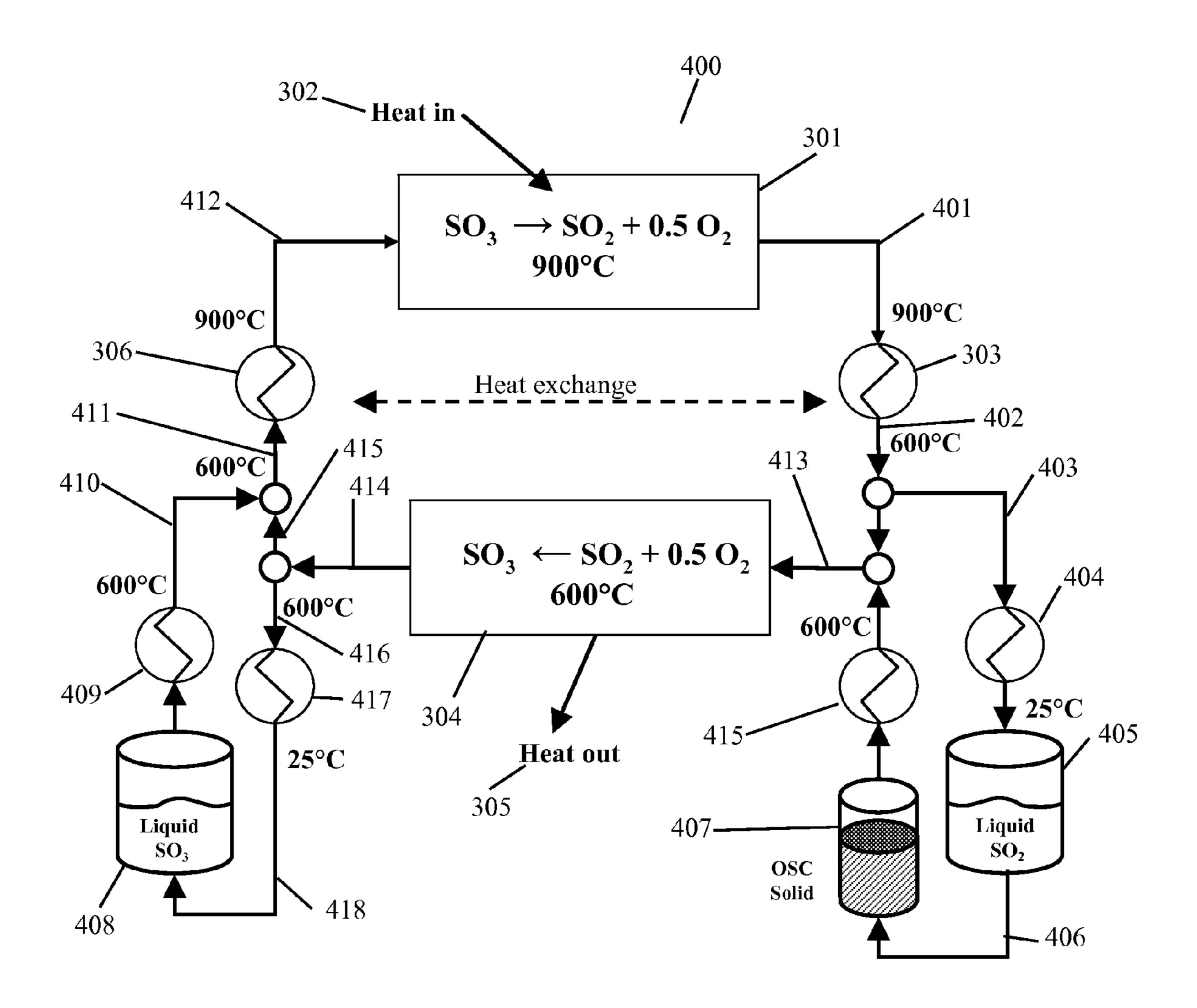
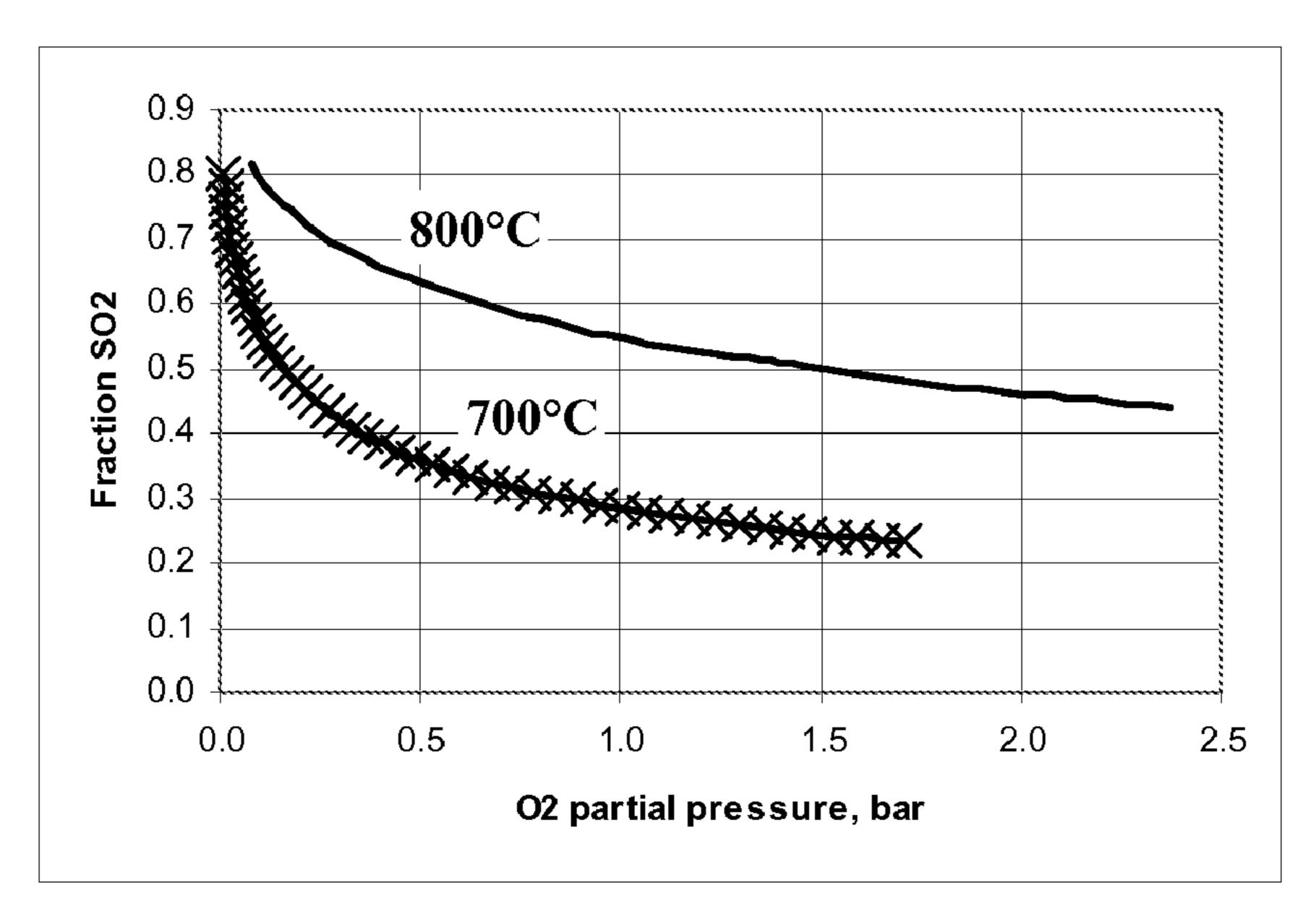


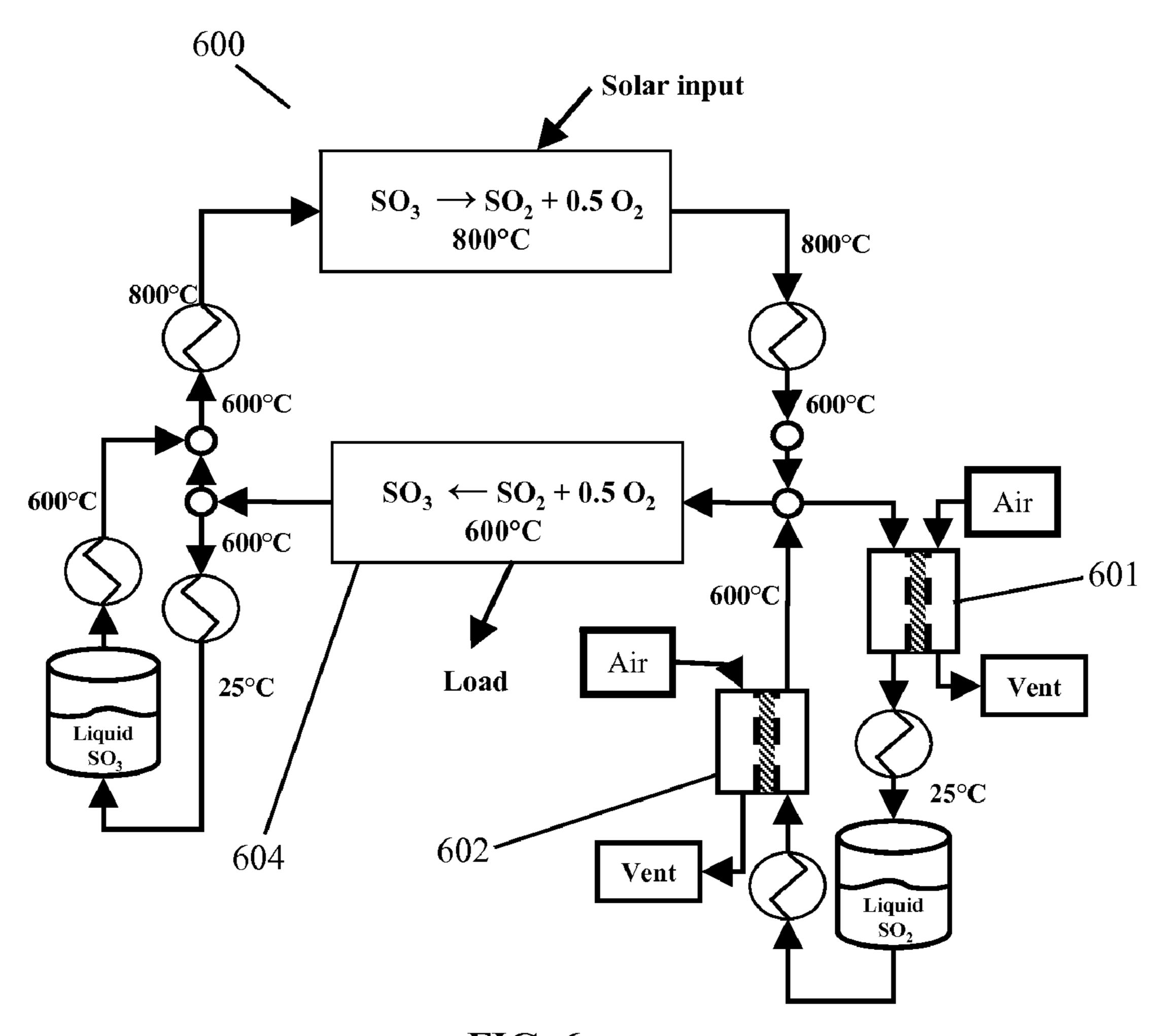
FIG. 3



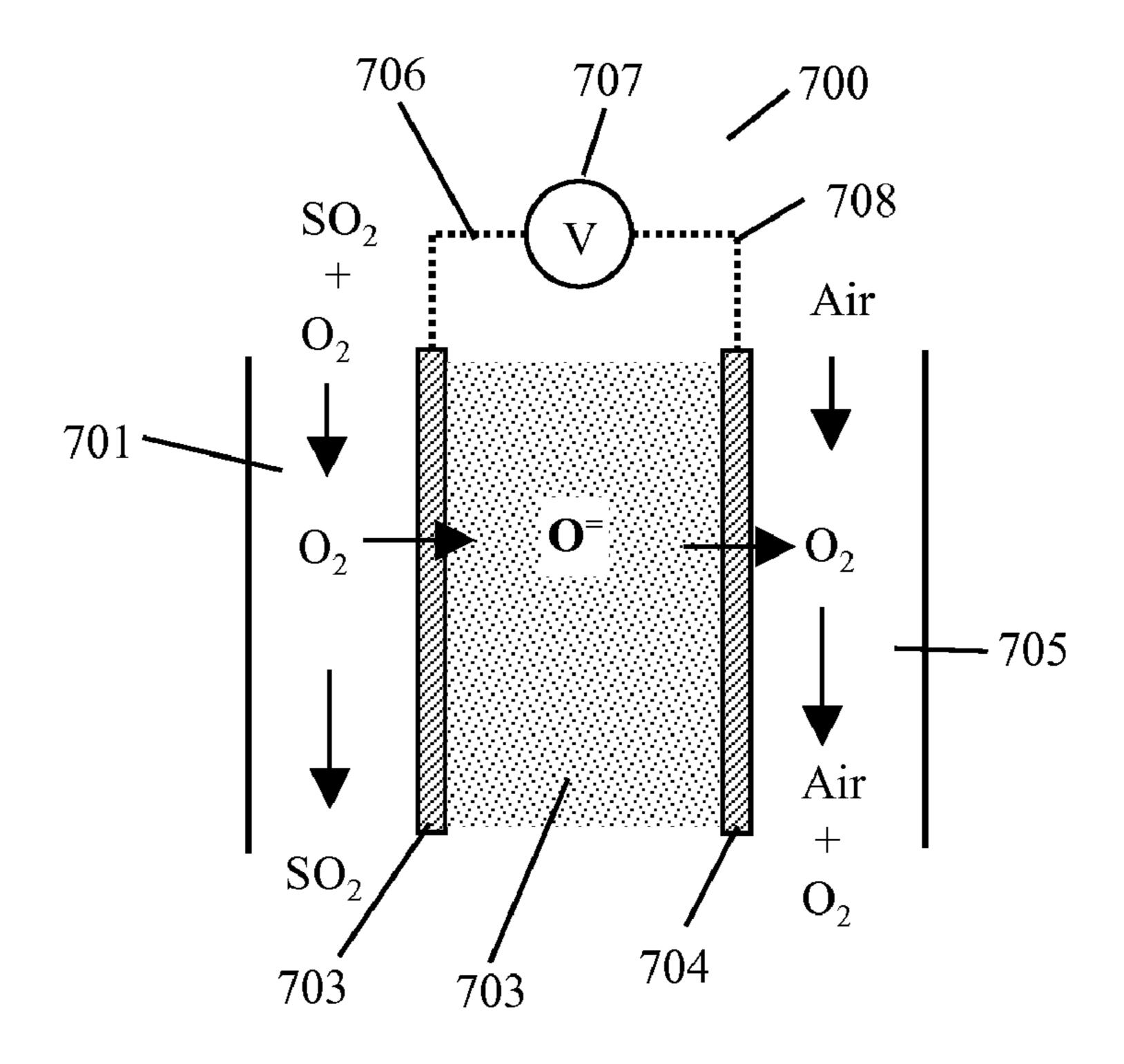
**FIG. 4** 



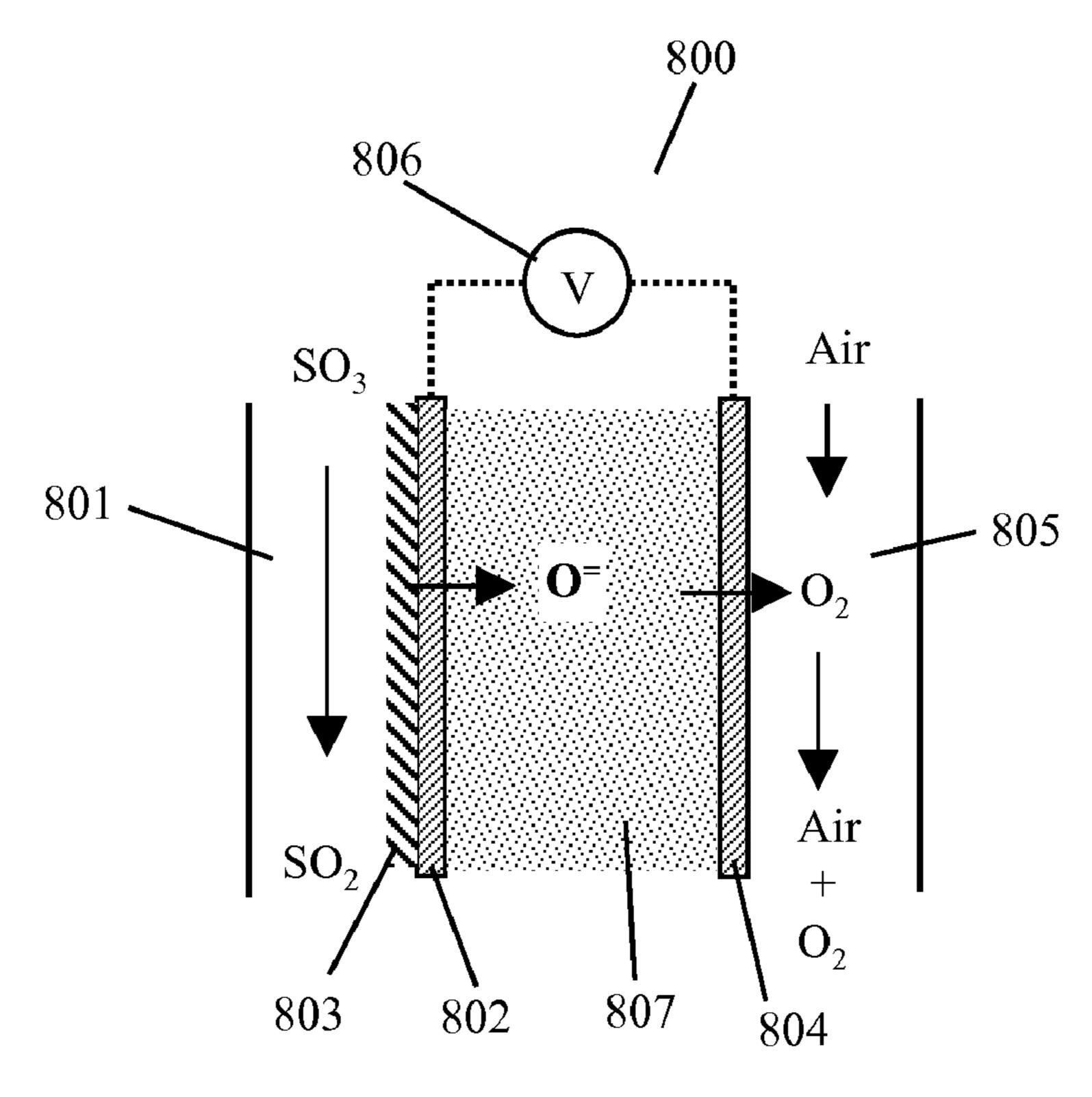
**FIG. 5** 



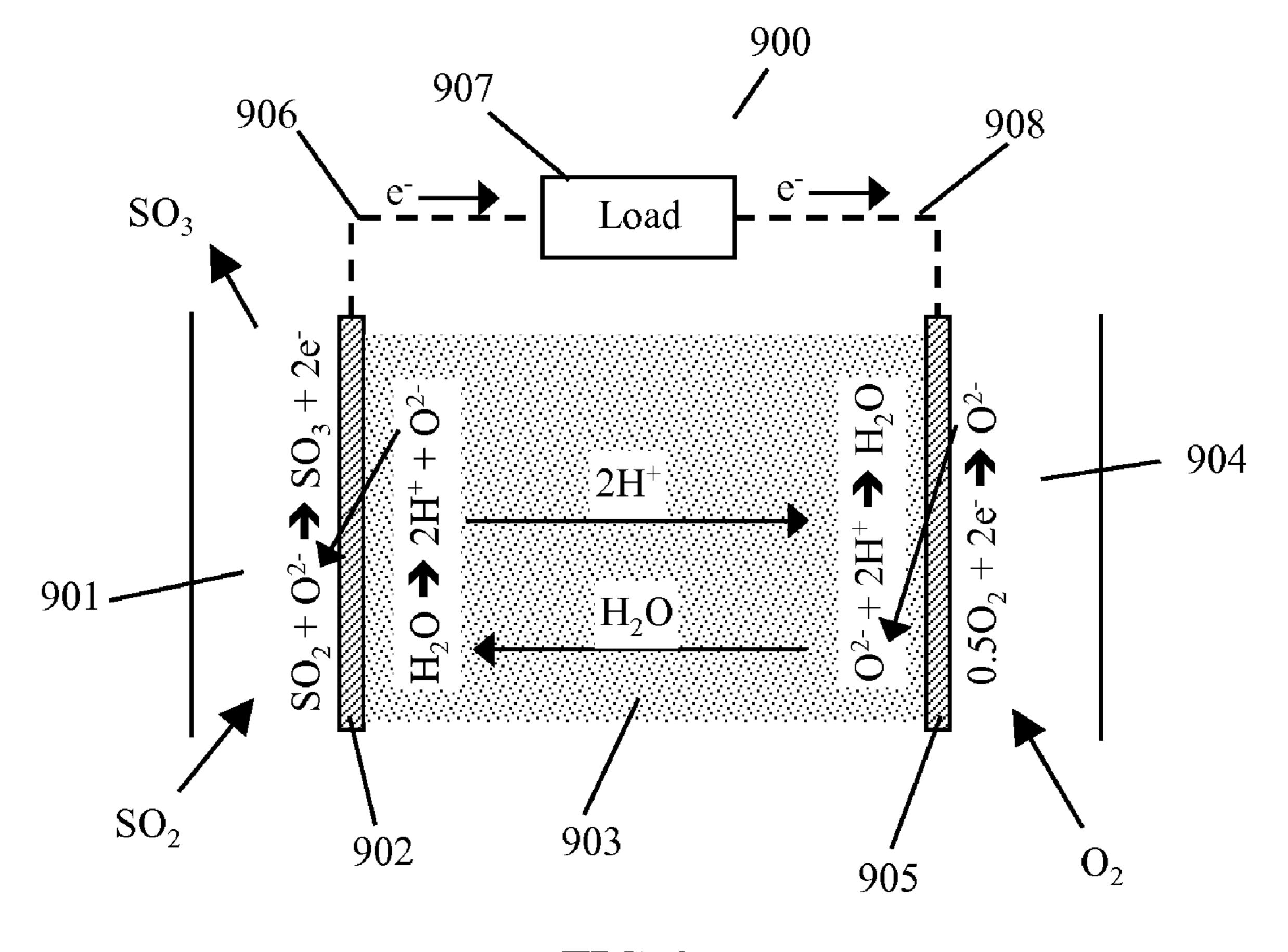
**FIG.** 6



**FIG.** 7



**FIG. 8** 



**FIG. 9** 

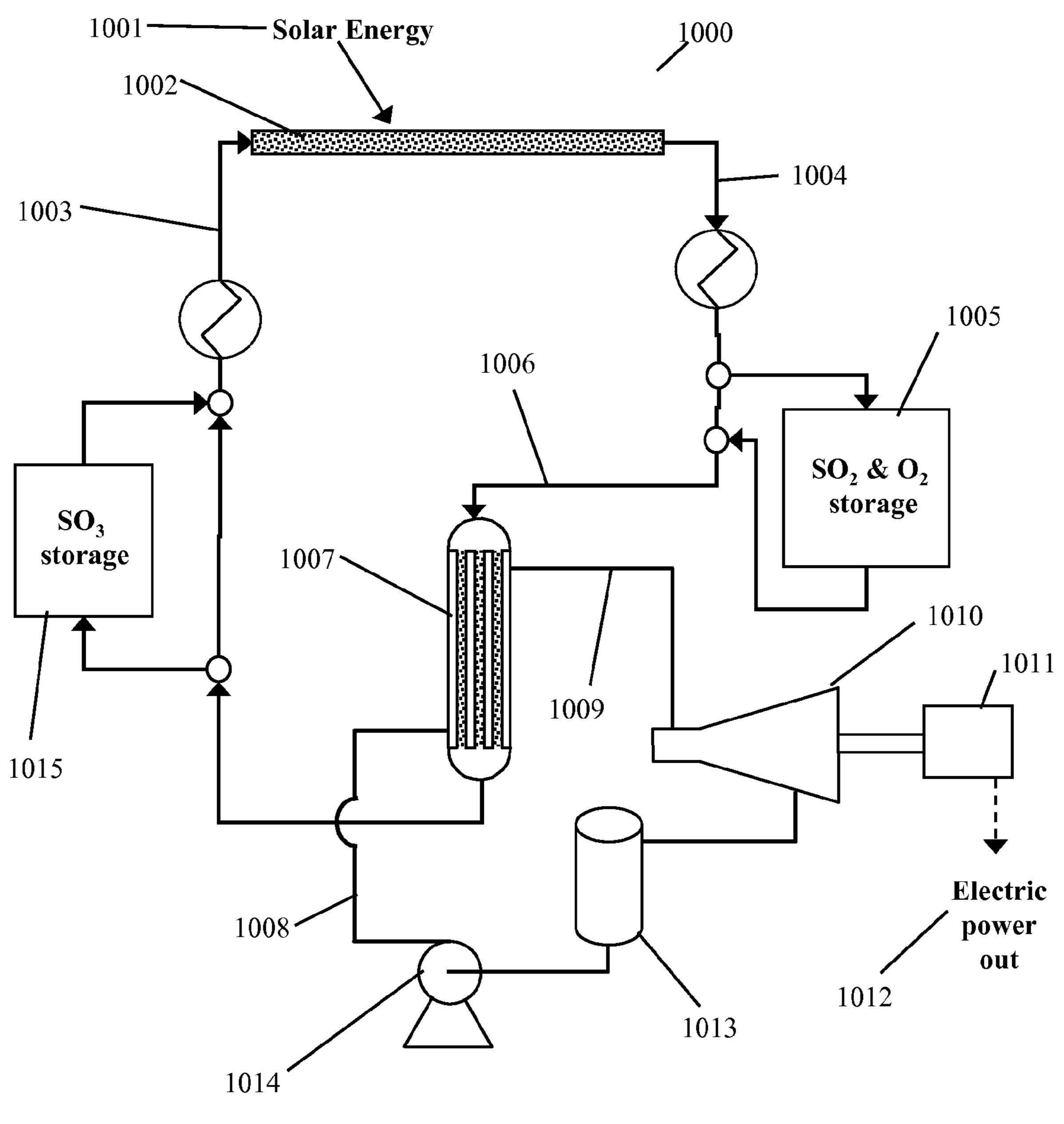
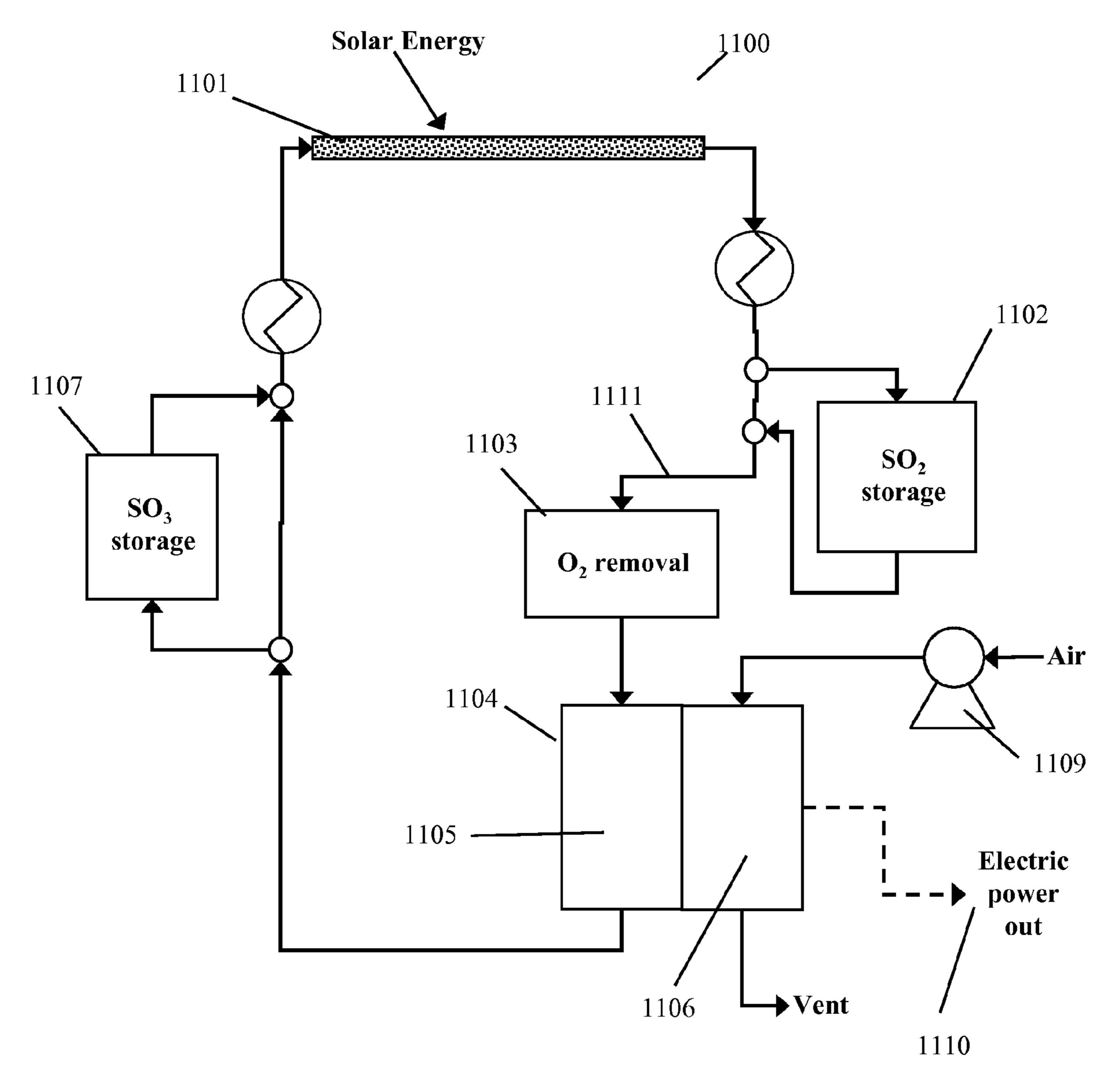


FIG. 10



**FIG. 11** 

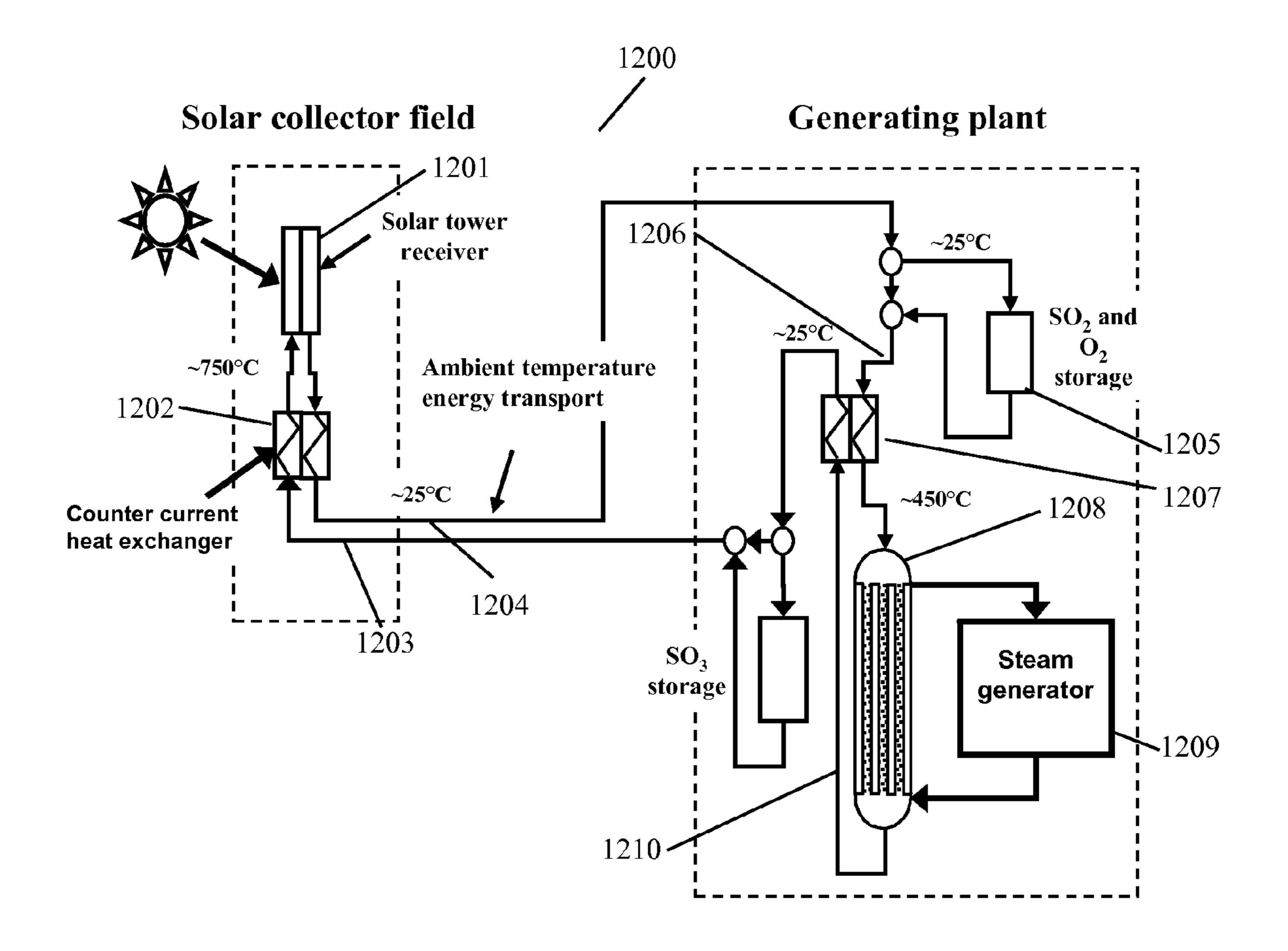


FIG. 12

#### THERMOCHEMICAL ENERGY STORAGE SYSTEM

#### FIELD OF THE INVENTION

[0001] The present invention relates to a concept and system design for thermochemical energy storage.

#### **BACKGROUND**

[0002] The headings used herein are for illustrative purposes and do not limit the interpretation of the following specification.

[0003] Thermal solar systems generate thermal energy when the sun is shinning but when the sun sets, solar thermal energy is not available and such system must either rely on conventional stored carbon based fuels or stored energy generated during the period when solar energy is available. Energy can be stored in a variety of ways such as electrical energy stored in batteries or capacitors, pumped storage where water is pumped to an elevated storage area for later use to generate electrical energy through a water powered turbine generator, pressurized storage of compressed gas etc. Another such energy storage technique is to use the solar energy to convert a chemical substance from a low energy state to a high energy state, storing this chemical compound and later use this chemical compound to generate energy through a chemical reaction that returns the chemical to it initial state and release energy, typically heat. One such process is the ammonia synthesis reaction and ammonia decomposition reaction shown in equations 1 and 2 as described in published articles by Luzzi, Lovegrove and coauthors (Solar Energy 66(2) pp. 91-101 (1999)).

$$H_2+3N_2 \rightarrow 2 NH_{3l+heat}$$
 (1)

$$3 \text{ NH}_3 + \text{heat} \rightarrow 3 \text{ H}_2 + \text{N}_2$$
 (2)

Both of these reactions are carried out over a catalyst. This reaction can transport energy from the solar collector where energy input dissociates HN<sub>3</sub> to H<sub>2</sub> and N<sub>2</sub>, The H<sub>2</sub> an N<sub>2</sub> are then transported to the location where the heat is needed where the ammonia synthesis reaction releases heat. To store energy for use during periods when solar energy is not available, for example at night or during cloudy periods, this system has to produce and store excess H<sub>2</sub> and N<sub>2</sub>. This is not an ideal situation since at ambient temperatures, H<sub>2</sub> and N<sub>2</sub> can only be stored as compressed gases since ambient temperature is above the critical temperature of these species. Storage as liquids would require additional energy input to liquefy these gases and storage at very low temperatures would have additional energy costs or require additional process equipment to deal with continual liquid evaporation. Also, storage of gas at high pressure results in high capital costs for the storage facilities and expenditure of considerable energy for compression as the gas is stored.

[0004] An alternative type of energy storage is described by Azpiazu and coauthors (Applied Thermal Engineering 23 (2003) p. 733-741) which uses the hydration of calcium oxide as the energy cycle. CaO is hydrated by reaction with water to release heat and then dehydrated using solar energy to regenerate the CaO and release the water.

 $CaO+H_2O\rightarrow Ca(OH)_2$ +heat

$$Ca(OH)_2$$
+heat $\rightarrow$ CaO+H<sub>2</sub>O (3)

 $SO_3$ +heat $\rightarrow SO_2$ +0.5  $O_2$  (5)

(4) 
$$SO_2+0.5 O_2 \rightarrow SO_3+heat$$
 (6)

In operation as a solar thermochemical storage system, the calcuim hydroxide is contained in a reactor as a packed bed or similar solid mass. Heat from solar energy collector is used to heat a gas or liquid heat transfer medium that transfers the heat energy to a heat exchange system installed in the calcium hydroxide bed to dehydrate the calcium hydroxide to calcium oxide and free water vapor at approximately 500° C. When heat energy is required from the stored chemical energy, the calcium oxide bed temperature is lowered to 25° C. and water added back to form calcium hydroxide and release the sorted energy. The energy is extracted by gas or fluid flow through a heat exchange system installed in the storage bed. A wide range of such adsorption-desorption reactions can be envisioned. U.S. Pat. No. 4,365,475 describes a number of such reaction couples for NH<sub>3</sub> adsorption-desorption. The reaction couple can be selected to collect energy and release energy over a variety of temperature ranges. The CaO—Ca(OH)<sub>2</sub> couple described above releases heat near ambient temperature so it would be a good cycle for space heating and adsorption chillers. Cycles described in U.S. Pat. No. 4,65,475 involving the formation of NH<sub>3</sub> complexes, could release energy at higher temperatures, for example 200 to 250° C., allowing the released heat to be used to generate steam for electrical power generation using a steam turbine. One major disadvantage of these adsorption-desorption systems is that the energy storage and release requires that a massive solid bed be heated and cooled to the required cycle temperatures thus wasting significant energy. Also, the large change in volume of the solid upon hydration-dehydration or ammonia complexation-decomposition make it difficult to prepare durable solid materials that can go through many cycles. Azpiazu and coauthors describe the calcium oxide-hydroxide couple as limited to 20 cycles before performance degradation was excessive.

[0005] Another system that has been described in U.S. Pat. Nos. 3,972,183 and 3,997,001 is the conversion of SO<sub>3</sub> into SO<sub>2</sub> and O<sub>2</sub> and the subsequent oxidation of SO<sub>2</sub> with O<sub>2</sub> to SO<sub>3</sub>. The process described uses a catalyst in the solar energy collector to decompose SO<sub>3</sub> to SO<sub>2</sub> and O<sub>2</sub> at about high temperature with the adsorption of heat, transport of the SO<sub>2</sub> and O<sub>2</sub> gas to the process or an energy storage unit where the SO<sub>2</sub> and O<sub>2</sub> is coverted to SO<sub>3</sub> and heat and the heat stored. These references describe the storage of energy as sensible heat in a thermal mass such as a bed of hot rocks or heat of phase change in a molten salt. Storage of energy as a hot thermal mass or as a hot molten salt requires insulation to retain the heat and also results in a slow loss of the stored energy through loss of heat.

#### **SUMMARY**

[0006] In embodiments of the invention, thermochemical energy storage cycles are provided that use the a reaction couple of a gaseous species that is catalytically decomposed to a less oxidized species and free oxygen with the adsorption of heat to store thermochemical energy followed by the catalytic oxidation of this less oxidized species to release energy. One embodiment of such the cycle is shown in equations 5 and 6.

Another embodiment of the cycle employs NO and NO<sub>2</sub> as shown in equation 7 and 8.

$$NO_2$$
+heat $\rightarrow NO+0.5 O_2$  (7)

NO+0.5 
$$O_2 \rightarrow NO_2$$
+heat (8)

[0007] For the SOx couple of equations 5-6, SO<sub>2</sub> is the stable species above 700 to 800° C. and SO<sub>3</sub> is the stable species below about 600° C. Thus, reaction 5 would be operated at 700 to 1000° C. with energy input from a solar or other energy source supplying heat and reaction 6 at 600° C. or lower with energy output to the target process. This would allow the production of high quality steam for power generation or process heat. The NOx cycle is similar with the NO<sub>2</sub> the preferred chemical species above 600° C. and NO the preferred chemical form below 300 to 400° C. SO<sub>2</sub> and NO plus O<sub>2</sub> provide thermochemical energy transport methods. The inventive cycles use the SO<sub>2</sub> to SO<sub>3</sub> interconversion combined with liquid storage of the SO<sub>2</sub> and SO<sub>3</sub> to provide a thermochemical energy storage system with a high energy density and low capital cost. Since the SO<sub>2</sub> and SO<sub>3</sub> can be stored at ambient temperature as a liquid, this eliminates the need for thermal energy storage at high temperature, eliminates the need for insulation to retain this high temperature stored heat and eliminates any loss of energy through slow loss of heat with time. In addition, inventive methods for the storage of the  $O_2$  or removal and supply of the  $O_2$  are described. In addition, an inventive electrochemical generator for the electrochemical oxidation of SO<sub>2</sub> into SO<sub>3</sub> and electricity is described.

[0008] A number of other aspects of these cycles will be described and systems described for energy transfer from the energy source (solar) to the process and for high density energy storage and release.

#### BRIEF DESCRIPTION OF THE DRAWINGS

[0009] FIG. 1 shows the equilibrium distribution of  $SO_3$ ,  $SO_2$  and  $O_2$  over a range of temperatures for 1 bar pressure starting with 1 mole  $SO_3$  and 0.5 mole  $O_2$ .

[0010] FIG. 2 shows the equilibrium distribution of  $NO_2$ , NO and  $O_2$  over a range of temperatures for 1 bar pressure starting with 1 mole  $NO_2$  and 0.5 mole  $O_2$ .

[0011] FIG. 3 is a schematic diagram of thermochemical energy cycle using  $SO_2$  and  $SO_3$  where the decomposition of  $SO_3$  to  $SO_2$  and  $O_2$  adsorbs thermal energy and the oxidation of  $SO_2$  to  $SO_3$  releases the thermal energy at the point of use.

[0012] FIG. 4 is a schematic diagram of a thermochemical energy cycle including energy storage using liquid  $SO_2$  and  $SO_3$  in one embodiment of the invention.

[0013] FIG. 5 is a graph showing the effect of oxygen partial pressure on the equilibrium fraction of SO<sub>2</sub> at 10 bar pressure for temperatures of 700 and 800° C. in one embodiment of the invention.

[0014] FIG. 6 is a schematic diagram of a thermochemical energy cycle including energy storage using an oxygen pump to remove and add  $O_2$  to the process stream in one embodiment of the invention.

[0015] FIG. 7 is a schematic diagram of an electrochemical O<sub>2</sub> pump in one embodiment of the invention.

[0016] FIG. 8 is a schematic diagram of an electrochemical  $O_2$  pump combined with a catalytic process to decompose  $SO_3$  into  $SO_2$  and  $O_2$  in one embodiment of the invention.

[0017] FIG. 9 is a schematic diagram of an electrochemical process for converting  $SO_2$  and  $O_2$  into  $SO_3$  and electric power in one embodiment of the invention.

[0018] FIG. 10 is a schematic diagram of a thermochemical energy cycle including energy storage and electrical power generation using thermal energy to generate steam and the steam to drive a turbine generator in one embodiment of the invention.

[0019] FIG. 11 is a schematic diagram of a thermochemical energy cycle including energy storage and an electrochemical generator that directly converts SO<sub>2</sub> and O<sub>2</sub> into electricity in one embodiment of the invention.

[0020] Use of the same reference numbers in different figures indicates similar or identical elements.

#### DETAILED DESCRIPTION

Basic System Design

[0021] FIG. 1 illustrates the thermodynamic species balance for the oxidation of SO<sub>2</sub> to SO<sub>3</sub> and FIG. 2 illustrates the thermodynamic balance for the NO to NO<sub>2</sub> couple, both at 1 bar pressure and with a starting composition of 1 mole of NO and SO<sub>2</sub> and 1 mole of O<sub>2</sub>. FIG. 1 shows that for the oxidation of SO<sub>2</sub> to SO<sub>3</sub>, equation 10, the equilibrium lies far to the left with SO<sub>2</sub> the preferred species at temperatures below about 750° C. and SO<sub>3</sub> the preferred species above 750° C. The oxidation of SO<sub>2</sub> to SO<sub>3</sub> is highly exothermic and the decomposition of SO<sub>3</sub> into SO<sub>2</sub> and O<sub>2</sub> is highly endothermic. By employing a catalyst that can facilitate the oxidation of SO<sub>2</sub> to SO<sub>3</sub> (equation 10) or the decomposition of SO<sub>3</sub> into O<sub>2</sub> and SO<sub>2</sub> (equation 9), this interconversion can form a thermochemical cycle.

$$SO_3 \rightarrow SO_2 + 0.5 O_2$$
 (9)

$$SO_2+0.5 O_2 \rightarrow SO_3$$
 (10)

[0022] FIG. 3 shows a system 300 using thermochemical energy cycle of  $SO_2$  and  $SO_3$ .  $SO_3$  is decomposed in a reactor 301 to  $SO_2$  and  $O_2$  at a temperature of about 900° C. with heat input 302 since the process is endothermic. The  $SO_2$  can then be circulated through a heat exchanger 303 to reduce the stream temperature to about  $600^\circ$  C. and then to a second reactor 304 which operates at a lower temperature where  $SO_3$  is thermodynamically preferred species and where the  $SO_2$  is oxidized to  $SO_3$  with the release of heat 305. To complete the cycle, the process stream from reactor 304 is circulated through heat exchanger 306 to raise the stream temperature to  $900^\circ$  C. and back to the  $SO_3$  decomposition reactor 301. This thermochemical cycle moves energy from one site where heat is adsorbed to a second site where energy is released.

[0023] This energy transfer cycle can also be an energy storage cycle by storing the high energy species, SO<sub>2</sub>. FIG. 4 shows a system 400 similar to system 300 of FIG. 3 but offers energy storage in one embodiment of the invention. System 400 similarly uses reactor 301 to decompose SO<sub>3</sub> to SO<sub>2</sub> and O<sub>2</sub> to produce process stream 401 which passes through heat exchanger 303 to reduce the stream temperature to 600° C. at point 402 prior to entering reactor 304. However, a portion of stream 402 can be split off to process stream 403 which then passes through heat exchanger 404 to drop the stream temperature to about 25° C. where a substantial portion of the SO<sub>2</sub> will change phase to a liquid and be collected in tank 405. The actual temperature of this stream and the temperature of the liquid SO<sub>3</sub> will depend on the system pressure and the overall process design. The temperature could range from

-50 to 250° C. In addition, the O<sub>2</sub> in process stream 403 will pass through tank 405, through line or process passage 406 to a vessel 407 containing an oxygen storage material which will adsorb or otherwise trap a substantial portion of the O<sub>2</sub>. Such an oxygen storage material is sometimes referred to as an oxygen storage compound (OSC). In addition, 407 can refer interchangeable to the oxygen storage material and the vessel containing the oxygen storage material. It should be noted that line and process passage are used interchangeably. In this manner, energy input 302 can be stored by producing a large amount of SO<sub>2</sub> and O<sub>2</sub> that is stored in vessels 405 and 407. Such an energy storage system must also have a source of SO<sub>3</sub> to convert to SO<sub>2</sub>. This is also shown in FIG. 4 where liquid SO<sub>3</sub> is stored in vessel **408**. During this energy storage operation, the liquid SO<sub>3</sub> in vessel 408 is vaporized and heated to 600° C. in heat exchanger 409 and then passes through line 410 and line 411 to heat exchanger 306 to be heated to 800° C. and then through line 412 to decomposition reactor 301 to complete the cycle. The overall process for energy storage then consists of vaporizing liquid SO<sub>3</sub> from vessel 408, heating this stream to 800° C., passing this process stream through reactor 301 to decompose the SO<sub>3</sub> to SO<sub>2</sub> and O<sub>2</sub> using a heat input source, then cooling this SO<sub>2</sub>+O<sub>2</sub> stream to 25° C. where the  $SO_2$  is condensed to be stored as a liquid and the  $O_2$ is stored in an oxygen storage material. This energy storage process can have a high energy density since the high energy phase, SO<sub>2</sub> is stored as a liquid and the O<sub>2</sub> can be stored as an absorbed phase in an oxygen storage material at reasonable pressures without the need for high pressure gas storage.

[0024] This stored energy can then be utilized by vaporizing  $SO_2$  in vessel 405, desorbing  $O_2$  from the oxygen storage material in vessel 407, passing this stream through heat exchanger 415 and on to reactor 304 where heat is generated. The  $SO_3$  is then passes through line 416 to heat exchanger 417 where it condenses to a liquid, then through line 418 to vessel 408 where the  $SO_3$  is stored as a liquid.

[0025] It should be noted that in operation, this process can operate in a variety of modes. All of the heat input from solar or other heat sources can be converted to heat output by directing all of the  $SO_2$  and  $O_2$  to process stream 413, through reactor 304 and then on through process stream 414 and 415 back to the decomposition reactor 301 where the cycle is completed. Another alternative is to split a portion of the  $SO_2$  and  $O_2$  to each of the process streams 403 and 413 so that some of the heat input is used to provide heat output and some of the energy input is stored as liquid  $SO_2$ . Of course the third operating mode is to have no heat input 302 and just produce heat output by using the stored  $SO_2$  in vessel 405 flowing through reactor 304 to produce heat output.

[0026] System 400 in FIG. 4 would be useful as a solar energy storage system where solar energy can be collected and supplied to reactor 301, this energy can be stored as liquid SO<sub>2</sub> in vessel 405 or used in reactor 304 to produce heat for any needed purpose. It can provide heat output when there is limited or no solar energy or other energy input by consuming stored SO<sub>2</sub> from vessel 405 to produce heat and then store the product SO<sub>3</sub> in vessel 408 for later use. Thus, this system can be used as a means to transfer energy from a solar collector to the point of use, store solar energy during periods when solar radiation exceeds current thermal needs, and produce thermal energy at the point of use from stored chemical energy when the solar radiation does not meet the thermal requirements.

[0027] To store energy using the process design shown in FIG. 4, the operating pressure has to be sufficiently high to

allow SO<sub>2</sub> and SO<sub>3</sub> to liquefy at reasonable temperatures. It is desirable to store the energy at ambient temperatures, for example below about 50° C. so that it can be stored for long periods without needing energy to maintain a low temperature or to thermally insulate or heat the storage vessels to maintain a higher temperature. If the process in FIG. 4 is operated at about 10 bar total pressure, then both SO<sub>2</sub> and SO<sub>3</sub> will condense to form a liquid if cooled below about 50° C. Pressures from 1 bar to above 100 bar could be used. In one embodiment, the operating pressure is in the range of 3 to 20 bar. In another embodiment, the operating pressure is in the range of 5 to 15 bar.

[0028] Reactors 301 and 304 may be thermal reactors or catalytic reactors. To obtain a fast rate of conversion from the input species to the output species, a catalyst may be used. Catalyst for the decomposition of SO<sub>3</sub> to SO<sub>2</sub> and O<sub>2</sub> and for the oxidation of SO<sub>2</sub> and O<sub>2</sub> to SO<sub>3</sub> are well known in the art and are discussed elsewhere in this specification. The temperature of operation of reactor 301 will depend on the level of conversion desired and on other process variables such as total pressure, partial pressure of O<sub>2</sub> and the temperature of the storage vessels 405 and 408. At 10 bar pressure, approximately 80% conversion of SO<sub>3</sub> to SO<sub>2</sub> can be obtained at about 1000° C. If the oxygen partial pressure is reduced, then the temperature for 80% conversion will decrease. The operating temperature of reactor 301 is in the range of 600 to 1200° C. In one embodiment, the operating temperature is in the range of 700 to 1000° C. In another embodiment, the operating temperature is in the range of 800 to 1000° C. Similarly, the operating temperature of reactor 304 is in the range of 300 to 800° C. In one embodiment, the operating temperature is in the range of 500 to 700° C. In another embodiment, the operating temperature is in the range of 500 to 600° C.

#### Operation at Reduced Oxygen Partial Pressures

[0029] At 10 bar pressure with excess O<sub>2</sub> in the circulating streams, reactor 301 must operate at quite high temperatures to obtain a high level of conversion from SO<sub>3</sub> to SO<sub>2</sub>. A high operating temperature may not be desirable for thermal solar systems since this would reduce efficiency and increase cost. One strategy to reduce the temperature of reactor 301 is to decrease the oxygen concentration. FIG. 5 shows the equilibrium fraction of sulfur species versus O<sub>2</sub> partial pressure at several temperatures and at 10 bar total pressure in one embodiment of the invention. At 800° C., if the O<sub>2</sub> partial pressure is below 0.25 bar, then the SO<sub>2</sub> fraction will be >70%. If the O<sub>2</sub> partial pressure can be reduced below 0.1 bar, then the SO<sub>2</sub> fraction could be greater than 80%. Similarly, at 700° C., the SO<sub>2</sub> fraction can be increased substantially if the O<sub>2</sub> partial pressure is reduced. By operating the process at lower O<sub>2</sub> partial pressure, the equilibrium conversion of SO<sub>3</sub> to SO<sub>2</sub> would be much higher. This could be achieved by operating the process with lower average O<sub>2</sub> pressure. A lower O<sub>2</sub> partial pressure could be accomplished by the choice of the oxygen storage material in vessel 407 or the operating temperature of this oxygen storage material. The actual magnitude of the O<sub>2</sub> partial pressure would be given by a tradeoff between the optimal pressure for the decomposition reaction, equation 10, in reactor 301 and the oxidation of SO<sub>2</sub>, equation 9, in reactor **304**. A lower O<sub>2</sub> partial pressure will drive equation 10 and allow reactor 301 to operate at a reasonable temperature, say 700 to 800° C. A higher O<sub>2</sub> partial pressure will drive equation 10 to the right producing

more  $SO_3$  so the actual choice of  $O_2$  partial pressure is a trade off between the needs of the two processes shown in equation 9 and 10. The main driver for the selection of the  $O_2$  partial pressure will probably be equation 9, the decomposition of  $SO_3$  to  $SO_2$ . The partial pressure of  $O_2$  for system 300 or 400 would be in the range of 0.01 to 3 bar. In one embodiment, the partial pressure of  $O_2$  for system 300 or 400 is in the range of 0.01 to 1.5 bar. In another embodiment, the partial pressure of  $O_2$  is in the range of 0.01 to 0.75 bar.

[0030] The O<sub>2</sub> partial pressure could be controlled by controlling the temperature of the oxygen storage material 407. This vessel containing oxygen storage material could be placed upstream of heat exchanger 404 to operate at 600° C. or heat exchanger 404 could be divided into several sections, the first section reducing the temperature of stream 403 to some intermediate temperature for oxygen storage material 407 placed in this location downstream of this first heat exchanger section. A second heat exchanger section would then reduce the temperature of this process stream to the temperature required to liquefy SO<sub>2</sub> for storage in vessel 405. Oxygen storage material containing vessel 407 could also be placed in other locations in the process loop. For example it could be placed just downstream of reactor 301 in line 401 for operation at very high temperature. Alternatively, the oxygen storage material 407 could be divided into several vessels and placed in different locations to control the O<sub>2</sub> partial pressure to different levels in different parts of the process stream.

#### Oxygen Storage Materials

[0031] The oxygen storage materials used in system 400 will depend on the specific system design and operating conditions. Metal oxides and peroxides offer one type of oxygen storage material. BaO and BaO2 is one example. At about 825° C., BaO<sub>2</sub> would have an O<sub>2</sub> partial pressure of about 1 bar. Lower temperatures would provide lower equilibrium O<sub>2</sub> partial pressures. Cerium oxides, manganese oxides, cerium and palladium oxides and mixed oxides can also provide embodiments of oxygen storage materials. One embodiment of a mixed oxide O<sub>2</sub> storage material is the mixed oxide REBaCo<sub>4</sub>O<sub>7</sub> as described by Motohashi et. al. (Materials Science and Engineering B 148 (2008) 196-198) where this material can store up to 3% of the oxygen storage material's weight as oxygen in the temperature range of 200 to 400° C. RE is one of the rare earth elements, in particular Y, Dy, Yb, and Lu (elements of the periodic table).

#### Electrochemical Control of O<sub>2</sub> Partial Pressure

[0032] The storage of oxygen is a critical aspect of the inventive process since storage as a gas would not be cost effective since it would require very large vessels or compression to high pressures which would consume energy and reduce efficiency. A significant observation is that the O<sub>2</sub> does not have to be stored within the process but can be removed from the process as excess SO<sub>2</sub> is generated and stored in vessel 405 and then added back to the process when the SO<sub>2</sub> is oxidized. FIG. 6 shows a system 600 similar to system 400 of FIG. 4 but replaces the oxygen storage material 407 with single or multiple electrochemical cells or electrochemical oxygen pumps in one embodiment of the invention. Electrochemical oxygen pump and electrochemical oxygen cell are used interchangeably in this disclosure. In system 600, electrochemical pumps 601 and 602 replace the oxygen storage material vessel 407. These electrochemical pumps function

as O<sub>2</sub> pumps. Cell **601** pumps O<sub>2</sub> out of the process when excess SO<sub>3</sub> is decomposed to SO<sub>2</sub> and O<sub>2</sub> moving the O<sub>2</sub> to an air purge stream and into the atmosphere. Cell 602 pumps O<sub>2</sub> into the process when the SO<sub>2</sub> is vaporized and is to be oxidized to SO<sub>3</sub> to generate heat. In this case, the electrochemical cell 602 pumps O<sub>2</sub> from the air purge stream into the process. FIG. 7 is a schematic diagram of such an electrochemical cell 700 in one embodiment of the invention. In FIG. 7, electrochemical pump 700 operates in the mode required for oxygen removal pump 601 of FIG. 6. SO<sub>2</sub> and O<sub>2</sub> pass through channel 701. Electrode 702 dissociates the O<sub>2</sub> into oxygen ions,  $O^{2-}$  and electrons, the oxygen ions are then transported through electrolyte 703 to the opposite electrode 704. At electrode 704 the oxygen ions are recombined with electrons to produce O<sub>2</sub> which is then extruded into the gas phase, a flowing purge stream of air in channel 705. The electrons travel from electrode 702 through conductors 706 and 708 to electrode 704. The pumping of O<sub>2</sub> from channel 701 to channel 705 is driven by the applied voltage, V, 707. The applied voltage controls the  $O_2$  pumping rate and the oxygen partial pressure that can be achieved in channel 701. By increasing the voltage, the  $O_2$  partial pressure can be driven to a very low value if desired and the pumping rate can be very high. For a given voltage, the pumping rate and the ultimate O<sub>2</sub> partial pressure can be also be influenced by the O<sub>2</sub> partial pressure in the air purge channel **705**. If the purge air is at 1 bar pressure, then the O<sub>2</sub> partial pressure would be approximately 0.21 bar. If a vacuum is applied to channel 705, then the O<sub>2</sub> partial pressure can be reduced to any desired value. For example, by operating the air purge channel 705 at 0.25 bar, the effective  $O_2$  partial pressure would be 0.05 bar in the process flow stream.

[0033] The electrochemical pump 602 in FIG. 6 is essentially similar to pump 700 shown in FIG. 7 except that the  $O_2$  flow is in the direction to move  $O_2$  from the air purge stream to the process flow stream, pumping  $O_2$  into the process flow stream. The current flow would be in the opposite direction and the effect of the  $O_2$  partial pressure on the air purge side would be opposite. A higher air pressure in the air purge channel 705 would help drive the  $O_2$  into the process flow stream and require a lower pumping voltage.

[0034] The use of an electrochemical pump to move O<sub>2</sub> from air into the process and from the process out to air would eliminate the need to store  $O_2$  in the process equipment and reduce the overall equipment size for energy storage. The only stored chemical is the liquid SO<sub>2</sub>. Ambient air becomes the source and sink for the required O<sub>2</sub> reactant. The design of the electrochemical oxygen pump can take many forms. A possible form is a zirconium oxide solid electrolyte cell such as that described in U.S. Pat. No. 5,378,345 and U.S. Pat. No. 4,877,506 which are incorporated into this application in their entirety. These cells use a stabilized zirconium oxide electrolyte that can transport oxygen ions and porous electrodes that act to dissociate  $O_2$  into oxygen ions and recombine the oxygen ions to form O<sub>2</sub> and also act as the electron conductor. A typical electrode material is a porous platinum layer. Other platinum group metals or mixtures of platinum group metals could be used.

[0035] An alternative embodiment using an electrochemical oxygen pump to remove oxygen from and add oxygen to the process stream would replace the ambient air purge at 601 and 602 of FIG. 6 with two vessels containing an oxygen storage material that would absorb and release the oxygen as the electrochemical pump removes it from or adds it to the

process stream. This could provide an advantage if any SO<sub>3</sub> or SO<sub>2</sub> were to leak across the electrochemical cell or pump since it would be trapped within the vessels holding the oxygen storage material and would not be released to the ambient air. The oxygen storage material could be contained in two vessels, one for each electrochemical pump 601 and 602 or it could be a single vessel connected to both electrochemical pump 601 and 602.

[0036] It should also be noted that while an electrochemical oxygen pump or cell is described herein, an alternative embodiment is to use an oxygen permeable membrane to selectively remove  $O_2$  from the process stream to an external purge stream. Oxygen could also be added back using a similar oxygen permeable membrane. The driving force for movement of  $O_2$  into the process stream or out of the process stream would be the  $O_2$  partial pressure on the opposite side of the membrane. A high  $O_2$  pressure or high air pressure would drive  $O_2$  into the process and a low  $O_2$  partial pressure or air pressure would drive  $O_2$  out of the process.

#### Electrochemical SO<sub>3</sub> Decomposition Reactor

[0037] The decomposition of SO<sub>3</sub>, equation 9, is limited by thermodynamics at the selected operating temperature. This is shown in FIG. 1 for a pressure of 1 bar as one embodiment of the invention. At the operating pressure of 5 to 15 bar, these curves would be shifted to the right and the equilibrium limited conversion to SO<sub>2</sub> can be low as shown in FIG. 5. Reducing O<sub>2</sub> partial pressure allows a higher equilibrium conversion. One approach to reducing the O<sub>2</sub> partial pressure is to make the reactor 301 in FIG. 4 a combined O<sub>2</sub> cell to pump oxygen and SO<sub>3</sub> decomposition reactor. FIG. 8 shows such a reactor 800 that can be placed in the location occupied by reactor **301** in FIG. **4** in one embodiment of the invention. The SO<sub>3</sub> enters reactor through channel **801**. One wall of the reactor is formed by the electrode 802 that is coated with a catalyst layer 803. The SO<sub>3</sub> reacts on the catalyst 803 to decompose into SO<sub>2</sub> and O<sub>2</sub>. The O<sub>2</sub> diffuses to electrode **802**, is dissociated into oxygen ions at electrode 802, is pumped through electrolyte 807 to the opposite electrode 804 where it is recombined to form O<sub>2</sub> and passes into channel **805** where it is swept away in the flowing air purge stream. By controlling voltage 806, the O<sub>2</sub> partial pressure at the interface between the catalyst layer 803 and electrode 802 can be controlled to the desired value. The O<sub>2</sub> partial pressure in the catalyst layer could be lowered to a very low value and as shown in FIG. 5, the equilibrium SO<sub>2</sub> conversion can be very high. This could also allow the reactor **800** to operate at a lower temperature ranging from 650 to 800° C.

Direct Electrochemical Conversion of SO<sub>2</sub> to Electrical Power

[0038] The process shown in FIG. 6 produces energy by oxidation of the stored SO<sub>2</sub> oxidation with O<sub>2</sub> to produce heat in reactor 604. This heat can then be used to produce steam which would power a steam turbine to produce electricity. Direct conversion of the SO<sub>2</sub> into electrical energy would be more desirable and could be substantially more efficient. FIG. 9 shows a system 900 with direction conversion of SO<sub>2</sub> into electrical energy in one embodiment of the invention. This device can be referred to as an SO<sub>2</sub> fuel cell or an SO<sub>2</sub> electrochemical generator. For the anode of the SO<sub>2</sub> fuel cell, SO<sub>2</sub> enters channel 901 and reacts on electrode 902 with oxygen ion, O<sup>2-</sup>, to form SO<sub>3</sub> and 2 electrons, 2e<sup>-</sup>. The O<sup>2-</sup> is

supplied through the electrode 902 from electrolyte 903. For the cathode of this fuel cell, O<sub>2</sub> enters channel **904** and reacts at electrode 905 to receive 2 electrons per O atom and form  $O^{-2}$  at the electrode. The electrons flow from the anode 902 through electric line 906 to the load 907 and electric line 908 to the cathode 905. The electrolyte 903 must transport  $O^{2-}$ from the cathode 905 to the anode 902. In one embodiment of the invention, this is done by the reactions shown in FIG. 9. At the cathode,  $O^{2-}$  reacts with  $2H^{+}$  in the electrolyte to form H<sub>2</sub>O. The H<sup>+</sup> is transported from the anode and H<sub>2</sub>O is transported to the anode through the electrolyte 903. At the anode, H<sub>2</sub>O dissociates to 2H<sup>+</sup> and O<sup>2-</sup> thus supplying the H<sup>+</sup>for transport to the cathode and oxygen ion,  $O^{2-}$ , for the anode reaction to oxidize SO<sub>2</sub> to SO<sub>3</sub>. An appropriate electrolyte for the SO<sub>2</sub> fuel cell of FIG. 9 would be sulfuric acid, H<sub>2</sub>SO<sub>4</sub> or any electrolyte that transports H<sup>+</sup> and H<sub>2</sub>O. The sulfuric acid could range from a dilute sulfuric acid to a nearly 100% concentrated sulfuric acid. The electrolyte could also be formed of a membrane or other porous material that is impregnated or filled with sulfuric acid. Alternatively the electrolyte could be a membrane or porous medium with the ability to transport H<sub>2</sub>O and H<sup>+</sup>. One such embodiment would be Nafion, a sulfonated tetrafluoroethylene copolymer. The transport of H<sup>+</sup>is equivalent to the transport of hydronium ion, H<sub>3</sub>O<sup>+</sup>. The overall electrochemical process shown in FIG. 9, identical to reaction equation 10, has a standard Gibbs free energy of reaction of -70.9 kJ/mol at 25° C. Using the Nernst equation, this equates to a standard cell potential of +0.37 volts. The operating temperature of this cell in one embodiment of the invention can vary over a wide range from 0° C. to 300° C. A lower temperature will provide a higher driving force and a larger cell voltage. In one embodiment, the operating temperature is in the range of 25° C. to 200° C.

[0039] An alternative embodiment of the invention to the process shown in FIG. 9 where the electrolyte transports protons,  $H^+$ , and water,  $H_2O$ , the electrolyte could transport only protons and use water supplied with the  $SO_2$  flow in channel 901. In this case the reaction of water to form  $H^+$  and  $O^{2-}$ ,  $H_2O \rightarrow 2H^+ + O^{2-}$ , would occur on the electrode in channel 901 using water supplied in the stream flowing through channel 901.

#### Thermal Solar Energy Storage System

[0040] The process and system design described above can be used as system for the capture and storage of solar energy. FIG. 10 illustrates one such of a system 1000 in one embodiment of the invention. In system 1000, solar energy 1001 is captured in the collector 1002 as high temperature heat. SO<sub>3</sub> flows into collector 1002 through stream 1003. Collector 1002 is filled with a catalyst for the conversion of SO<sub>3</sub> to SO<sub>2</sub> and  $O_2$  and captures the thermal energy from solar radiation. The SO<sub>2</sub> and O<sub>2</sub> flow through process stream **1004** and then is split into section 1005 or to process stream 1006. In section 1005, the SO<sub>2</sub> and O<sub>2</sub> are stored for use during low solar energy periods. The SO<sub>2</sub> and O<sub>2</sub> that flow through process stream 1006 flow into reactor 1007 where the SO<sub>2</sub> and O<sub>2</sub> react to produce SO<sub>3</sub> and release thermal energy, heat. Reactor 1007 could be a heat exchange reactor such as a tube and shell reactor containing a catalyst for the conversion of SO<sub>2</sub> and O<sub>2</sub> into SO<sub>3</sub> and heat. Water or low temperature steam could flow into reactor through line 1008 to produce high temperature steam that exits reactor 1007 through line 1009 to steam turbine 1010 where the energy is extracted to drive electric generator 1011 to produce electric power output

1012. The steam or condensed water flows out of the steam turbine 1010 to storage vessel 1013 and to circulation pump 1014 and back to reactor 1007. The  $SO_2$  and  $O_2$  storage section 1005 and the  $SO_3$  storage section 1015 are not shown in detail as they can take any of the forms described in other parts of this specification and other forms that can be implemented by those familiar with the art.

[0041] The system shown in FIG. 10 is one example of a solar energy generation system using the aspects described in this specification. This system uses the solar energy to generate high temperature steam that is used to drive a steam turbine to produce electrical power. This system is only described for illustration purposes and the system can take many forms. In other embodiments of the invention, the solar energy collection components can be a central tower structure with surrounding solar reflectors that concentrate the solar energy on the central energy collector. An alternative embodiment is a parabolic trough collector that concentrates the solar energy on a linear tube reactor. A third embodiment would be a circular parabolic mirror that concentrates the solar energy on a central thermal collector. The SO<sub>2</sub> and O<sub>2</sub> storage system could be a system that liquefies the SO<sub>2</sub> and stores the SO<sub>2</sub> as a liquid at temperatures near ambient. The O<sub>2</sub> can be stored by absorption on an oxygen storage material or by adsorption on the surface of an adsorbent. The gaseous O<sub>2</sub> can be compressed at high pressure to store the  $O_2$  in a pressure vessel. An alternative embodiment is to use an oxygen transfer membrane to extract O<sub>2</sub> from the process stream and move it into the surrounding air. Such a membrane could take the form of an electrochemical cell or pump that transfers the  $O_2$  as oxide ions such as stabilized ZrO<sub>2</sub> membranes operating at high temperatures. The electrical power generation process can also take many forms. It can take the form of a thermal process wherein the oxidation of SO<sub>2</sub> to SO<sub>3</sub> produces heat that is used to produce steam that then drives a turbine and electric generator as shown in FIG. 10. Alternatively the heat from the decomposition reactor can be used to heat a heat transfer fluid which is then used to generate steam in a boiler or to heat other fluid to drive a turbine or other engine to produce electrical power. The engine can take the form of a turbine, reciprocating engine, sterling engine etc.

#### Direct Electrochemical Solar Energy System

[0042] FIG. 11 illustrates an alternative solar energy system in one embodiment of the invention. System 1100 is similar to system 1000 in FIG. 10 but with a modification in the method of electric power generation from the oxidation of SO<sub>2</sub> to SO<sub>3</sub>. The SO<sub>2</sub> generated in solar energy collection reactor 1101 will flow to SO<sub>2</sub> storage system 1102 or to the O<sub>2</sub> removal section 1103 and electrochemical generator 1104 which produces an electrical power output 1110. Electrochemical generator 1104 could be similar to that described in FIG. 9. The SO<sub>2</sub> stream flows to the anode side 1105 of the cell and an oxygen containing stream such as air flows to the cathode side of the cell 1106. The SO<sub>3</sub> oxidation product from the electrochemical generator 1104 is either recycled to the solar energy collector 1101 or stored in SO<sub>3</sub> storage section 1107 where the SO<sub>3</sub> is stored for use during low solar energy periods. The cathode side 1106 of the electrochemical generator 1104 can be supplied with oxygen by a purge air stream using blower 1109. Other oxidants could be used. The O<sub>2</sub> removal section 1103 could be similar to the oxygen storage material or the electrochemical pump described elsewhere in the specification and shown in FIG. 7. Alternatively, it could

be a membrane based system that uses oxygen partial pressure difference to extract  $O_2$  from stream 1111. The level of oxygen removal required will depend on the sensitivity of the anode 1105 to  $O_2$ . In general, the  $O_2$  level does not have to be extremely low since any oxygen present at the anode will probably just react directly with the  $SO_2$  to form  $SO_3$  thus bypassing the electrochemical conversion and lead to a lower electrical generation efficiency. It is not expected that low levels of  $O_2$  at the anode will inhibit the anode electrochemical reaction.

[0043] In one embodiment of this invention, catalysts for the oxidation of SO<sub>2</sub> to SO<sub>3</sub> would be platinum, palladium or other platinum group metals supported on a oxide or other high surface area support such as aluminum oxide, silicon oxide, titanium oxide, zirconium oxide or mixtures of these oxides with or without additional additives. Vanadium is also a preferred catalyst, again supported on oxides such as described for the platinum group metal catalysts. An optimal catalyst used commercially for this reaction is vanadium oxide supported on silica support containing potassium, sodium and aluminum oxide additives.

[0044] The decomposition of SO<sub>3</sub> to SO<sub>2</sub> and O<sub>2</sub> would be done on similar catalysts as for the oxidation as described above.

[0045] Anode materials for the electrochemical generator described in FIG. 9 would be similar to the catalysts used for the catalytic oxidation of SO<sub>2</sub> and described above. A preferred catalyst would be a porous platinum layer since this would act as an electron conducting layer as well as a catalyst for the oxidation of SO<sub>2</sub> using oxide ions transported by the electrolyte.

[0046] Cathode catalysts for the electrochemical generator of FIG. 9 would be similar to those described for the hydrogen oxygen fuel cell and are well described in the literature and well known.

#### Alternative System Design

[0047] One alternative embodiment is shown in FIG. 12. In this embodiment, system 1200 shows a solar tower or parabolic trough solar collector 1201 located proximate to a counter current heat exchanger 1202 that reduces the process flow streams 1203 and 1204 to a temperature near ambient temperature so that the process streams connecting the solar collectors to the generating plant will be near ambient temperature. This provides the advantage of reducing cost and complexity of the piping system by eliminating the need for high temperature insulation and high temperature materials. The counter current heat exchanger 1202 can be located at each solar collector unit or at groups of closely spaced solar collectors depending on collection system design and cost requirements. Heat exchanger 1202 could be combined with solar collector 1201 as a single unit or an integrated unit.

[0048] Stream 1204 from the solar collector field will then either split a portion of the flow to the SO<sub>2</sub> and O<sub>2</sub> storage section 1205 or to process stream 1206 and then to heat exchanger 1207. Heat exchanger 1207 could also be a counter current heat exchanger which would increase the temperature of the process stream 1206 to the required temperature for the SO<sub>2</sub> oxidation reactor 1208 where the chemical energy is converted to heat. This temperature of reactor 1208 would be in the range of 350 to 600° C. The SO<sub>3</sub> leaving the SO<sub>2</sub> oxidation reactor 1208 flows through process stream 1210 back to heat exchanger 1207 where the temperature is

adjusted to near ambient temperature for SO<sub>3</sub> storage or return to the solar collector field through process stream 1203. [0049] In one embodiment steam generator section 1209 could consist of a steam turbine and an electric power generator. In another embodiment the heat release and electric power generating section shown as reactor 1208 and steam generator 1209 could be replaced with the direct electrochemical generator 1104 shown in FIG. 11.

[0050] In other embodiments, the heat exchangers shown in FIG. 12 could be co-current heat exchangers or combinations of heat exchangers. Also, the heat exchangers could be separated into several heat exchangers to provide an intermediate temperature for  $SO_2$ ,  $O_2$  and  $SO_3$  storage or for other purposes.

- [0051] Various other adaptations and combinations of features of the embodiments disclosed are within the scope of the invention. Numerous embodiments are encompassed by the following claims.
- 1. A thermochemical system that transports energy from a first location to a second location where the energy is used to generate steam for a steam turbine, comprising:
  - a thermal source providing a first thermal energy;
  - a first reactor at the first location converting the first thermal energy to chemical energy by decomposing SO<sub>3</sub> into SO<sub>2</sub> and O<sub>2</sub>;
  - a second reactor at the second location converting the chemical energy to a second thermal energy by oxidizing SO<sub>2</sub> with O<sub>2</sub> to produce SO<sub>3</sub>, the second reactor evaporating water into the steam for the steam turbine;

liquid SO<sub>2</sub> storage of; and

liquid SO<sub>3</sub> storage of;

- wherein an output of the first reactor is coupled to both an input of the second reactor and the liquid SO<sub>2</sub> storage, and an output of the sec reactor is coupled to both an input of the first reactor and the liquid SO<sub>3</sub> storage;
- wherein a flow split of SO<sub>2</sub> and O<sub>2</sub> from the first reactor to the second reactor and the liquid SO<sub>2</sub> storage is variable, and a flow split of SO<sub>3</sub> from the second reactor to the first reactor and the liquid SO<sub>3</sub> storage is variable.
- 2. The system of claim 1 wherein the thermal source is a solar concentrator that focuses sunlight on a receiver to produce a temperature high enough to decompose SO<sub>3</sub> into SO<sub>2</sub>.
- 3. The system of claim 1 wherein at least one of the first reactor and the second reactor comprises a catalyst for converting SO<sub>3</sub> into SO<sub>2</sub> and O<sub>2</sub> or SO<sub>2</sub> and O<sub>2</sub> to produce SO<sub>3</sub>.
- 4. The system of claim 1 wherein the first reactor converts  $SO_3$  into  $SO_2$  and  $O_2$  at a temperature in the range of 600 to  $1200^{\circ}$  C.
- 5. The system of claim 1 wherein the second reactor converts  $SO_2$  and  $O_2$  to produce  $SO_3$  at a temperature below about  $800^{\circ}$  C.
- 6. The system of claim 1 further comprising an oxygen storage material that chemically binds and stores a substantial portion of  $O_2$  produced by the first reactor converting  $SO_3$  into  $SO_2$  and  $O_2$ .
- 7. The system of claim 1 wherein pressures in the first reactor and the second reactor are maintained in the range of 1 bar to 100 bar.
- **8**. The system of claim 7 wherein the pressures are maintained in the range of 5 to 15 bar.
- 9. The system of claim 6 wherein at least one of the oxygen storage material and a temperature of the oxygen storage material is chosen to adsorb and release  $O_2$  at an  $O_2$  partial pressure in the range of 0.01 to 3 bar.

- 10. The system of claim 1, further comprising:
- a first electrochemical cell being located downstream of the first reactor, the first electrochemical cell being exposed on one side to a process stream and on the other side to the ambient air, the first electrochemical cell removing O<sub>2</sub> formed by the first reactor from the process stream and pumps it into the ambient air; and
- a second electrochemical cell being located upstream of the second reactor, the second electrochemical cell begin exposed on one side to the process stream and on the other side to the ambient air, the second electrochemical cell pumping O<sub>2</sub> from the ambient air into the process stream to be used by the second reactor.
- 11. The system of claim 1, further comprising:
- a first electrochemical cell being located downstream of the first reactor, the first electrochemical cell being exposed on one side to a process stream and on the other side to a vessel containing an oxygen storage material; and
- a second electrochemical cell being located upstream of the second reactor, the second electrochemical cell being exposed on one side to the process stream and on the other side to an other vessel containing an other oxygen storage material wherein:
  - the first electrochemical cell removes O<sub>2</sub> formed decomposing SO<sub>3</sub> into SO<sub>2</sub> and O<sub>2</sub> from the process stream and pumps it into the vessel containing the oxygen storage material;
  - the second electrochemical cell pumps  $O_2$  from the other vessel containing the other oxygen storage material into the process stream to be used in oxidizing  $SO_2$  and  $O_2$  to produce  $SO_3$ .
- 12. The system of claim 1 wherein the first reactor includes an electrochemical cell that removes oxygen from the first reactor to allow a higher conversion of SO<sub>3</sub> to SO<sub>2</sub>.
- 13. The system of claim 1, wherein the second reactor includes an electrochemical cell that produces electrical power from the conversion of SO<sub>2</sub> and O<sub>2</sub> to produce SO<sub>3</sub>.
- 14. A method of transporting energy from a first location to a second location where the energy is used to generate steam for a steam turbine, the method comprising:
  - converting the thermal energy at the first location to chemical energy in a first reactor by decomposing SO<sub>3</sub> into SO<sub>2</sub> and O<sub>2</sub>;
  - converting the chemical energy to one of thermal energy or electrical energy in a second reactor at the second location by oxidizing SO<sub>2</sub> with O<sub>2</sub> to produce SO<sub>3</sub>;
  - using stored SO<sub>3</sub> liquid for the first reactor and storing a portion of SO<sub>2</sub> from the first reactor as a liquid when more energy is available at the first location then is required at the second location; and
  - using stored SO<sub>2</sub> liquid for the second reactor and storing SO<sub>3</sub> liquid from the second reactor when more energy is needed at the second location then is available at the first location;
  - wherein using stored SO<sub>3</sub>, storing a portion of the SO<sub>2</sub>, using stored SO<sub>2</sub> liquid, and sorting SO<sub>3</sub> liquid comprise:
    - controlling a flow split of SO<sub>2</sub> and O<sub>2</sub> from the first reactor to the second reactor and a liquid SO<sub>2</sub> storage; and
  - controlling a flow split of SO<sub>3</sub> from the second reactor to the first reactor and a liquid SO<sub>3</sub> storage.

- 15. The method of claim 14, further comprising using a solar concentrator to focus sunlight on a receiver to produce a temperature high enough to decompose SO<sub>3</sub> into SO<sub>2</sub>.
- 16. The method of claim 14 wherein at least one of the first reactor and the second reactor comprise a catalyst for converting SO<sub>3</sub> into SO<sub>2</sub> and O<sub>2</sub> or SO<sub>2</sub> and O<sub>2</sub> to produce SO<sub>3</sub>.
- 17. The method of claim 14, further comprising operating first reactor at a temperature in the range of 600 to 1200° C.
- 18. The method of claim 14, further comprising operating second reactor at a temperature below about 800° C.
- 19. The method of claim 14, further comprising chemically binding at least a portion of  $O_2$  produced in the first reactor by the decomposition of  $SO_3$  into  $SO_2$  and  $O_2$  in an oxygen storage material.

- 20. The method of claim 14, further comprising maintaining pressures in the first reactor and the second rector are in the range of 1 to 100 bar.
- 21. The method of claim 20 wherein the pressures are maintained in the range of 5 to 15 bar.
  - 22. The method of claim 14, further comprising:
  - pumping, with a first electrochemical cell, O<sub>2</sub> produced in the first reactor by the decomposition of SO<sub>3</sub> into SO<sub>2</sub> and O<sub>2</sub> to one of ambient air and a vessel containing an oxygen storage material;
  - pumping, with a second electrochemical cell, O<sub>2</sub> from one of the ambient air and another vessel containing another oxygen storage material to react with SO<sub>2</sub> in the second reactor to produce SO<sub>3</sub>.

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