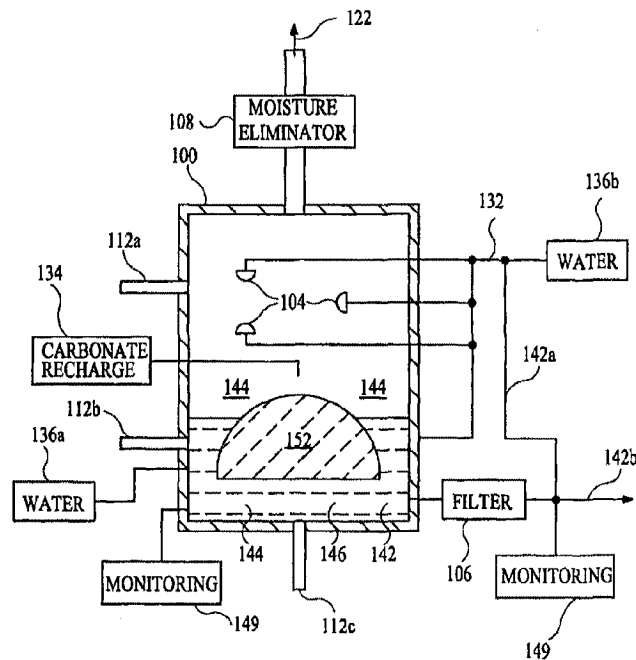


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(54) Title: METHOD AND APPARATUS FOR EXTRACTING AND SEQUESTERING CARBON DIOXIDE

(57) Abstract

A method and apparatus to extract and sequester carbon dioxide (CO<sub>2</sub>) from a stream or volume of gas (112a) wherein said method and apparatus hydrates CO<sub>2</sub>, and reacts the resulting carbonic acid (144) with carbonate (152). Suitable carbonates include, but are not limited to, carbonates of alkali metals and alkaline earth metals, preferably carbonates of calcium and magnesium. Waste products are metal cations and bicarbonate in solution (146), providing an effective way of sequestering CO<sub>2</sub> from a gaseous environment.



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METHOD AND APPARATUS FOR EXTRACTING  
AND SEQUESTERING CARBON DIOXIDE

STATEMENT OF GOVERNMENT INTEREST

5           The United States Government has rights in this invention pursuant to Contract No. W-7405-ENG-48 between the U.S. Department of Energy and the University of California.

CROSS-REFERENCE TO RELATED APPLICATIONS

10           This application claims the benefit of U.S. Provisional Application No. 60/096,846, filed 8/18/98, and U.S. Application No. 09/314,220, filed 5/19/1999.

TECHNICAL FIELD

15           The present invention relates generally to a method and apparatus for extracting carbon dioxide (CO<sub>2</sub>) from a stream or volume of gas, and sequestering said CO<sub>2</sub> from the atmosphere or other gaseous environment. The invention particularly relates to a method and apparatus that utilize carbonate and water to sequester said CO<sub>2</sub> as bicarbonate.

BACKGROUND ART

20           A variety of chemical means exist or have been proposed which consume CO<sub>2</sub> contained in emissions from fossil fuel combustion or other gas streams, thus reducing the potential atmospheric CO<sub>2</sub> burden (reviews by: H. Herzog and E. Drake, "Carbon Dioxide Recovery and Disposal From Large Energy Systems", Annual Reviews of Energy and Environment Vol. 21, p 145-166, 1996; X. Xiaoding and J.A. Moulijn, "Mitigation of CO<sub>2</sub> by Chemical Reactions and Promising Products", Energy and Fuels, Vol. 10, p 305-325, 1996).

25           Among these chemical approaches, the exposure and reaction of such waste CO<sub>2</sub> to certain naturally occurring or artificially formed calcium-, magnesium-, sodium-, and/or silica-rich minerals has been explored as reviewed below. The reaction of certain carbonate and silicate minerals with CO<sub>2</sub> is a well-known "rock weathering" phenomenon that plays a major role in controlling atmospheric CO<sub>2</sub> on geologic time scales (R.A. Berner, A.C. Lasaga, and R.M.

30           Garrels, "The Carbonate-Silicate Geochemical Cycle and its Effect on Atmospheric Carbon Dioxide Over the Last 100 Million Years", American Journal of Science, Vol. 283, p 42-50, 1983). Over the very long term such process are expected to eventually consume most of the CO<sub>2</sub> emitted by man's activities. The problem is that such natural processes occur on the order of >1,000 year time scales and thus will have little immediate impact on the rapidly increasing

CO<sub>2</sub> emissions and atmospheric CO<sub>2</sub> burden in the coming centuries. Nevertheless, several researchers have proposed that certain weathering reactions be used to sequester CO<sub>2</sub>, in particular those reactions which lead to CO<sub>2</sub> sequestration or storage in the form of solid carbonates.

5           For example, fixation and storage of CO<sub>2</sub> by artificial weathering of waste concrete in combination with coccolithophorid algae cultures was reported by H. Takano and T. Matsunaga, "CO<sub>2</sub> Fixation by Artificial Weathering of Waste Concrete and Coccolithophorid Algae Cultures", *Energy Conversion Management*, Vol. 36, No. 6-9, p 697-700, 1995. It was shown that CO<sub>2</sub> can be sequestered into biologically produced carbonate and biomass. Various  
10 mechanisms of rock weathering to fix CO<sub>2</sub> was discussed by T. Kojima, "Evaluation Strategies for Chemical and Biological Fixation/Utilization Processes of Carbon Dioxide", *Energy Conversion Management*, Vol. 36, No. 6-9, p 881-884, 1995. Studies of CO<sub>2</sub> fixation by silicate rock weathering were reported by T. Kojima, A. Nagamine, N. Ueno and S. Uemiya, "Absorption and Fixation of Carbon Dioxide by Rock Weathering", *Energy Conversion  
15 Management*, Vol. 38, Suppl., p S461-S466, 1997. Sequestering of CO<sub>2</sub> as carbonate by reaction with minerals rich in calcium and magnesium oxides was reported by K.S. Lackner, C.H. Wendt, D.P. Butt, E.L. Joyce, D.H. Sharp, "Carbon Disposal in Carbonate Minerals", *Energy*, Vol. 20, No. 11, p 1153-1170, 1995. Reacting flue gas CO<sub>2</sub> with water and soil to ultimately precipitate and sequester the CO<sub>2</sub> as carbonate was explored by T. Chohji, M.  
20 Tabata, and E. Hirai, "CO<sub>2</sub> Recovery From Flue Gas by an Ecotechnological (Environmentally Friendly) System", *Energy*, Vol. 22 No. 2/3, p 151-159, 1997. A study by H. Kheshgi ("Sequestering Atmospheric Carbon Dioxide by Increasing Ocean Alkalinity", *Energy*, Vol. 20, No. 9, p 912-922, 1995) looked at the option of adding calcium oxide to the ocean as a means of increasing the CO<sub>2</sub> absorption capacity of the ocean. The preceding approaches often  
25 require elevated temperatures or pressures, significant energy, land, or other resource inputs, and/or have negative environmental impacts. The cost of implementing these technologies is therefore often prohibitive.

As reviewed by H. Herzog and E. Drake, (*Annual Reviews*, loc. cit.) several chemical means exist for separating and concentrating CO<sub>2</sub> from gas streams. U.S. Patent 4,376,101  
30 (Sartori et al) discloses the removal of CO<sub>2</sub> from a gaseous stream via use of an aqueous solution containing an alkali metal salt or hydroxide and an activator or promoter system comprising an amine compound. While such processes remove or separate CO<sub>2</sub> from a waste stream, they offer no downstream method of ultimately sequestering the CO<sub>2</sub> from the

atmosphere. They also often require elevated temperatures or pressures, exotic chemicals, and/or significant inputs of energy or resources.

Gas/water/calcium carbonate (limestone) reactors have been used in desulfurization of power plants exhaust as reviewed by H. N. Soud and M. Takeshita, "FGD Handbook, IEA Coal Research, London, 438p., 1994. Such reactors differ from the present invention in three important aspects: 1) The volume of SO<sub>2</sub> in the gas streams to which desulfurization is applied is vastly smaller than the CO<sub>2</sub> content in the same gas stream; 2) The hydration step in carbonate desulfurization involves combining SO<sub>2</sub> with H<sub>2</sub>O to form the strong acid H<sub>2</sub>SO<sub>3</sub>. In contrast, the hydration of CO<sub>2</sub> envisioned here forms carbonic acid H<sub>2</sub>CO<sub>3</sub>, a weak acid which has a slower reaction rate with carbonate than does H<sub>2</sub>SO<sub>3</sub>. 3) The reaction of H<sub>2</sub>SO<sub>3</sub> with carbonate (e.g., CaCO<sub>3</sub>) and oxygen forms a solid, CaSO<sub>4</sub>, and a gas, CO<sub>2</sub>, whereas the H<sub>2</sub>CO<sub>3</sub> with carbonate reaction forms cations and bicarbonate in solution, does not require supplemental oxygen, produces little or no solid waste, and consumes rather than generates gaseous CO<sub>2</sub>.

U.S. Patent 5,100633 (Morrison) describes a process for scrubbing acid-forming gases which include SO<sub>2</sub> and CO<sub>2</sub> from an exhaust gas stream through reactions with alkaline solutions formed from the waste ash from biomass burning. The resulting alkali metal salts are then precipitated or dewatered forming solid, possibly useful waste products. This process does not provide a system for net CO<sub>2</sub> sequestration, however, considering that the molar ratio of carbon to alkali metals or to alkaline earth metals in the end products is many times lower than that ratio in the original biomass burned to form the alkaline ash. That is, only a very small fractional equivalent of the CO<sub>2</sub> released in biomass combustion can be sequestered by this process, and therefore when initial ash and CO<sub>2</sub> formation are considered the overall process is a net source rather than a net sink for CO<sub>2</sub>.

The chemical reactions involving CO<sub>2</sub> gas, water, and carbonate minerals (principally calcium carbonate) have been extensively studied as reviewed by J.W. Morse and F.T. Mackenzie ("Geochemistry of Sedimentary Carbonates", Cambridge, Amsterdam, 707p., 1990) and by T. Arakaki and A. Mucci ("A Continuous and Mechanistic Representation of Calcite Reaction-Controlled Kinetics in Dilute Solutions at 25°C and 1 Atm Total Pressure", Aquatic Geochemistry, Vol. 1, p 105-130, 1995). However, the context of these studies has been to describe the dissolution or precipitation of solid carbonate under various conditions, not the consumption and sequestration of CO<sub>2</sub>.

#### DISCLOSURE OF THE INVENTION

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