[0004] It has been predicted that over the coming few decades (2010-2060), the cumulative amount of atmospheric CO_2 will increase, up to approximately 496 gigatons, due to fossil fuel combustion in the existing infrastructure. Thus, there is an urgent need for both alternative energy sources and improved control of the rate of CO_2 emissions. However, due to high global energy demands, there is no immediate alternative to replace or substantially reduce production of fossil fuels. The problem is further exacerbated by the low cost of fossil fuels. Fortunately, research into the electrocatalytic reduction of CO_2 has produced a remarkable number of advances over the past few years, yet there is still no known solution that can harvest CO_2 directly from the air and other point sources and convert collected CO_2 emissions into value-added chemicals.

SUMMARY

[0004a] According to a first aspect of the present invention, there is provided an active CO_2 capture unit for capturing CO_2 from an input gas, comprising:

an inlet through which an input gas is introduced into the unit;

a non-aqueous region comprising a non-aqueous CO_2 binding organic liquid containing OH^- arranged to be in contact with the input gas to chemisorb CO_2 from the input gas and convert the chemisorbed CO_2 into HCO_3^- by reacting with OH^-;

an aqueous region arranged downstream of the non-aqueous region, wherein at an aqueous region interface, the HCO_3^- interacts with H_2O and decomposes to CO_2 and CO_3^{2-};

an anion exchange membrane disposed between the non-aqueous region and the aqueous region to facilitate HCO_3^- diffusion and migration from the non-aqueous region to the aqueous region; and

a captured CO_2 outlet disposed downstream of the aqueous region.

[0004b] According to a second aspect of the present invention, there is provided an active CO_2 capture unit for capturing CO_2 from an input gas, comprising:

an inlet through which an input gas is introduced into the unit;

a non-aqueous, dry region into which the input gas is introduced,

a cathode arranged upstream of the dry region and in fluid communication with an H_2O source and in fluid communication with the dry region, such that the cathode is configured to decompose H_2O received from the H_2O source into H_2 and OH^- and the OH^- is flowed to the dry region, wherein in the dry region OH^- interacts with the input gas to chemisorb CO_2 from the input gas and convert the chemisorbed CO_2 into HCO_3^- by reacting with OH^-;

an aqueous region comprising a humidified gas arranged downstream of the non-aqueous region, wherein at an aqueous region interface, the HCO_3^- interacts with H_2O and decomposes to CO_2 and CO_3^{2-};

an anion exchange membrane disposed between the non-aqueous region and the aqueous region to facilitate HCO_3^- diffusion and migration from the non-aqueous region to the aqueous region;

an anode arranged downstream of the aqueous region; and

a captured CO_2 outlet disposed downstream of the aqueous region.

[0004c] According to a third aspect of the present invention, there is provided a system for active CO_2 capture and CO_2 reduction to a product gas, comprising:

the active CO_2 capture unit of any one of the first and second aspects;

a reduction unit arranged downstream of the active CO_2 capture unit such that the reduction unit receives captured CO_2 from the CO_2 capture outlet, the reduction unit comprising:

a reduction unit inlet for receiving the captured CO_2 from the CO_2 capture outlet, the reduction unit comprising a catalyst for reduction of the captured CO_2 arranged such that the captured CO_2 is flowed through the reduction unit into contact with the catalyst, wherein upon contact with the catalyst, the captured CO_2 is reduced to a product gas comprising one or more of C_2H_4, C_2H_5OH, CH_3COOH, CH_3OH, CH_4, C_3H_6, CO, and H_2;

a catalyst for oxidation of H_2O arranged downstream of the reduction unit and in fluid communication with the reduction unit to generate protons for CO_2 reduction and O_2 as a byproduct, wherein the catalyst for oxidation is an anode;

a separator arranged between the catalyst for reduction and the catalyst for oxidation; and

an energy source.

[0004d] According to a fourth aspect of the present invention, there is provided a method of capturing CO_2 using the active CO_2 capture unit of any one of the first and second aspects, comprising:

flowing the input gas into the inlet and into the non-aqueous region for chemisorbing CO_2 and conversion of the CO_2 to HCO_3^- by OH^- present in the non-aqueous region;

flowing the HCO_3^- across the anionic exchange membrane and into the aqueous region, wherein the flow of HCO_3^- is driven at least in part by a gradient of moisture across the anion exchange membrane, wherein upon flow in the HCO_3^- into the aqueous region, the HCO_3^- interacts with H_2O present in the aqueous region and decomposes to CO_2 and CO_3^{2-}; and

flowing the CO_2 from the aqueous region to the captured CO_2 outlet.

[0004e] According to a fifth aspect of the present invention, there is provided a method for capturing CO_2 and reducing CO_2 to a product gas comprising one or more of C_3H_6, C_2H_4, C_2H_5OH, CH_3COOH, CH_3OH, CH_4, CO, and H_2 using the system of the third aspect, comprising:

flowing the input gas into the inlet and into the non-aqueous region for chemisorbing CO_2 and conversion of the CO_2 to HCO_3^- by OH^- present in the non-aqueous region;

flowing the HCO_3^- across the anionic exchange membrane and into the aqueous region, wherein the flow of HCO_3^- is driven at least in part by a gradient of moisture across the anionic exchange membrane, wherein upon flow in the HCO_3^- into the aqueous region, the HCO_3^- interacts with H_2O present in the aqueous region and decomposes to CO_2 and CO_3^{2-} thereby resulting in captured CO_2;

flowing the captured CO_2 from the aqueous region to the capture CO_2 outlet;

flowing the captured CO_2 from the capture CO_2 outlet into the reduction unit, wherein upon contact with the catalyst the captured CO_2 is reduced to the product gas comprising one or more of C_3H_6, C_2H_4, C_2H_5OH, CH_3COOH, CH_3OH, CH_4, CO, and H_2; and

flowing the product gas to a reduction unit outlet.

[0004f] According to a sixth aspect of the present invention, there is provided a system for capture and reduction of CO_2 from a dilute source, comprising:

a CO_2 capture unit, comprising:

an inlet through which an input gas is introduced into the unit,

a non-aqueous region comprising a non-aqueous CO_2 binding organic liquid containing OH^- arranged to be in contact with the input gas to chemisorb CO_2 from the input gas and convert the chemisorbed CO_2 into HCO_3^- by reacting with OH^-;

an aqueous region arranged downstream of the non-aqueous region, wherein at an aqueous region interface, the HCO_3^- interacts with H_2O and decomposes to CO_2 and CO_3^{2-};

an anion exchange membrane disposed between the non-aqueous region and the aqueous region to facilitate HCO_3^- diffusion and migration from the non-aqueous region to the aqueous region,

a cathode arranged upstream of the capture unit and comprising an H_2O source inlet, an H_2 outlet, and a OH^- outlet in fluid communication with the non-aqueous region of the capture unit to flow OH^- to the non-aqueous region of the capture unit;

a reduction unit arranged immediate downstream of the aqueous region to receive a flow of the captured CO_2 from the capture unit, the reduction unit comprising a catalyst for reduction of the captured CO_2 and a product gas outlet such that the captured CO_2 is flowed through the

reduction unit into contact with the catalyst, wherein upon contact with the catalyst the captured CO_2 is reduced to a product gas comprising one or more of C_2H_4, C_2H_5OH, CH_3COOH, CH_3OH, C_3H_6, CH_4, CO, and H_2 and the product gas if flowed out the product gas outlet;

an anode arranged downstream of the reduction unit and adapted to be a catalyst for oxidation of H_2O wherein oxidation of H_2O generates protons for the reduction unit and O_2 as a byproduct, wherein the anode is in fluid communication with the reduction unit to flow the protons to the reduction unit and an O_2 outlet spatially separate from the product gas outlet to remove O_2;

a separator arranged between the reduction unit and the anode; and

an energy source.

[0005] Water-driven CO_2 capture techniques are attractive for their low energy penalty. Hydrate-based CO_2 separation is a water-driven technology where CO_2 forms hydrates with water or water-miscible solvents under high pressure and can be separated from a feed with a mixture of gasses. Moisture swing technologies capture CO_2 directly from the air where a quaternary amine ion-exchange resin supported on a polymeric backbone acting as an anion-exchange membrane absorbs CO_2 in a water-deprived (dry) environment in the form of bicarbonates and carbonates, and releases it at CO_2 in a wet environment by virtue of the carbonate-bicarbonate equilibrium. This mechanism can be exploited by keeping a constant water-deprived environment on one side of the anion-exchange membrane with a constant supply of CO_2 and a wet environment on the other side, thereby establishing a gradient of concentration of water across the anion-exchange membrane.

[0006] Disclosed herein is an active CO_2 capture unit that utilize a CO_2 binding organic liquid present in a non-aqueous region of the unit to chemisorb CO_2 from an input gas and convert it to HCO_3^-. The unit includes an anion exchange membrane arranged at an interface between the non-aqueous region and an aqueous region to facilitate transfer of the HCO_3^- to the aqueous region where the HCO_3^- interacts with H_2O and is decomposed to CO_2 and CO_3^{2-}.

[0007] Also disclosed herein is a system for CO_2 capture and conversion of the captured CO_2 into one or more of C_3H_6, CH_4, C_2H_4, C_2H_5OH, CH_3OH, CO, H_2 and CH_3COOH. Byproducts of the system can include syngas (CO and H_2) and O_2. Systems of the disclosure can be fully integrated systems that combine CO_2 capture and conversion into a single process. The process can be sustainable and energy-efficient.

WHAT IS CLAIMED:

1. An active CO_2 capture unit for capturing CO_2 from an input gas, comprising:

an inlet through which an input gas is introduced into the unit;

a non-aqueous region comprising a non-aqueous CO_2 binding organic liquid containing OH^- arranged to be in contact with the input gas to chemisorb CO_2 from the input gas and convert the chemisorbed CO_2 into HCO_3^- by reacting with OH^-;

an aqueous region arranged downstream of the non-aqueous region, wherein at an aqueous region interface, the HCO_3^- interacts with H_2O and decomposes to CO_2 and CO_3^{2-};

an anion exchange membrane disposed between the non-aqueous region and the aqueous region to facilitate HCO_3^- diffusion and migration from the non-aqueous region to the aqueous region; and

a captured CO_2 outlet disposed downstream of the aqueous region.

2. The active CO_2 capture unit of claim 1, further comprising a cathode arranged upstream of the non-aqueous region, an anode arranged downstream of the aqueous region, and wherein the aqueous region comprises an aqueous electrolyte such that an electric field is adapted to be generated within the capture unit.

3. The active CO_2 capture unit of any one of the preceding claims, wherein the non-aqueous CO_2 binding organic liquid comprises an ionic liquid, optionally wherein the ionic liquid is imidazolium based or phosphonium based, or

wherein the non-aqueous CO_2 binding organic liquid comprises one or more ionic liquids selected from the group consisting of choline hydroxide, tetrabutylphosphonium methanesulfonate, 1-Butyl-3-methylimidazolium hexafluorophosphate, optionally wherein non-aqueous CO_2 binding organic liquid comprises a non-aqueous polar organic solvent comprising one or more alcohols, organic amidine bases, and guanidine bases.

4. The active CO_2 capture unit of any one of the preceding claims, wherein the non-aqueous region comprises an alkali metal hydroxide dissolved in the non-aqueous CO_2 binding organic liquid, and/or the aqueous region comprises one or both of an aqueous electrolyte and water.

5. An active CO_2 capture unit for capturing CO_2 from an input gas, comprising:

an inlet through which an input gas is introduced into the unit;

a non-aqueous, dry region into which the input gas is introduced,

a cathode arranged upstream of the dry region and in fluid communication with an H_2O source and in fluid communication with the dry region, such that the cathode is configured to decompose H_2O received from the H_2O source into H_2 and OH^- and the OH^- is flowed to the dry region, wherein in the dry region OH^- interacts with the input gas to chemisorb CO_2 from the input gas and convert the chemisorbed CO_2 into HCO_3^- by reacting with OH^-;

an aqueous region comprising a humidified gas arranged downstream of the non-aqueous region, wherein at an aqueous region interface, the HCO_3^- interacts with H_2O and decomposes to CO_2 and CO_3^{2-};

an anion exchange membrane disposed between the non-aqueous region and the aqueous region to facilitate HCO_3^- diffusion and migration from the non-aqueous region to the aqueous region;

an anode arranged downstream of the aqueous region; and

a captured CO_2 outlet disposed downstream of the aqueous region.

6. The active CO_2 capture unit of claim 5, further comprising an H_2 outlet configured to allow for flow of H_2 generated at the cathode during decomposition of the H_2O away from the cathode.

7. The active CO_2 capture unit of any one of the preceding claims, wherein the input gas is an anthropogenic CO_2 source, optionally wherein the anthropogenic CO_2 source is air or flue gas.

8. The active CO_2 capture unit of any one of the preceding claims, wherein the anion exchange membrane comprises a hollow-fiber structure, or wherein the anion exchange membrane comprises quaternary amines or phosphonium attached to a polymer backbone resin with hydroxide, carbonate, and/or bicarbonate moieties, optionally wherein the polymer backbone is polystyrene.

9. A system for active CO_2 capture and CO_2 reduction to a product gas, comprising:

the active CO_2 capture unit of any one of the preceding claims;

a reduction unit arranged downstream of the active CO_2 capture unit such that the reduction unit receives captured CO_2 from the CO_2 capture outlet, the reduction unit comprising:

a reduction unit inlet for receiving the captured CO_2 from the CO_2 capture outlet, the reduction unit comprising a catalyst for reduction of the captured CO_2 arranged such that the captured CO_2 is flowed through the reduction unit into contact with the catalyst, wherein upon contact with the catalyst, the captured CO_2 is reduced to a product gas comprising one or more of C_2H_4, C_2H_5OH, CH_3COOH, CH_3OH, CH_4, C_3H_6, CO, and H_2;

a catalyst for oxidation of H_2O arranged downstream of the reduction unit and in fluid communication with the reduction unit to generate protons for CO_2 reduction and O_2 as a byproduct, wherein the catalyst for oxidation is an anode;

a separator arranged between the catalyst for reduction and the catalyst for oxidation; and

an energy source.

10. The system of claim 9, further comprising an outlet for the O_2 byproduct arranged spatially separated from an outlet for the product gas.

11. A method of capturing CO_2 using the active CO_2 capture unit of any one of claims 1 to 8, comprising:

flowing the input gas into the inlet and into the non-aqueous region for chemisorbing CO_2 and conversion of the CO_2 to HCO_3^- by OH^- present in the non-aqueous region;

flowing the HCO_3^- across the anionic exchange membrane and into the aqueous region, wherein the flow of HCO_3^- is driven at least in part by a gradient of moisture across the anion exchange membrane, wherein upon flow in the HCO_3^- into the aqueous region, the HCO_3^- interacts with H_2O present in the aqueous region and decomposes to CO_2 and CO_3^{2-}; and

flowing the CO_2 from the aqueous region to the captured CO_2 outlet.

12. The method of claim 11, further comprising flowing H_2O into the cathode for decomposition of the H_2O into H_2 and OH^-, and flowing the OH^- into the non-aqueous region and/ or further comprising applying an electric field across the anion exchange membrane to increase a rate of transfer of HCO_3- across the anion exchange membrane.

13. A method for capturing CO_2 and reducing CO_2 to a product gas comprising one or more of C_3H_6, C_2H_4, C_2H_5OH, CH_3COOH, CH_3OH, CH_4, CO, and H_2 using the system of any one of claims 9 to 10, comprising:

flowing the input gas into the inlet and into the non-aqueous region for chemisorbing CO_2 and conversion of the CO_2 to HCO_3^- by OH^- present in the non-aqueous region;

flowing the HCO_3^- across the anionic exchange membrane and into the aqueous region, wherein the flow of HCO_3^- is driven at least in part by a gradient of moisture across the anionic exchange membrane, wherein upon flow in the HCO_3^- into the aqueous region, the HCO_3^- interacts with H_2O present in the aqueous region and decomposes to CO_2 and CO_3^{2-} thereby resulting in captured CO_2;

flowing the captured CO_2 from the aqueous region to the capture CO_2 outlet;

flowing the captured CO_2 from the capture CO_2 outlet into the reduction unit, wherein upon contact with the catalyst the captured CO_2 is reduced to the product gas comprising one or more of C_3H_6, C_2H_4, C_2H_5OH, CH_3COOH, CH_3OH, CH_4, CO, and H_2; and

flowing the product gas to a reduction unit outlet.

14. A system for capture and reduction of CO_2 from a dilute source, comprising:

a CO_2 capture unit, comprising:

an inlet through which an input gas is introduced into the unit,

a non-aqueous region comprising a non-aqueous CO_2 binding organic liquid containing OH^- arranged to be in contact with the input gas to chemisorb CO_2 from the input gas and convert the chemisorbed CO_2 into HCO_3^- by reacting with OH^-;

an aqueous region arranged downstream of the non-aqueous region, wherein at an aqueous region interface, the HCO_3^- interacts with H_2O and decomposes to CO_2 and CO_3^{2-};

an anion exchange membrane disposed between the non-aqueous region and the aqueous region to facilitate HCO_3^- diffusion and migration from the non-aqueous region to the aqueous region,

a cathode arranged upstream of the capture unit and comprising an H_2O source inlet, an H_2 outlet, and a OH^- outlet in fluid communication with the non-aqueous region of the capture unit to flow OH^- to the non-aqueous region of the capture unit;

a reduction unit arranged immediate downstream of the aqueous region to receive a flow of the captured CO_2 from the capture unit, the reduction unit comprising a catalyst for reduction of the captured CO_2 and a product gas outlet such that the captured CO_2 is flowed through the reduction unit into contact with the catalyst, wherein upon contact with the catalyst the captured CO_2 is reduced to a product gas comprising one or more of C_2H_4, C_2H_5OH, CH_3COOH, CH_3OH, C_3H_6, CH_4, CO, and H_2 and the product gas if flowed out the product gas outlet;

an anode arranged downstream of the reduction unit and adapted to be a catalyst for oxidation of H_2O wherein oxidation of H_2O generates protons for the reduction unit and O_2 as a byproduct, wherein the anode is in fluid communication with the reduction unit to flow the protons to the reduction unit and an O_2 outlet spatially separate from the product gas outlet to remove O_2;

a separator arranged between the reduction unit and the anode; and

an energy source.

15. The system of claim 9 or 14, wherein the catalyst for reduction is a copper mesh; optionally wherein the copper mesh has a mesh size of about 40 to about 120 mesh; or

wherein the catalyst comprises metal nanocrystals, optionally wherein the metal nanocrystals comprise Cu nanocrystals, optionally wherein the Cu nanocrystals are Cu nanocubes having dominate (100) facets, optionally wherein the metal nanocrystals are grown on a mesh substrate; or

wherein the catalyst for oxidation comprises one or more oxides of Ni, Fe-Ni, Pt-coated Ti, Ir, and Ru.

16. The system of any one of claims 9, 14 or 15, wherein the input gas is an anthropogenic CO_2 source selected from air or flue gas.

17. The system of any one of claims 9 or 14 to 16, wherein the anion exchange membrane comprises a hollow-fiber structure, or

wherein the anion exchange membrane comprises quaternary amines or phosphonium attached to a polymer backbone resin with hydroxide, carbonate, and/or bicarbonate moieties, optionally wherein the polymer backbone is polystyrene.

18. The system of any one of claims 9 or 14 to 17, wherein the aqueous region comprises an aqueous electrolyte such that an electric field is adapted to be generated within the capture unit; and/or

wherein the non-aqueous region comprises an alkali metal hydroxide dissolved in the non-aqueous CO_2 binding organic liquid.

19. The system of any one of claims 9 or 14 to 18, wherein the non-aqueous CO_2 binding organic liquid comprises one or more ionic liquids selected from the group consisting of choline hydroxide, tetrabutylphosphonium methanesulfonate, and 1-Butyl-3-methylimidazolium hexafluorophosphate.

20. The system of claim 19, wherein the non-aqueous CO_2 binding organic liquid comprises one or more non-aqueous polar organic solvents selected from the group consisting of ethylene glycol, methanol, and ethanol.

21. The system of any one of claims 9 or 14 to 20, wherein the aqueous region comprises one or both of an aqueous electrolyte and water.