SHORT COMMUNICATION

An effective and ecofriendly suggestion to decrease atmospheric carbon dioxide by using NH₃ gas

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ABSTRACT: Global warming is increasing permanently, because the concentration of CO₂ in the atmosphere is rising continuously. According to National Oceanographic and Atmospheric Administration, the concentration of CO₂ in the atmosphere was 407 ppm in June 2016 and 413 ppm in April 2017 as a last record for now. If the effects of other greenhouse gases, such as CH₄, N₂O, SF₆, NF₃, chlorofluorocarbons, hydrofluorocarbons, perfluorocarbons are added, the effective concentration may reach or exceed 550 ppm CO₂-equivalent. According to the United Nations Intergovernmental Panel on Climate Change-2014 Climate Change Report, this is about two times higher than 278 ppm CO₂ concentration in the pre-industrial year 1765. Thus, very urgent solutions must be found. The aim of this article is to suggest a vital, fast and very meticulous solution using NH₃ gas in the atmosphere in order to decrease the atmospheric CO₂ without delay. The laboratory experiments in the gas phase for (NH₃+CO₂) reaction showed us that to use NH₃ gas in the atmosphere will be a very fast, effective method for decreasing CO₂ concentration of atmosphere. (NH₃+CO₂) reaction is also quantitative in the cold atmosphere strata and there will be no more free ammonia in the atmosphere and no public health problem.

KEYWORDS: Atmospheric CO₂; Chemical CO₂absorption; Decreasing CO₂concentration; Global warming; NH₃ gas.

INTRODUCTION

In June 2016, the concentration of CO_2 in the atmosphere reached 407 ppm and in April 2017 reached 413 ppm according to NOAA. Measurements were made by two independent CO_2 monitoring programs (NOAA and Scripps) at the Mauna Loa Observatory in Hawaii, about 3400 meters above sea level. In the 2015 Paris Climate Conference (COP 21), the World Leaders agreed to stabilize global warming at maximum 2°C above pre-industrial level, at the end of the twenty-first century. But, if GHGs emissions increase with the present growth rates, "IPCC 2014- AR5-Scenerio RCP 8.5" says

*Corresponding Author Email: h.eryilmaz@artvin.edu.tr Tel.: +90 4662151040 Fax: +90 4662151057 that the warming will be about 4° C (2.6-4.8°C) (IPCC, 2014) at the end of the twenty-first century. Additionally, if all "Intended Nationally Determined Contributions" (INDCs) of UN countries to mitigate GHGs emissions are calculated totally, it seems that the global warming will be 2.7-3.0 °C, i.e. certainly above 2°C. So, COP 21 Agreement could not solve the problem exactly, but simply postponed it, to solve after 2020s. Also, the withdrawal of the USA from the Paris Climate Treaty in 2017 makes the situation even more difficult. For a successful worldwide solution, the lifestyle of human being should be changed. This means using less consumption goods and especially, saving as much as possible the carbon based energy. Apart from these precautions, science and technology can provide more effective methods in solving this

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problem. For instance, some of them are Carbon Capture and Storage (CCS) methods (Spigarelli and Kawatra, 2013; Zaman and Lee, 2013; Wang *et al.*, 2011). In the specialized literature, there is a wide and comprehensive review (Boot-Handford *et al.*, 2014) for these CCS procedures. These methods can be divided into two main groups:

1) Physical methods: Liquefaction of CO₂ at high pressure and low temperature is a known process and it may be possible for gas mixtures with a high percentage of CO₂ but, not proper for atmospheric CO₂ because of its very low concentration. It is necessary to use some physical adsorbents in this case. A large number of adsorbents have been proposed for CCS. The widely investigated adsorbents are aluminosilicatebased zeolites, metal-organic frameworks, activated carbons, carbon-based decomposition materials, and amine-based materials, amine-modified mesoporous silicas. Another alternative physical method is separation of CO₂ from gas mixtures with membranes made from polyphenyleneoxide, polymethylsiloxane, polypropelen, ceramic (Rubin et al., 2012), polyimide, polycarbonate, polyacetylene, poly ethylene oxide, poly aniline, poly sulfone, etc. (Spigarelli and Kawatra, 2013). Absorptions in a solvent (for example; selexol, rectisol, fluor, purisol, etc.) under high pressure-low temperature and then regenerations of CO₂ with pressure-swing or temperature-swing are also investigated methods. But operations need high pressures and usually, solvents are not cheap (Spigarelli and Kawatra, 2013).

2) Chemical absorption methods: In the case of chemically CO₂ absorption CCS methods, it is necessary to use some basic materials to bind the atmospheric CO₂ directly as a chemical compound. Some of them are MgO, CaO, Ca(OH), NaOH, Na, CO, K₂CO₃, NH₃, organic amine compounds (Piperazine (PZ), 2-Methyl PZ, Aminoethyl PZ, Hydroxyethyl PZ, 2-Amino-2-methyl propanol, Monoethanolamine (MEA), Methyldiethanolamine, bis-Aminoethylether, etc.),ionic liquids (as cations: 1,3-dialkylimidazolium, N,N-dialkylpyrrolidinium, alkylopyridinium, tetraalkylammonium, tetraalkylphosphonium, etc. and as anions: bis(trifluoromethylsulfonyl)imide, trifluoromethanesulfonate, hexafluorophosphate, tetrafluoroborate, etc.), But these CCS methods are applicable only for power plants using huge amounts

carbondioxide capturing basic compounds, ammonia is a very cheap one and very much on the market and so, there are very many articles about it, but all of them are with aqueous NH₃ solution in liquid phase (Darde et al., 2010; Darde et al., 2012; Zhao et al., 2012; Chen et al., 2012; Dong et al., 2012; Darde et al., 2012(10); Shuangchen et al., 2013; Han et al., 2013; Zhang and Guo, 2013; Li et al., 2014; Li et al., 2014; Qi and Wang, 2017). NH₃ has minimum molecular weight (MW), and because of this, has the biggest theoretical Carbon Capture Capacity (CCC) as (kg CO₂/kg base). One of these amine compounds, monoethanolamine (MEA) which is mostly used base for CCS, has only 0.72 CCC but NH, has bigger 2.59 CCC, if calculated 1 mol base for 1 mol CO₂ theoretically. It means that NH, has 3.6 times CCC of MEA. But in practice, only 3.0 times CCC has been reached with NH, solution (Yeh et al., 2005), just a little less than theoretical 3.6 value. Another advantage is that the price of NH₃ is about one fifth of the MEA's price. So, regardless of other financial factors, the NH, method will be fifteen $(3 \times 5 = 15)$ times cheaper than the MEA method approximately. Additionally, MEA and its bicarbonate salt are poisonous compounds and cannot be used directly. Separation of CO₂ from the MEA salt and regeneration of MEA require a lot of energy. It has been estimated that its energy consumption is about 1/3 of a power plant (Spigarelli and Kawatra, 2013). But in the NH₃ method, regeneration will be not necessary. After capture of CO_{2} , its storage is necessary. Several ideas have been proposed for the long-term storage of captured CO₂. These proposals include storing CO₂ in various geologic formations e.g., oil and gas fields, coal beds, and saline aquifers, injecting CO, into the deep ocean.Unfortunately, because of ocean currents and supersaturation in deep waters, a large fraction of the injected CO₂ will be released to the atmosphere after a few hundred years. Additionally, direct ocean storage may affect marine-ecosystems. In CO₂ injection into the deep ocean, CO₂ hydrates $(5.75 \text{ H}_2\text{O} \cdot \text{CO}_2)$ are formed. These compoundes are nonstoichiometric crystalline compounds that form at high pressures and low temperatures by trapping CO, molecules in hydrogen-bonded cages of H₂O (House et al., 2006). The researching of CO₂ hydrate stability is still going on. The effect of pressure (100-250 bar), temperature (274-288 K), and salinity (3.5%

of fossil fuel, or for extremely big industrial plants

producing iron, steel, cement etc. Among these

w/w electrolytes) on hydrate dissolution rates in the ocean have been investigated (Daeseung et al., 2015). They found that higher pressure, lower temperature and high salinity significantly decrease the hydrate breakdown and dissolution. In the NH, method, pH of sea water and marine-eco system will not change. Because, CO₂ is an acidic substance, NH₃ is a basic substance, but NH4HCO3 formed from CO2 and NH₃ is a neutral substance. As can be found in the catalogs of chemical companies such as Merck; J.T. Baker; Sigma-Aldrich etc., pH of a 0.1 N solution of NH₄HCO₂ in water is 7.8 at 25 °C. Also seawater pH was given to a range between 7.8 and 8.0 (Ohline et al., 2007). Clearly, 7.8 is approximately between 7.5-8.0 and the concentration of NH₄HCO₃ in sea waters will be always under 0.1 N. Thus, the pH of sea water and marine-eco system will not be changed.

Chemical data for the NH, method

The first study using aqueous NH₃ for CCS was done in 1997 (Bai and Yeh, 1997). But, the following data have been received from (Ma et al., 2014): If CO₂ gas is passed through aqueous NH₃ solution, mainly NH₄HCO₃ is formed. The other particles in the resulting solution are CO₂, NH₃, O₂, H₂O, NH₄⁺, H⁺, OH⁻, NH₂COO⁻, HCO₃⁻, and CO_3^{2-} . In the simplest way, reaction steps are (CO_2) \rightarrow NH₂COOH \rightarrow NH₂COONH₄ \rightarrow NH₄HCO₃). The yield of CO₂ absorption is 90% at 54°C temperature and in 4-5 seconds. The absorption temperature is a very important factor in the yield. According to another study (Yeh et al., 2005); if the 40°C is decreased to 10°C, the yield increases by 12%, in 28% NH₃ solution. The thermal decomposition of 20% NH₄HCO₃ solution is (0% at 4°C), (6% at 49°C), (16 % at 60°C), and (29% at 71°C). This means that NH₄HCO₂ is very stable in the cold. Also, the Standard Free Energy of formation value, ΔG° , for NH₄HCO₃(s) is $\Delta G^{\circ} = -26.5$ kcal/mol (Zhang et al., 2003). These value means that the reaction takes place spontaneously and easily. Especially in the cold conditions, it shifts completely to the right side. NH₂COONH₄ formation reaction is very fast under atmospheric pressure and in room temperature, because of its low activation energy, 48.6 kJ/mol (Ma et al., 2013). The reaction rate is as Eq. 1.

$$R_{R} = 1.66 \text{ x } 10^{11} \exp\left(\frac{-6100}{RT}\right) [CO_{2}] [NH_{3}] \text{ mol } m^{3}/\text{s}$$
 (1)

Also, NH_4HCO_3 , formation (the last step) is a fast

reaction with the low activation energy. As a result, all steps of NH_4HCO_3 formation from $(CO_2 + NH_3)$ are fast and completely resulted reaction.

Suggestion of a new method to decrease atmospheric CO, with NH₂

The CCS studies are still in progress, but for the time being, these methods cannot be used for CO₂ emitted from vehicles, buildings, etc. and not also for atmospheric CO₂. Although it is believed that CCS difficulties may be solved in the following years, the decrease in GHGs emissions will not be sufficiently enough, and the concentration of CO₂ in the atmosphere will be higher and higher. CO, concentration emission from various sources can be found in IPCC 2014 AR5 report. Also, daily atmospheric CO₂ concentrations can be taken from the web page, (https://www.co2.earth/global-co2emissions). In 2010, total World GHGs emission was 49 Gt CO₂-equivalent and only 76 % of it was CO₂ in itself, according to IPCC 2014 AR5 report (IPCC, 2014). And for the same year 2010, the atmospheric CO₂ concentration was 388 ppm according to NOAA. So, we can make a prediction using the values given by these two authorized organizations. Assuming that the ratio 76 %, above mentioned for year 2010, was the same ratio in April 2017; and the concentration of CO₂ was 413 ppm in April 2017 as mentioned above, it can be calculated easily with a simple mathematical proportion that, the total effective GHGs concentration in the year 2017 was approximately 543 ppm CO₂equivalent as shown in Eq. 2.

$(413 \text{ ppm CO}_2) \times (100 / 76) = 543 \text{ ppm CO}_2$ -equivalent (2)

This is about 2 times that of the pre-industrial 278 ppm. In Fig.1, the NOAA graphics show that, this calculation approximately is correct; and nowadays, total effective GHGs concentration must be around 500 ppm as a prediction. In terms of CO_2 equivalents, all GHGs in the atmosphere in 2016 were 489 ppm as shown in Fig. 1, of which 403 ppm was CO_2 alone, and the rest 86 ppm was from other GHGs. Certainly, the global warming will increase constantly under these circumstances.

In that case, what is the solution? Although the forests and the oceans absorb the atmospheric $CO_{2,}$ this is not enough to overcome the problem. One of the appropriate ways, perhaps the best way for

decreasing CO₂, seems to use NH₃ gas in the upper layers of the atmosphere directly, by beginning with small amounts of NH₃ and then increasing its dose gradually. The reaction of NH, solution with CO_2 to produce NH₄HCO₃ is a known reaction in laboratory and industry. Because of this, we have repeated this reaction in laboratory, only in the gas phase, but not in a solution. It is necessary to apply the experiments in the high altitude of atmosphere to make a complete and perfect method. However, these experiments have not been performed due to the inadequate opportunities. Perhaps, it may be possible for us later in the further studies. Thus, the experiments of (NH_3+CO_2) gas phase reactions were made only in the laboratory of Artvin Çoruh University, Science-Technology Research and Application Center in 2018.

MATERIALS AND METHODS

The gas phase reaction of (NH_2+CO_2) was made in a glass desiccator. "Air CO₂ntrol 3000" instrument of Dostmann Electronic Gmbh was used to measure the CO₂ concentration of the air in the desiccator. A firefighting tube was used as CO₂ source in the desiccator. In the beginning, 25% NH₃ solution in a test tube and the CO₂ measuring instrument were placed in an empty desiccator separately and the desiccator was closed tightly. CO, of the air in desiccator was reacted with NH₃ vapors coming from the solution and the values of remaining unreacted CO₂ were read with several minutes' intervals. As shown in Fig. 2 (curve B), decreasing of CO₂ was very slow and, after about six h, became zero. It was very surprising. After similar experiments, the reaction was repeated with 25% NH₂ solution in a wide watch glass, instead of



Fig. 2: Decreasing CO₂ concentrations in time by the reaction with NH₃.

narrow test tube. This time, the reaction took place very fast, because of fast evaporation of NH_3 gas from NH_3 solution in the wide watch glass. The Fig. 2 (curve A) shows the results of the experiment starting with high CO₂ concentration, above 3000 ppm (H in the instrument). It is clear that the reaction will be faster by direct injection of ammonia into the cold atmosphere strata.

RESULTS AND DISCUSSION

It can be seen from Fig. 2 curves that, to use NH_3 gas in the atmosphere will be a very fast, effective method for decreasing CO_2 concentrations and perhaps, stopping global warming. Anyone can think by looking at the curves that, all the CO_2 in the atmosphere will be removed. No, it never happens. Because, the amount of removed CO_2 will be equivalent only to the amount of injected ammonia into the atmosphere. Some advantages of NH_3 gas method can be listed as follows:

- Among the basic substances previously mentioned (MgO, CaO, Ca(OH)₂, NaOH, Na₂CO₃, K₂CO₃, NH₃, amine compounds and zeolites); only NH₃ is a gas, and it diffuses easily in the atmosphere.
- Among them, NH₃ has the minimum MW and the maximum CCC in (kg CO₂ / kg base).

3. Only NH₃ can be sprayed as 100% pure substance. The amine compounds are poisonous. All other basic substances, except NH₂, must be solved or dispersed in water and then sprayed. Because of this dilution, their concentrations will be decreased, and additionally they have bigger MWs than NH,, so their CCC will be much less. For example, MW of NaOH is 40 g/mol, and it can be solved only 42 g in 100 g water at 0°C (temperature of atmosphere around 3 km above). So, 1 kg of saturated 30% NaOH solution can catch only 0.34 kg CO₂ according to (NaOH \rightarrow NaHCO₂) transformation, but pure 1 kg NH, can catch 2.59 kg CO_2 according to $(NH_3 \rightarrow NH_4HCO_3)$ transformation. It is about 7.6 times of that NaOH solution as CCC. So, NH₃ has the largest CCC compared to all other basic substances. In the 3-4 km height of atmosphere, temperature is under 0°C and so, $(NH_3 \rightarrow NH_4HCO_3)$ transformation will be 100% because of negative ΔG° value. Because of the crystal NH₄HCO₃ density is 1.58 g/cm³, NH₄HCO₃ crystals produced in the atmosphere falls into the oceans without or with very little decomposition, and into the warmer lands only with 5-6% decomposition. But, the rest of NH₄HCO₃

511

will be absorbed by the earth and oceans as a fertilizer and because of very low NH4HCO3 concentration, there will not be any eutrophication in the oceans. Some very fine NH₄HCO₂ crystals may be suspended in the air for a while but, because of its high solubility in water, NH4HCO3 will fall into oceans and lands as rain droplets. NH₃ less than 25 ppm in air is not harmful, and in water for example, the average of the mean acute toxicity values for 32 freshwater species is 2.79 mg NH₃/L, about 3 ppm (Serezli et al., 2016). Essentially, free NH, concentration in the atmosphere is very low and under ppm level, because of CO, and other acidic gases. Nowadays, it is about 20 ppb (Vikipedi, 2018). Essentially, NH, is a natural gas. The primary natural resources of NH, gas are agriculture, livestock, plant decomposition, some volatilization from soils and oceans. Even so, its concentration is always under ppm level, because, very high amounts CO_2 and other acidic gases make with it ammonium compounds and in this way, free NH₃ concentration in the atmosphere remains always below the ppm level and completely harmless for public health. After NH, was sprayed in the atmosphere and the reaction completed with excessive CO₂ of the atmosphere, NH₃ concentration of air will also be under ppm level and completely harmless. NH₄HCO₃ falling down on oceans will not change the pH of sea water, because, pH of 0.1 M NH₄HCO₃ solution is only 7.8 at 25°C and approximately, this pH value is the same with pH of sea water. This means that, NH₄HCO₃ is not an acidification material for soil and water. Additionally, it is impossible for NH₄HCO₂ concentration to be more than 0.1 M in the sea water, on the contrary, it will be much less, perhaps in the ppm range. In addition CO₂, other acidic pollutants such as NO_x, SO_x, HF, HCI, HBr, H₂S, and also Hg species will be captured from the atmosphere by NH, because of its basic properties. These ammonium salts, e.g., ammonium sulfate, ammonium nitrate, also scatter incoming solar radiation and make cooling effect on global heating and so, this is an added benefit of the ammonia method (Zhu *et al.*, 2015). NH₄HCO₃, falling down on sea and land, will accelerate (CO₂ Biomass) transformation as an N and C fertilizer, particularly in seas, lakes and oceans. In a new study (Samimi and Moghadam, 2018), an aquatic plant, *Lemna gibba*, has been used very effectively to remove NH, from petrochemical waste water below 15 ppm. This shows also that dilute NH₃ under 15 ppm concentrations in water is certainly

harmless for nature. NH_4HCO_3 is also used both as a fertilizer in the soil and as baking powder in the production of cakes, biscuits, pastry, etc. in industry and at home because of this harmless. It must not be forgotten that, in baking systems, NH_3 concentration is much more than the atmospheric concentration of NH_3 gas.

Application conditions

A cold and moist atmosphere is necessary to activate (NH₃ + CO₂ + H₂O \rightarrow NH₄HCO₃) reaction. This means 3-4 km altitude in the troposphere. But, an altitude of 5-6 km may be necessary to give sufficient time for completion of the reaction before falling down on seas and lands. In the practice, experiments must be started certainly with low NH, concentrations. Although starting with low concentrations, NH, spraying operations must be applied in high altitude of atmosphere in certain places, such as cold and remote regions of the vast Pacific Ocean in south of the World, for safe and harmless application. After NH₃ has been sprayed from an aeroplane, because of transporting and mixing processes by the winds, NH, will spread slowly in the atmosphere but, in a short time, will react with acidic CO₂, NO₂, SO₂, HF, HCI, HBr, H₂S gases and will turn into ammonium compounds. Also by this way, all other acidic pollutants in the air will be cleaned. Application must be started with the small amounts of NH, and must be increased step by step. The conditions of application should always be under control and observation. All applications must be controlled very tightly and constantly by official organizations such as UN, NASA, NOAA etc. Of course, the NH, which would be used in this method, must be produced industrially with hydrogen coming preferably from a non-carbon process, e.g., from water electrolysis. Coal, coke, natural gas, petroleum and CO are cheap natural resources for hydrogen production but, in this case, again CO₂ occurs. But at the beginning, it is not necessary for first experiments in the atmosphere.

CONCLUSION

According to the IPCC statement, the main cause of increasing global warming is represented by human activities. Unfortunately, the world population and energy needs increase consistently, so very urgent solutions are necessary. The NH₃ gas spraying method can be a fast and effective solution, if it is applied

carefully. At least, the current study method will be useful to keep atmospheric CO_2 concentration and global warming under a constant level, which is already the target of the COP 21 agreement. Of course, this will be an expensive application, but its cost must be paid by United Nations, related organizations and the other NGO's to save future of the peerless planet "EARTH".

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CONFLICT OF INTEREST

The author declares that there is no conflict of interests regarding the publication of this manuscript.

ABBREVIATIONS

AR 5	5. Assessment report
°C	The degree Celsius
CaO	Calcium oxide
$Ca(OH)_2$	Calcium hydroxide
CCC	Carbon capture capacity
CCS	Carbon capture and storage
CH_4	Methane
CO_2	Carbon dioxide
COP 21	Twenty- first Conference of the Parties, Paris 2015
ΔG^{o}	Standard free energy of formation
GHGs	Greenhouse gases
g/mol	Gram per mole
Gt	Giga ton
h	Hour
HBr	Hydrogen bromide
HCl	Hydrogen chloride
HF	Hydrogen fluoride
Hg	Mercury
IPCC	Intergovernmental Panel on Climate Change
Κ	Kelvin scale, absolute thermodynamic temperature scale
K_2CO_3	Potassium carbonate
Kg	Kilogram
kJ	Kilojoule

kJ/mol	Kilojoule per mole
km	Kilometre
М	Molarity
MEA	Monoethanolamine
mg	Milligram
MgO	Magnesium oxide
mol m ³/s	Mole x Cubic metre per second
MW	Molecular weight
Na_2CO_3	Sodium carbonate
NaOH	Sodium hydroxide
NASA	National Aeronautics and Space Admin- istration of USA
NF ₃	Nitrogen trifluoride
NGO	Non-Governmental Organization
NH ₃	Ammonia
NH ₄ HCO ₃	Ammonium hydrogen carbonate
N_2O	Dinitrogen monoxide
NO_x	Nitrogen oxides
NOAA	National Oceanic and Atmospheric Administration
R_{R}	Reaction rate
ppb	Parts per billion
ррт	Parts per million
PZ	Piperazine
SF_6	Sulfur hexafluoride
SO_x	Sulfur oxides
UN	United Nations

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