Use of Aqueous Ammonia in Silencer for removal of CO2, SO2 and NOx from exhaust gases of I.C. Engines

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Abstract

This paper consists of experimental research work for implementing aqueous ammonia solution as an absorber for the reduction of CO2, SO2 and NOx from exhaust gases of I.C. Engines. The aqueous ammonia process can simultaneously remove CO2, SO2, NOx and also hydrocarbons that may present in the exhaust gas. There could be oxidation of SO2 and NOx prior to contacting the aqueous ammonia absorbent. A concept pertaining to the ammonia/carbon dioxide reaction in an exhaust system is presented.

Index Terms— ammonia, CO2, SO2, NOX,

1. Introduction

All Reduction of green house effect gases (GHGs) has taken the attention of researchers and scientists around the globe. In recent years, these concerns have risen than ever before. The large amounts of carbon dioxide (CO2) being emitted into the atmosphere could cause severe global climate changes [2]. Recent observations confirm atmospheric that the concentration of CO2 in the atmosphere has increased by nearly 30% for the last 150 years, with an accelerating trend in last year's. In 1997, world community including India accepted Kyoto Protocol. Its importance and possible implementation was emphasized in 2005.

The objective was to address the problem of climate changes occurring due to human activities. Protocols were defined to follow the footwork of UN Framework Convention on Climate Change (UNFCCC) [1]. This is the largest increase observed for any decade in at least the last 200 years. From 1999 to 2005, global CO2 emissions from fossil fuel increased at the rate of roughly 3% by year [9]. Through human activities, mainly due to burning of fossil fuels and by cutting down of forests adds to green house effect and rise sea level. The CO2 concentration

level in the atmosphere should therefore be stabilized or reduced.

Following are the advantages of aqueous ammonia process for CO2 separation from flue gases:

The rate of corrosion due to ammonia is low, i.e. aqueous ammonia does not pose a corrosion problem.

Aqueous ammonia has high loading capacity. Approximately 1.2kg of CO2 can be absorbed per kg of ammonia.

The energy requirement for ammonia regeneration is predicted to be much lower.

The major byproducts from the Aqua Ammonia Process include ammonium sulphate, Ammonium nitrate and Ammonium bicarbonate. Ammonium sulphate and Ammonium nitrate are well known fertilizers for certain crops. Ammonium bicarbonate has been utilized by certain developing countries as a crop fertilizer for over 30 years with proven results in farmland practice which enhanced crop root development and leaf growth [4]. Ammonium bicarbonate was rated by Kirk-Othmer [5] as having definite fertilizing value.

Considering all these advantages of ammonia, a concept is visualized to implement ammonia as a replacement for Platinum and Iridium catalysts in mufflers or silencers. Ammonia absorbs SO2, NOx, and CO2 so as to reduce the atmospheric pollution. The main advantage of this exhaust system is that the whole other systems are unaltered, so this concept can be implemented on existing automobiles too. It is currently envisioned that the aqua ammonia process can be used in automobile exhaust systems to capture all three major acid gases (SO2, NOX, CO2) and also HF, which may be present in the flue gases.

2. Experimental Work

a) Construction:

The designed equipment consists of three cylinders viz. the absorber, the storage cylinder and the filter. The absorber has a provision for continuous flow of aqua ammonia solution from a reservoir into the absorber in the direction opposite to that of the gas flow. A gas spreader is provided at the outlet of the combustion chamber. The spreader is a small porous cup that confirms the dissolution of gases into the aqueous ammonia and avoids formations of big gas bubbles. The spreader is so adjusted that it dips under a constant level of ammonia flowing.

The storage cylinder serves as storage in case of rise in the level of aqueous ammonia due to pressure rise. The filter cylinder consists of gas filters so as to absorb the ammonia vapours formed due to the temperature of the exhaust gases. There is a provision of cooling these vapours and recollecting for reuse (not shown in figure). The remaining unabsorbed gases are then ejected to the atmosphere.

b) Working:

The exhaust gases containing SO2, NOx, and CO2 from the combustion chamber enter the absorber through the spreader. A definite flow rate of aqueous ammonia is capable of absorbing maximum amount of CO2 and the others. The direction of flow of aqueous ammonia is anti-parallel to that of the flue gases. There is continuous flow of aqueous ammonia as shown in the figure.

The function of storage cylinder is to hold aqueous ammonia in case of rise in level due to pressure of the exhaust gases. Further the gases, flowing through the pipe and a pair of filters in the filter cylinder, are ejected to the atmosphere.

The gases that are ejected to the atmosphere are found to contain 10% to 25% less carbon dioxide, carbon monoxide and hydrocarbons.

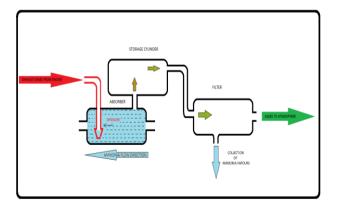


Figure 1: Block Flow Diagram for Aqua Ammonia and Exhaust Gases

The Elementary chemical reactions in the CO2–NH3– H2O system are shown in the following table [3]:

Equation	Process	No.
$CO_2 + NH_3 \longleftrightarrow NH_2COO^- + H^+$	formation of ammonia carbamate	(1)
$CO_2 + OH^- \longleftrightarrow HCO_3^-$	formation of bicarbonate by combination of CO ₂ with hydroxyl ions	(2)
$\text{HCO}_3^- \longleftrightarrow \text{CO}_3^{2-} + \text{H}^+$	formation of carbonate	(3)
$\mathrm{NH}_3 + \mathrm{H}_2\mathrm{O} \longleftrightarrow \mathrm{NH}_4^+ + \mathrm{OH}^-$	hydrolysis of ammonia	(4)
$H_2O \longleftrightarrow OH^- + H^+$	dissociation of water	(5)

Table No.1 Reactions

The chemical reactions that take place in the cylinders are as shown in the table. Further research work will also be done to know the exact concentration of ammonia to absorb the gases.

c) Regeneration:

In the regeneration process aqueous ammonia, the absorption by-products are thermally decomposed to release CO2 from the solution of ammonium compounds. Three potential reactions could be responsible for liberation of CO2 during the thermal regeneration.

2NH4HCO3 (aq) \leftrightarrow (NH4)2CO3 (aq) + CO2 (g) + H2O Δ Hrx = 6.4 kcal/mol(6)

NH4HCO3 (aq) \leftrightarrow NH3 (aq) + H2O + CO2 (g) Δ Hrx = 24.1 kcal/mol(7)

(NH4)2CO3 (aq) \leftrightarrow 2NH3 (aq) + H2O + CO2 (g) Δ Hrx = 15.3 kcal/mol(8)

The heat of reaction for the process has been reported to be 20.0kcal/mol.

From Eq. 6, it is shown that ammonium bicarbonate transforms into ammonium carbonate and CO2. It has the lowest enthalpy of dissociation for CO2 release. Ammonium carbonate can be further dissociated to release more CO2, but the required cost per mole of CO2 is high

d) Discussion of results:

It was seen that the carbon dioxide content of exhaust gases was reduced by an average of 25% as compared to that of the ordinary silencer. Also reduction in the emission of gases such as nitrogen oxides was noted. Along with the above gases, absorption of hydrocarbons from the exhaust gases was seen.

3. Conclusions

Aqua Ammonia with proper concentration can be very useful for reducing the rate of pollution from I.C. engines. Future research work will be modifications in this concept and design for efficient absorption of carbon monoxide, carbon dioxide, sulphur dioxide and nitric oxides in the ammonia solution. Work will also be conducted to determine the mechanism of the reactions involved in the absorption and regeneration reactions.

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