ISSN: 0975-833X

Available online at http://www.journalcra.com

International Journal of Current Research
Vol. 8, Issue, 05, pp.31587-31589, May, 2016

INTERNATIONAL JOURNAL OF CURRENT RESEARCH

RESEARCH ARTICLE

AUTOMOTIVE EXHAUST EMISSION CONTROL USING DOPED PEROVSKITE BASED CATALYSTS (A LITERATURE REVIEW)

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ARTICLE INFO

Article History:
Received 18th February, 2016
Received in revised form
25th March, 2016
Accepted 18th April, 2016
Published online 30th May, 2016

Key words:

Catalyst,
Reaction Engineering.

ABSTRACT

Carbon monoxide, oxides of nitrogen, unburnt hydrocarbons and particulate matter are the principal air quality pollutant emission from automobiles. Number of automobiles are rocketed in recent years all over the world. Carbon monoxide it reduces the blood oxygen-carrying capacity which can reduce the availability of oxygen to key organs. Extreme level of exposure can be fatal. Maximum 8 hours average CO level allowed by WHO is 10mg/m3 Thus, it is very necessary to control exhaust emission from automotive source for environment. Catalytic converters are used for this purpose which converts carbon monoxide into less harmful carbon dioxide. Catalytic converters are not only useful for automobile but also in forklifts , mining equipments, generator sets, locomotives, motor cycles, airplanes etc. Three way oxidation- reduction catalytic converters are used in gasoline-powered vehicle for strict vehicle emission regulations. Research has been carried out and still going on for development of more economical and efficient catalyst. Originally very expensive metals Pt and Rh were used. Perovskites are one of these development but low surface area is its limitation. To overcome this limitation doped perovskite is the solution which reduces 70% decrease in precious metal usage. To develop doped perovskites using different material and different method to make catalyst more economical and efficient will be goal of the research work.

Citation: Shilpa Patel, 2016. "Automotive exhaust emission control using doped Perovskite based catalysts (A literature review)", *International Journal of Current Research*, 8, (05), 31587-31589.

INTRODUCTION

The principal air quality pollutant emission from automobiles (petrol, diesel and alternative fuel engines) are carbon monoxide, oxides of nitrogen, unburnt hydrocarbons and particulate matter. Number of cars sold worldwide from 1990 to 2015(in million units) are 39.2 to 72.41. Number shows that sale of cars has rocketed and so the numbers of the other vehicles like light duty trucks, heavy duty trucks (both diesel and gasoline fueled) motor cycles have also increased with time. Annual pollution emission of carbon monoxide from a passenger car in 2000 in USA was 261 kg. 2/3 of carbon monoxide emission comes from transportation sources, with the largest contribution coming from cars.

Motivation for research work

Effect of carbon monoxide on human being is, it reduces the blood oxygen-carrying capacity which can reduce the availability of oxygen to key organs. Extreme level of exposure can be fatal. Maximum 8 hours average CO level allowed by WHO is 10mg/m^3. Thus, it is very necessary to control exhaust emission from automotive source for environment and to follow regulations. I want to contribute in controlling of such dangerous carbon monoxide from automobile Emission by this research work.

Preliminary studies

To control exhaust emissions two types of systems are used, the air injection system and the exhaust gas recirculation (EGR) system. For chemical engineering point of view, catalytic converters are used. It consists of an insulated chamber containing catalyst. Exhaust gases are passed through the catalyst and converted hydrocarbons, carbon monoxide and nitrogen oxides in water vapor carbon dioxide and nitrogen. This system is not completely effective; during warm up the temperature is so low that emission can not be catalyzed completely. Due to strict regulation it is necessary to develop more efficient exhaust emission control system. The catalytic converter was invented by Eugene Houdry, a French mechanical engineer involved with catalytic oil refining. In 1973, former General Motor President Robert Stempel decided

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to begin implementation of the catalytic converter in automobiles. Since then many scientists have been working on the development of the catalytic converter. Catalytic converter have become mandatory in all cars. Although catalytic converter are primarily used in exhaust system in automobiles, it is also modified and used for trucks, buses, fork lifts, mining equipments, generator sets, locomotives, motor cycles, airplanes etc.

Different variation of catalytic converters

The key type of catalytic converters are listed below:

1. Two way oxidation catalytic converter: Here, Oxidation of carbon monoxide to carbon dioxide and oxidation of hydrocarbon to carbon dioxide and water occurs. It is not able to control oxides of nitrogen.
2. Diesel oxidation catalytic converter: It is used for diesel engines. This device uses oxygen in the exhaust gas stream to convert carbon monoxide to carbon dioxide and hydrocarbon to water and carbon dioxide. Efficiency is 90%.
3. Three way oxidation- reduction catalytic converters are used in gasoline- powered vehicle for strict vehicle emission regulations. It is one of the important inventions in the history of the automobiles. They perform these simultaneous tasks, reduction of nitrogen oxides to nitrogen, oxidation of carbon monoxide to carbon dioxide and oxidation of unburnt hydrocarbon to carbon dioxide and water

Different types of catalyst used in three way catalytic converter; their advantages & limitations

Original three-way catalytic converter used the combination of Pt and Rh typically in a 5:1 ratio. Further development has led to a multitude of formulas including Pt/Pd/Rh, Pd/Rh, Pd only etc. They provide high number of active sites but can agglomerate. In oxide formit has tendency to react with alumina and form solid solution, to minimize it, zirconia is used with the alumina. Current generation automotive catalyst material consists of a ceramic or metallic substance coated by an aluminium oxide (Al_2O_3) based wash coat. The most commonly found converters contain a ceramic substrate (cordierite: $2Al_2O_32SiO_2$, 5MgO) coated with a precious metal containing wash coat. This wash coat contains a combination of platinum group metals (Pt, Pd, Rh) with other rare earth oxides such as CeO_2, ZrO_2 etc. Alumina is used because it is relatively inert and provides high surface but it does not possess the thermal, physical and chemical properties ideal for the perfect activated catalyst layer. Beria, lantana, zirconia, strandhana are added to enhance the thermal characteristics of the alumina. Catalyst upon which active catalyst layer is placed are substrate. They are monolith, honeycomb and unitary body. Both ceramic and metallic forms are used here. Required properties are large amount of geometric surface. This surface does not react with catalyst layer but allow secure attachment of the catalyst layer. Ceramic Substrate: it is made up from silicon dioxide, talc and Kaolin at 1200°C.

Metallic Substrate: They are formed from thin metal sheets of metal alloys containing Fe, Cr and Al stabilized with Y and Ce that have excellent oxidation resistance. Advantage of this over its ceramic counterpart is that either geometric surface can be formed into smaller volume or same volume and geometric surface will provide less pressure drop. It can provide 186 to 248 cells/cm^2. The advantage here is lower exhaust gas pressure drop can result in increased horse power and performance.

Perovskites

The general chemical formula for perovskite compounds is ABX_3 where A&B are two cations of very different sizes, and X is an anion that ends to both. The 'A' atoms are larger than the 'B' atoms. The most common mineral in the earth is bridgmanite, a magnesium rich silicate with adopts the perovskite structure at high pressure. Perovskite like LaCO3 LaMNO3, LaFeO3 prepared by ceramic, citrate complexion, sol-gel co precipitation, microemulsion, spray-drying/freeze drying, flame hydrolysis, reactive grinding.

Problems with perovskites (Limitations)

Low surface area of perovskite is most important limitations. Various synthesis routes have been attempted to improve surface area, yet this was much inferior than the noble metal catalyst

Objective of Research Work

Objective of proposed research work is to develop doped perovskite catalyst for CO oxidation in the exhaust emission.

- Preparation of catalyst by suitable doping material on perovskite.
- Finding out developed catalyst for reducing amount of CO from the exhaust emission.
- Enhancement of activity of selected catalyst
- Characterization of selected catalyst
  Kinetic study of selected catalyst

Finding out most suitable parameters for the working of catalyst.

Doped Perovskite

Doping: Doping means the introduction of impurities (Foreign substance) into the semiconductor crystals. Doping reduces 70% decrease in precious metal usage compared with conventional automotive catalyst.
e.g.

- Strontium doped $LaCoO_3$ perovskite catalyst.
- Cerium doped $LaCoO_3$ perovskite catalyst.
- Cobalt doped LaMnO3 perovskite oxidation
- Platinum doped LaMnO3 perovskite
- Palladium doped perovskite catalyst $BaCe_{1-x}Pd_xO_{3-\delta}$
- Perovskite BaCeO3 material with low level of substitution of Pd(II) on the Ce site and a corresponding number of oxygen vacancies.

These and other kind of doped perovskite catalysts will be studied carefully and selected for research work.

Methods for perovskite doping

(1) Sol-gel method for perovskite doping

Synthesis of perovskite crystals structure phase of strontium doped rare earth manganese using sol-gel method. Polycrystalline perovskite of R0.6 $Sr0.4MnO_3$ nanocomposites (R=La, Nd or Sm) can be synthesized using the sol-gel method in the presence of citric acid and polyethylene glycol as chelating agents respectively. The synthesized gel was calcined at 800°C for 5hrs.

(2) Grinding method for perovskite doping

Pd doped perovskite catalyst

$Bace_{1-x}Pd_xO_{3-}$ (x=0.0,0.05,0.1) were prepared by grinding together BaO_2(99% ceral), CeO_2(99.9% ceral) and PdO(99.95% ceral) in appropriate stoichiometric ratio, pelletizing the ground powder and heating them in flowing oxygen at 1000°C for 10 hours. The pellets were then reground, pelletized and heated for another 10hrs at 1000°C in flowing oxygen, These samples are called 'as- prepared'. Reduction of 'as-prepared' samples was carried out in flowing 5% H_2 in N_2 at 1000°c for 1hr. Reoxidation of the reduced sample was carried out in flowing O_2 at 1000°C for 1hr.

(3) Co-precipitation method for perovskite doping

Nickel doped $LaCoO_3$ perovskite by Co-precipitation method. The $LaCoO_3$, Ni_{1-x} CoO_3 (X=0.1,0.2,0.3) and $LaCo_{1-y}$ Ni_yO_3(y=0.1,0.2,0.3) are prepared by the Co - precipitation method. For this, concentrations solution of Lanthanum, Nickle and Cobalt nitrates are prepared. The concentrations are selected in order to obtain La:Ni:Co atomic ratio of (1-X):X:1 and 1:(1-Y):Y. where, X and Y are the substitution degree to the mixed metal nitrate solution. Excess of ammonium hydroxide (NH_4OH) is added so that metal oxy- hydroxide precipitate are formed. Precipitates are washed with distilled water to PH-7 value filtered and dried in oven for overnight at 110°C .Dried precipitate ground and kept calcinations in muffle furnace at 800°C for 8 hrs.

Characterizations of prepared catalyst

For Characterizations of prepared catalyst following procedure will be done.

1 X-ray diffraction analysis (XRD) pattern provides information of unique fingerprint of crystal.
2 Fourier Transform Infrared Spectroscopy (FT-IR spectra) to examine the chemical bonding.
3 UV v/s DRS study (Diffusion Reflection Spectra) is used to find bond gap of material
4 Scanning Election Microscopy(SEM).

These and other kind of doped perovskite catalysts will be prepared by various methods and studied for the purpose of exhaust emission control.

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