

## Influence of catalytic mass content in catalytic combustion of isopropyl alcohol using aerosol nanocatalysis technology

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*Abstract.* This paper studies the effect of catalytic combustion of isopropyl alcohol according to the principles of aerosol nanocatalysis technology on a vibrating fluidized bed reactor, using a metal oxide catalyst in the form of  $\text{Fe}_2\text{O}_3$ . Catalytic reactions under this technology eliminate the need for catalytic supports, while implementing in situ a continuous mechanical and chemical activation (MCA) of the catalyst surface by the mobile inert material. The effect of catalytic mass concentration in the reactor was analyzed to ascertain the best mass content of catalyst needed to achieve complete high % volume content of  $\text{CO}_2$  in the combustion gases product. This study revealed that under this technology, complete combustion can be achieved at a catalytic mass content of 0.0002 grams and 0.0004 grams, as results of the experiment showed that there needs to be a full saturation of the inert material with the catalytic component, for the combustion reaction to be favorable towards  $\text{CO}_2$  generation. This experiment was conducted by varying the amount of catalytic content being introduced into the reactor, by altering the mass of the catalyst from 0.0001 grams – 0.0005 grams, while using a temperature of 400 °C and MCA frequency of 3 Hz, the MCA frequency of 3Hz provides the reactant the opportunity to fully interact with the pores of the catalytic surface under mild reactor bed vibrations while under the impact of atmospheric pressure.

*Key words:* MCA, combustion, in-situ, vibrating fluidized bed reactor, inert material

### INTRODUCTION

As a result for concerns that have risen due to toxic gas emissions into the environment, combustion engine technology have seen a surge in research, this is due to the pressure by environmental agencies on researchers to find ways to combat the detrimental effects that appear when fuels are not combusted properly [1]. There has been an increase in the enactment of severe regulations that pertain to emissions from industries, countries situated in Europe and United States have been forerunners in this aspect. This has made research in this field imperative, as

it is very important that fuel economy needs to be improved [2]. It is now very common to find out combustor engine research that is focused on eliminating or decreasing toxic and dangerous emissions have been on a steady incline as seen in the Institute of Combustion engines and Control engineering.

Alcohols in recent times have been used as fuels for combustion engines in place of fossil fuels and natural gas, because they are oxygenated compounds. Aliphatic alcohols in the form of methanol, ethanol, propanol and butanol have obtained a lot of attention as forms of fuel due to their ability to be synthesized chemically and biologically, and this gives them the advantage of being able to be used in internal combustion engines. Alcohols have a general chemical formula  $\text{C}_n\text{H}_{2n+1}\text{OH}$ . However, most researches have focused on using ethanol as a result of its extremely high octane number, but it comes with its caveat which is low density, and other negative parameters, that have made it not to be an ideal replacement for diesel fuel in combustion engines [3].

Methanol on the other hand has also been used as fuel, but it poses a serious challenge due to its tendency to have vapor locks in power supply systems at elevated temperatures; the need to develop a complex homogeneous mixing system in cylinders; its difficulty to readily ignite a cold engine and its high wear and tear which in turn reduces engine life [4].

Based on observed literature, there has been little to no research on the use of isopropyl alcohol as a fuel, therefore this paper is focused on understanding the feasibility on the usage of isopropyl alcohol as a fuel for catalytic combustion that proceeds completely to produce  $\text{CO}_2$  and water.

Catalytic combustion has been ushered as a viable option to mitigate and reduce not only  $\text{NO}_x$  emissions that arise from high temperature but other VOC's (Volatile Organic

Compounds) in the form of UHC (unburned hydrocarbons), CO<sub>2</sub>, CO, prompt NO<sub>x</sub>, and thermal NO<sub>x</sub>. These harmful emissions were synonymous with earlier combustion processes called “thermal or flame combustion” in which combustion processes were executed at high temperature [5]. Catalytic combustion involves using a catalyst to speed up the reaction process at a relatively low temperature, so as to obtain better by products and a more efficient economy.

It has been well noted that catalyst used in conventional combustion processes are usually in the form of noble metals: platinum (Pt), palladium (Pd), metal oxides or a mixture of both. The noble metal catalyst are very expensive, prone to sintering at elevated temperatures of 800 °C or above, while also requiring catalytic supports in the form of silica gel, gamma alumina or alumina oxide. The role the support plays is to provide a platform for the catalyst to attach to, in order to provide porosity (micro) and sustainable structural characteristics. However these supports have a prominent drawback which is that they are highly hygroscopic [6]. In addition, most of the catalytic combustors used in the industry employ fluidized bed or adopt a water shirt that strongly cools the catalyst in the combustion zone, as a result there is a surge in emission of CO, NO<sub>x</sub>, when unlocking any of the sections of the shirt, as the temperature increases steeply [7].

The technology of aerosol nanocatalysis is based on a vibrating fluidized bed which is comprised of a reactor operating in a vibrating mode with its reactor bed pseudofluidized in nature. The catalytic component placed in the reactor experiences in situ by exerted mechanical chemical activation. This produces a constant pulverizing of the catalytic particles to nanoscale and this creates a sustained high activity over an indefinite duration of time. [8]. Due to the ability to control the reaction process, by altering the speed of the reaction, chemical transformations can be tailored for each reaction specification and this can be achieved by regulating the frequency Hz of the mechanical chemical activation, so as to attain the desired reaction. Technically this provides the opportunity to increase selectivity for target reactions by influencing the forced interactions at a nanoscale [9]. Researchers in the earlier years were able to study aerosol nanocatalysis and model the process mathematically, this enabled them to understand how to optimize the reaction processes, in order to create a control system that is operated automatically [10, 11]. The quintessential nature of aerosol nanocatalysis is the absence of a support. The catalytic system comprises of glass beads which acts as a dispersing material, this initiates the mechanical chemical activation of the catalyst particle that is contained in the reactor. The catalyst particles are 200 microns initially in size, however under the influence of mechanical chemical activation, during reactions the size is 8–100 nm. The non existence of a catalytic support creates high activity on the surface of the catalyst that is 10<sup>4</sup>–10<sup>6</sup> higher than in catalyst that require supports. Factors which make aerosol nanocatalysis technology advantageous and contribute to its high catalytic activity are [12]:

- Catalytic concentration applied is less than what is applied in the industries, as the concentration is 1 g/m<sup>3</sup> of the reactor volume.

- The small amount of catalyst used; regeneration processes can be periodically excluded or avoided thereby reducing cost.
- There can be the avoidance of the use of steam, since the catalyst does not use supports, thereby reaction products will not clog on to the catalyst.
- In-situ mechanical chemical activation which generate high activity.
- The catalytic component can be applied for longer duration of time in comparison to the catalyst used in industries; i.e. high stability

For this reason the catalytic concentration commonly used in aerosol nanocatalysis technology is predominantly 0.0001 gm, and this was applied in the combustion of isopropyl alcohol and results obtained were encouraging, however it was necessary to study the effect of increasing the catalytic content mass (grams) to ascertain if increasing the catalytic content leads to improved combustion processes (complete combustion). Therefore the catalytic concentration in this research was varied from 0.0001 gm to 0.0005 gm, and experiments were conducted at a constant mechanical chemical activation frequency and temperature to ascertain the best catalytic concentration to achieve complete combustion to CO<sub>2</sub>. Experiments were also conducted on the inert material (without catalyst concentration) which is the glass beads, this was done to create an understanding on how catalyst concentration on the glass beads (inert material) affects combustion processes, this was done on a definite mechanical chemical activation frequency, but also on various temperature ranges. This study will broaden the horizon on the role catalyst concentration plays.

## MATERIALS AND METHODS

The experiment was necessary, as it enables the obtaining of data that indicates the degree of conversion of fuel, which was taken as isopropyl alcohol, as well as the presence of harmful substances in the composition of combustion gases.

The catalyst used in this research is Fe<sub>2</sub>O<sub>3</sub>, which is a metal oxide catalyst and this is a deviation from the norm that is commonly practiced in the industries, this catalyst was chosen, as it is cheaper and easily attainable. Although the use of single metal oxides in catalytic combustion has been researched by [13]. Most metal oxides are prone to sintering at temperatures above 1000 °C and need materials made of wash coat to support them in order to maintain good dispersion. Wash coat materials include silica, alumina, zirconia and all these wash coats are known to be ineffective at temperatures above 1000 °C.

Common metal oxides are currently used as supports and these are iron oxide Fe<sub>2</sub>O<sub>3</sub>, silica oxide SiO<sub>2</sub>, titania TiO<sub>2</sub>, ceria CeO<sub>2</sub> and alumina Al<sub>2</sub>O<sub>3</sub>. Although these oxide's pore surface area are known to vary depending on their method of preparation. For commercial benefits, it is essential that these metal oxides have a high surface area (>100 m<sup>2</sup>/g) and are highly porous, so as to take advantage of the expensive active metals that will be dispersed on them [14].

The essence of these metal oxides is that they can act as Lewis acid sites, thereby affecting the conversion and selectivity process by accepting electrons in pairs. By accepting electron pairs they can invariably affect the conversion and selectivity, and this is how they act as Lewis acid.

Alumina and iron oxide are strong Lewis acids because they have  $Al^{3+}$  and  $Fe^{3+}$  ions in their structure as a result reveal a strong affinity to oxidize substrates that have a preference for increased activity rather than selectivity.

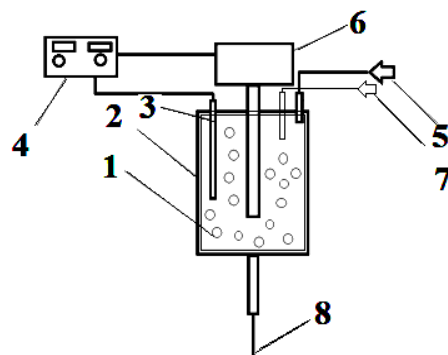
Magnesium Oxide which is a member of alkaline earth metal can also be used as a support and this was shown to be compatible with platinum, as platinum had an inclination to form small metals [15]. These metal oxides apart from providing support for catalysts, also provide the lattice oxygen that enables combustion while gaseous oxygen from the air introduced via the combustor replenishes the vacancy created in the absence of basic conditions [14]. Lewis acid sites and lattice oxygen are the main components when it comes to combustion of alcohol and this elucidates the difference in activity among various metal oxides [16].

However since the technology of aerosol nanocatalysis offers the opportunity to carry out reactions without the presence of supports, metal oxide catalyst ( $Fe_2O_3$ ) was chosen.

The reactor is made of heat-resistant material, capable of withstanding vibrational oscillations, as well as temperature fluctuations of 400 °C or higher. Also, the reactor has a grid inside which does not allow the catalyst to fall out of the reaction zone; a metal pocket for thermocouple to control temperature fluctuation; the thermocouple is insulated. All tubes for supply of raw materials and combustion products are tight. Rotameter and dispenser are calibrated. The laboratory installation is designed to conduct combustion reactions. The experiment is conducted by introducing isopropyl alcohol via (7) to the reactor (1), while air is also introduced to (1) simultaneously via the aid of (5), the electric heater (2) provides the heat and high temperature for the reaction to take place, this high temperature is measured via the thermocouple (3) that is attached to the reactor (1). The combustion gases are collected via (8) through the help of rubber pipes that are connected to plastic bottles that contain water which enables the trapping of the gases.

But before the procession of the above experiment, the catalytic component should be pre-adsorbed onto the surface of the dispersing material. The procedure is carried out as follows, for the experiment a certain amount of dispersing material (in laboratory experiments, a glass bead in the dimension range of diameter 1.0–1.2 mm) which occupies 50 % of the reactor volume. The dispersing material is loaded into a glass flask in an amount that is more than necessary. Catalyst powder was weighed on an analytical balance to the nearest 0.0001 gm in an amount that was excess, and this exceeds the adsorption capacity of the dispersing material. This excess catalyst powder is introduced into the glass flask. The flask was covered and shook vigorously at room temperature for 10–15 mins. The catalyst and the dispersing material are loaded into the vibrating reactor

(1), after the shaking process, the reactor is operated at a vibration frequency using (6) to enable the total dispersion and heating of the catalyst on the dispersing material via the vibration of the reactor. Subsequently the catalytic system (catalyst + dispersing material) is unloaded into a flask and later into a sieve in order to separate the catalyst which was not adsorbed. The separated catalyst is weighed on an analytical balance. If there is a differentiation in mass between the initial amount of catalytic system before introduction into the reactor and after unloading from the reactor, a new measured catalyst is added to the catalytic system and then shook vigorously and later re-loaded into the reactor, which is allowed to vibrate on a non-definite MCA (mechanical chemical activation) frequency for 1 hour. This procedure is repeated until there is no change in mass from the unloaded catalytic component (dispersing material and catalyst) after heating in the reactor, in comparison with the previously weighed catalytic system (dispersing material and catalyst) after shaking in a flask.



1 – reactor with inert material and catalyst inside, 2 – electric heater, 3 – thermocouple pocket, 4 – control unit, 5 – line of air supply using a compressor, 6 – magnetic vibration device, 7 – line of supply of isopropyl alcohol using a dispenser, 8 – line for the removal of gases of oxidation

**Fig. 1.** Schematic representation of the laboratory setup

Owing to the goal of this research, the catalytic mass content was varied in mass from 0.0001 gm, 0.0002 gm, 0.0003 gm, 0.0004 gm and 0.0005 gm. Each time the catalytic content was increased, the whole above procedure was repeated; following that, is the introduction of the various masses of catalytic content in the form of 0.0002 gm, 0.0003 gm, 0.0004 gm and 0.0005 gm periodically (after satisfactorily conducting experiments on each individual catalytic mass content).

## METHOD OF DETERMINING PRODUCTS

### Determination of products from combustion reactions

The determination of concentration of hydrogen, oxygen, CO, CO<sub>2</sub>, and methane in gas products from laboratory and pilot plants for the disposal of industrial waste, was done by LHM-8 (Laboratory Chromatography 8).

Measuring range of concentrations: 1) Hydrogen – 0.01–7 % Volume; 2) Oxygen – 0.05–20.95 % Volume; 3) CO – 0.02–25 % Volume; 4) CO<sub>2</sub> – 0.01–10 % Volume; Methane – 0.01–30 % Volume.

Relative total measurement error for each component does not exceed 15 %.

Measuring component of H<sub>2</sub>, O<sub>2</sub>, CO, CO<sub>2</sub> and methane in the gas products is founded on direct chromatography by the determination on a chromatograph with a thermal conductivity detector. Separation is conducted on a chemical modification of active coal (SKT fraction) 0.25–0.5 mm.

During the performance of measurement in laboratory there should be the observance of the following conditions: 1) Temperature of air 20±5 °C; 2) Moisture of air Not more than 80 % during temperature 25 °C; 3) Line voltage 220±10 V.

Chromatography parameters: Temperature of thermostat 130 °C; Temperature of column 150 °C; Bridge 100 current; Output of gas-in relation with N<sub>2</sub>, 30 cm<sup>3</sup>/min; Volume of sample 1–2 cm<sup>3</sup>; Speed of chart 600 mm/hr; Ratio of signal/noise, not less 10:1.

For calibration of device use standard artificial mixture, that is fed to the chromatograph with the aid of gas tap dispatcher.

Calibration coefficient for H<sub>2</sub>, O<sub>2</sub>, CO is calculated by the formula

$$K_1 = \frac{C}{h \times m}.$$

Calibration coefficient for methane and CO<sub>2</sub> K<sub>2</sub> is calculated by

$$K_2 = \frac{C}{h \times b \times m},$$

where *C* is a concentration of substance in artificial mixture, mg/dm<sup>3</sup>; *h* is a height of chromatograph peak, mm; *b* is a width of chromatograph peak on medium height, mm; *m* is a scale factor

Calibration coefficient is determined for each analyzed substance in redistribution required for significant concentration.

Calibration of device is conducted at least 1 time in 2 months, in redistribution required for significant concentration.

#### PROCESSING OF MEASUREMENT RESULTS

Concentration of H<sub>2</sub>, O<sub>2</sub>, CO is calculated by the formula

$$C_1 = h \times b \times m \times K_1.$$

Concentration of methane and CO<sub>2</sub> is calculated by the formula

$$C_2 = h \times b \times m \times K_2,$$

where *h* – height of the peak, mm; *b* – Width of the peak not medium height, mm; *m* – Scale factor; K<sub>1,2</sub> – Coefficient of analyzed substance according to claim 9.4. [17, 18].

#### CONTROL OF ACCURACY OF RESULTS OF MEASUREMENT

Control of convergence of the output signals of the chromatograph, control of the parameters is always in relationship with the output signals of chromatograph during three input samples. Control is carried out during calibration; during periodic control calibration coefficient, and also during measurement.

Result control recognized as positive when condition is met.

$$K_1 = \frac{S_{\max} - S_{\min}}{S} \times 100 \leq 12,$$

where *S*<sub>max</sub> is a maximum square chromatograph peak, mm<sup>2</sup>; *S*<sub>min</sub> is a minimum square chromatograph peak, mm<sup>2</sup>; *S* is an average arithmetic value square peak, mm<sup>2</sup>.

If obtained value is > 12 %, then it is necessary to check parameter analysis correctly, good condition of chromatograph.

$$K_1 = \frac{H_{\max} - H_{\min}}{H} \times 100 \leq 12,$$

where *H*<sub>max</sub> is a maximum height chromatography peak, mm; *H*<sub>min</sub> is a minimum height chromatography peak, mm; *H* is an average arithmetic value height peak, mm.

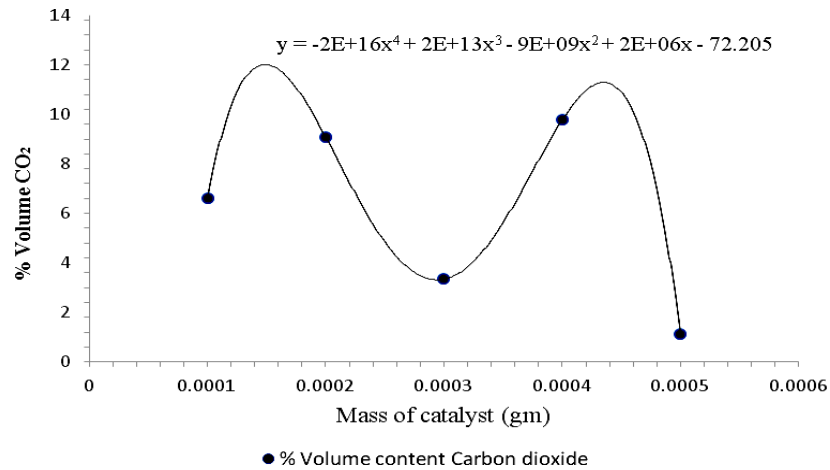
If obtained value >12 %, it is necessary to check good condition of analysis parameter.

#### RESULTS AND DISCUSSION

The table 1 reveals data as obtained from chromatography results, shows the % changes in the amount of gases obtained from combustion of isopropyl alcohol based on changes in mass of catalyst used in the reaction process. The temperature (°C) and frequency (Hz) were held constant, as this enables us to understand the importance of alterations in catalyst mass. Several experiments were carried out, and the average results were compiled in order to interpolate the data used in constructing the graph below in Fig. 2. It can be concluded that with a mass of free catalyst at 0.0002 gm and 0.0004 gm, the highest content of CO<sub>2</sub> in the oxidation products is observed. This shows that although catalytic combustion reactions using technology of aerosol nanocatalysis is possible using a catalytic mass of 0.0001gm, better combustion reactions that lead to more production of CO<sub>2</sub>, will require the catalytic component be increased, so as to maximize this reaction.

The Table 2 shows that although combustion reactions took place, the % volume of CO<sub>2</sub> gas contained was relatively minute in comparison with the inert material combined with catalyst, the frequency of vibration was kept constant, while the temperature was varied.

From the above results, it can be inferred that combustion reactions will always take place, when there is high temperature, and significant vibration of the vibrofluidized reactor bed containing the inert materials under the technology of aerosol of nanocatalysis, but the combustion reactions leads to infinitesimal % volume of CO<sub>2</sub> produced, thus for combustion reactions to be maximized, it is imperative to add a catalytic content to the inert material, that way the efficiency of the reaction can be attained. However, at a high temperature of 520 °C, the highest % volume of CO<sub>2</sub> was seen, while the lowest was observed at 400 °C. This result substantiates the theory of combustion reactions under “flame technology” where combustion reactions were only possible at very high temperatures in the range of 800 °C – 1000 °C, but with the technology of aerosol nanocatalysis, such temperature range is significantly reduced as seen in Fig. 3.



**Fig. 2.** Changes in the %composition of CO2 gas from changes in the amount of catalyst in the reaction zone

**Table 1.** Data of studies with variation in volume of catalyst

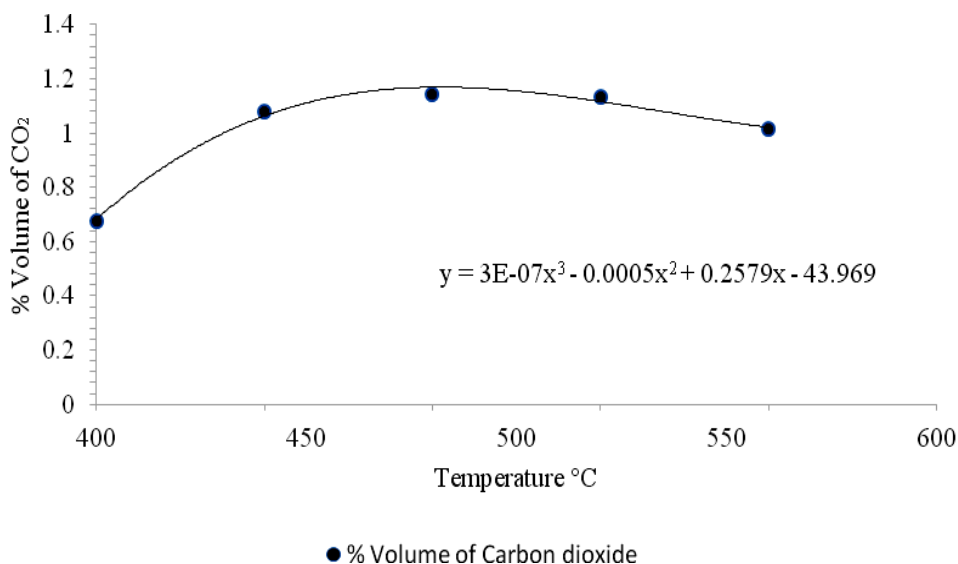
Nos of experiments	Gm	Hz	°C	Time of sampling from the beginning of the experiment (Seconds)	CO <sub>2</sub>	CH <sub>4</sub>	H <sub>2</sub>	O <sub>2</sub>	CO
1	0.0001	3	400	10	7.52	0.03	0.1	10.05	-
2	0.0001	3	400	20	6.99	0.03	0.12	11.84	-
3	0.0001	3	400	30	6.21	0.015	0.07	14.37	-
4	0.0001	3	400	40	5.68	0.015	0.09	15.37	-
5	0.0002	3	400	10	8.54	0.03	0.3	5.88	-
6	0.0002	3	400	20	9.32	0.06	0.41	5.46	-
7	0.0002	3	400	30	9.39	0.06	0.41	3.57	-
8	0.0002	3	400	40	9.1	0.045	0.38	5.3	-
9	0.0003	3	400	10	4.2	-	0.02	16.37	-
10	0.0003	3	400	20	3.18	-	-	18.69	-
11	0.0003	3	400	30	3.03	-	-	18.95	-
12	0.0003	3	400	40	2.96	-	-	19.27	-
13	0.0004	3	400	10	9.88	0.045	0.26	4.2	-
14	0.0004	3	400	20	9.39	0.06	0.37	5.88	-
15	0.0004	3	400	30	9.88	0.06	0.35	4.09	-
16	0.0004	3	400	40	9.95	0.06	0.31	3.52	-
17	0.0005	3	400	10	1.2	-	-	20.69	-
18	0.0005	3	400	20	1.27	-	-	20.49	-
19	0.0005	3	400	30	0.99	-	-	20.99	-
20	0.0005	3	400	40	1.02	-	-	20.79	-

**Table 2.** Data of combustion reactions in the presence of inert material (glass beads) in the reactor

Number of experiments	Mass of catalyst (gm)	F (Hz)	Temp (°C)	Time of sampling from the beginning of the experiment (Seconds)	CO <sub>2</sub>	H <sub>2</sub>	O <sub>2</sub>	CO
1	2	3	4	5	6	7	8	9
21		3	400	10	0.75	21.53	2.41	
22		3	400	20	0.69	21.58	2.42	
23		3	400	30	0.59	21.47	2.43	
24		3	440	10	1.01	21.29	2.51	
25		3	440	20	1.34	21.58	2.52	

Cont. table

1	2	3	4	5	6	7	8	9
26		3	440	30	0.89	21.48	2.53	
27		3	480	10	1.19	21.58	2.61	
28		3	480	20	1.08	21.37	2.62	
29		3	480	30	1.16	21.69	2.63	
30		3	520	10	1.23	21.67	2.71	
31		3	520	20	0.97	21.51	2.72	



**Fig. 3.** Change in the % composition of CO<sub>2</sub> contained in the experiment in the presence of inert material in the reactor

### CONCLUSIONS

1. Metal oxides in the form of Fe<sub>2</sub>O<sub>3</sub> can be used in catalytic combustion reactions under the technology of aerosol nanocatalysis, this is a deviation from the normal practices currently used in the industries, which apply noble metal catalyst in the form of platinum (Pt), palladium (Pd) and Rhodium (Rd), the use of metal oxide as catalyst helps save cost of reactions.
2. The volume of catalyst applied in combustion reactions under the technology of aerosol nanocatalysis, is relatively small in comparison with conventional catalytic combustion reactions, under this technology, catalytic combustion can take place at relatively small amounts in the range of 0.0001 gm – 0.0005 gm, this is possible due to the exclusion of catalytic supports, as a result the reactant and air have equal access to the surface of the catalyst, and thus reaction proceeds without any challenges.
3. In as much as, the volume of catalyst in the reaction zone does not need to be large, combustion under this technology, has shown that the amount of catalyst in the reaction zone has to be increased from the standard 0.0001 gram, that is usually applied in this technology, as shown in the results above in Fig. 2, whereby the best catalytic combustion reactions were achieved at a catalytic concentration of 0.0002 gram and 0.0004 gram.

This further highlights the importance of catalytic saturation on the surface of the inert material, as when there is higher saturation of the catalyst, the % volume of CO<sub>2</sub> increases in the combustion gases product. This further backs the theory behind aerosol nanocatalysis; which is the presence of an exclusion of intra-diffusion stages of catalysis thereby an increase of activity of catalytic dust (when applied to the inert material).

4. Technology under AnC using a vibrating fluidized bed experiences continuous regeneration, as a result this leads to a high efficiency of processes which are guided according to the above principles and specific properties of nanoparticles.
5. Combustion under aerosol nanocatalysis technology targets combustion by lowering the oxygen requirements for the reactant to undergo a chemical transformation. Lower air demands, provides an environment for complete combustion to occur and this produces more energy, by creating a domain for better burning of the alcohol or hydrocarbon.

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