

EMISSIONS FROM GAS BURNER, THEIR IMPACT ON THE ENVIRONMENT AND ABATEMENT TECHNIQUES

Mohammad Nazri Mohd. Jaafar
Department of Automotive and Aeronautics
Faculty of Mechanical Engineering
Universiti Teknologi Malaysia

ABSTRACT

The growing concern over increasing regional and local emissions and concentrations of nitrogen oxides has led to action to reduce emissions by the introduction of legislation in many countries. Recently, there have been significant developments in nitrogen oxides control legislation, both nationally and internationally. All of these legislation and regulation limits on NO_x emissions has led to the development of low NO_x combustors especially for new plants. Basically there are two methods of reducing NO_x . The first involved modifications to the combustion processes to prevent the formation of NO_x . The second method is to treat the combustion gas chemically, after the flame, to reduce NO_x to N_2 .

1.0 INTRODUCTION

The use of fossil fuels is a major source of air pollution, in particular, emissions of oxides of nitrogen, oxides of carbon, volatile organic compounds (VOCs) and also

oxides of sulphur. The combustion of fossil fuels is a major contribution to four pressing environmental concerns, namely, global warming, acid rain, photochemical smog and ozone depletion.

This concern has prompted many governing bodies to legislate new regulations regarding emissions from combustion processes in order to abate the growing environmental problems. United States of America and Japan were among the first to issue legislation regarding emission's level from combustion processes either from stationary or mobile sources. The first federal legislation enacted was the Air Pollution Control Act of 1955 (Public Law 84-159, July 14, 1955) [2]. Thereon, many more amendments and acts were amended and enacted, respectively, and led to the Clean Air Act of 1963. The Air Quality Act of 1967 has initiated a two-year study on the concept of national emission standards from stationary sources that served as the basis for the 1970 legislative act. The concern then spread to other countries such as Europe and United Kingdom. The United Kingdom now has the Environmental Protection Act 1990. It is in the hope that these environmental problems will be reduced by the actions.

2.0 ATMOSPHERIC POLLUTION

2.1 Acid Rain

Rain falling through a perfectly unpolluted atmosphere will arrive at the earth with a pH value of about 5.6. This value of pH is achieved due to reactions of the rain drops with carbon dioxide in the atmosphere since pure water has a pH of 7.0. Generally, any rain with a pH of less than 5.6 is considered acidic.

Acid rain has several damaging effects on ecological system especially mountain lakes and stream. Lake acidification tends to reduce the population of its inhabitant especially fish and this is apparent when the pH of about 4.5 or less is reached since this causes the decimation or malformation of fish. Many lakes

around the world are losing their fish population due to lake acidification and some of these lakes have a pH as low as 2.4, which is equivalent to that of a vinegar. These problems have caused losses of millions of dollars each year to the country involved and this has become a major problem. Acid rain also causes damage to plants and forests, has an adverse effect on human health, and reduces visibility.

The main sources of acid rain are oxides of sulphur and nitrogen, predominantly from combustion processes. Over a period of time after those gases are emitted, usually hours or days, they are carried along by wind currents and combine with water molecules in the water vapour of the atmosphere to form tiny drops, mainly of nitric acid, HNO_3 , and sulphuric acid, H_2SO_4 . These aerosolised acids are then returned to the earth when they encounter rain producing clouds, hence as acid rain.

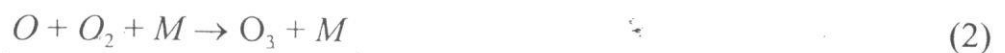
In general SO_2 contributes about 60 percent of the acidity of such precipitation, whereas NO_x contributes about 35 percent [14]. However, the SO_2 emission can be reduced by desulphurisation processes that include desulphurisation of fuel, desulphurisation of waste gas, use of sulphur absorbing additives, and new combustion systems with coal gasification/gas desulphurisation, or with sulphur retention in the combustion chamber. Thus, the NO_x contribution tends to rise [21].

2.2 Ozone Depletion

The atmospheric ozone layer is a layer of relatively high concentration of ozone (O_3) located in the stratosphere (at the height of about 10 to 30 miles). This layer is very important to the society and the environment for a number of reasons. Firstly, it has the ability to absorb ultra-violet (UV) radiation from the sun. A direct UV ray (unblocked by ozone layer) can kill plants and animal cells. It can also cause skin cancer for the human. Secondly, when the ozone absorbs ultra-violet radiation it heats the atmosphere in the vicinity; a change in the abundance of ozone may change the temperature structure of the atmosphere and hence alter the atmospheric

circulation. Thirdly, ozone in the troposphere is a greenhouse gas. It absorbs the infra-red radiation and will trap heat hence cause the earth to become warmer by the greenhouse effect since the surface temperature of the earth is the result of a balance between incoming solar radiation and outgoing infra-red radiation emitted by the earth.

Ozone is produced directly in the stratosphere by the action of ultra-violet radiation ($h\nu$) with a wavelength of less than 243 nm on oxygen molecules (O_2) (UK Stratospheric Ozone Review Group, 1987), which then photodissociates to form free oxygen atoms (O). These then react with further oxygen molecules to form ozone:



where M is any other molecules.

Ozone itself is photodissociated by both UV and visible light:



The production of ozone (processes 1 and 2) can be balanced against the reactions that destroy ozone. Until 1950s, chemical loss of odd oxygen (i.e. O and O_3) was attributed only to the reaction given below:



which is originally proposed by Chapman in 1930. However, it is now realised that ozone in the stratosphere is removed predominantly by catalytic chain reactions (i.e. reactions in which the molecule initiating the reaction sequence is regenerated and is therefore able to initiate further reactions) involving molecules. These catalytic chain reactions are the reactive chlorine (ClO_x) cycle, the reactive nitrogen (NO_x) cycle and

the reactive hydrogen or hydroxyl (HO_x) cycle.

The formation and destruction of ozone in the atmosphere should always be in equilibrium to preserve the ozone layer that is protecting the earth. However, the increased in the concentration of the human-made gases such as oxide of nitrogen from combustion processes and from agricultural activities has led to an imbalance in this equilibrium by removing ozone at a faster rate than it is being produced. The first clear sign of O_3 depletion, popularly known as the "ozone hole" was reported in 1984 over the Antarctic [10]. In 1987 when the "hole" was very severe the total amount of O_3 recorded at Halley Bay in the Antarctic was half of 1970 levels. Similar losses have been recorded each year since then. Total O_3 , measured annually, has fallen by 5 percent since 1979 over the Antarctic.

2.3 Global Warming

Global warming or sometimes referred to as the greenhouse effect is a phenomenon described by a rise in the earth atmospheric temperature. This rise in temperature is caused by the imbalance of the energy of the earth. The sun is the only significant source of energy reaching the earth. The solar energy is delivered by means of ultraviolet, visible and infrared radiation having a wavelength of about 0.2 - 4 μm with a peak at 0.6 μm . Of that incident upon the earth, a portion of the energy is reflected back to the outer space (about 30%) without absorption and does not cause the earth heating [15]. The ultraviolet radiation penetrates the earth's atmosphere and is reflected back as infra-red radiation. Greenhouse gases play an important role in the absorption of these radiations. Greenhouse gases are carbon dioxide (CO_2), methane (CH_4), ozone (O_3), nitrous oxide (N_2O) and chlorofluro-carbons (CFCs).

Gases present in the atmosphere, especially the greenhouse gases, absorb and then re-emit most of the thermal radiation from the earth's surface. However, the atmosphere emits less energy into space than the earth's surface would because the atmosphere's average temperature is lower, roughly -18°C . Thus, the difference

between the energy emitted by the earth and that emitted by the atmosphere is absorbed by the atmosphere. This energy-trapping process is called the greenhouse effect; here the atmosphere plays the same role as the glass pane in a greenhouse. The greenhouse effect is vital for mankind's survival; it keeps the global temperature at a habitable 15°C rather than a hostile -18°C [16]. However, when energy trapping is enhanced, for example, as resulting from an increased concentration in atmospheric carbon dioxide causes more of the thermal radiation emitted from the earth's surface to be absorbed in the atmosphere. This will increase the global temperature higher than needed and causing global climate change. The effect of the doubling in carbon dioxide concentration will raise the global average surface temperature by 2 ± 1 K [5].

2.4 Photochemical Smog

Photochemical smog is a condition that happened when there are air stagnation, abundant sunlight and high concentrations of hydrocarbon and oxides of nitrogen in the atmosphere; and this is usually the conditions of most major metropolitan cities of the world. That is why photochemical smog was first observed in Los Angeles in the mid-1940s. Ever since then, this phenomenon has been detected in other major cities throughout the world.

Smog arises from photochemical reactions in the lower atmosphere by the interaction of hydrocarbons and nitrogen oxide released by exhausts of motor vehicle and some stationary sources. This interaction results in a series of more complex reactions producing secondary pollutants such as ozone, aldehydes, ketones and peroxyacyl nitrates. These reaction mechanisms are too complex and are not fully understood.

The starting mechanism is the absorption of ultraviolet light from the sun by NO_x . This causes nitrogen dioxide to decompose into nitric oxide and highly reactive atomic oxygen [25]:



The atomic oxygen initiates oxidising processes or quickly combines with molecular oxygen to form ozone, which itself is reactive and acts as an oxidant:



In the absence of other species, a steady state is achieved in which the ozone concentration is given by the so-called photostationary state relation [27]:

$$[O_3] = \frac{k_1[NO_2]}{k_3[NO]} \quad (8)$$

Under normal conditions, the ozone formed will be quickly removed by reaction with NO to provide NO_x and O_2 as shown by equation (7). However, in the present of hydrocarbons in the atmosphere this mechanism is partially eliminated as NO reacts with the hydrocarbon radical peroxyacyl ($RCO_3\bullet$) and this resulted in the builds up of ozone concentrations to dangerous levels.

3.0 COMBUSTOR EXHAUST EMISSIONS

Generally there are four major pollutants from any combustion processes. These are oxides of nitrogen (NO_x), carbon monoxide (CO), unburned hydrocarbon (UHC) and

smoke. Oxides of sulphur (SO_x) are present, however, if the fuel used during the combustion processes contained sulphur. Since in the present work only natural gas was used as primary fuel, emissions of oxides of sulphur will not be discussed here. Also, since natural gas has a very high content of methane, the tendency of soot formation is negligible even under fuel-rich conditions. Therefore, only oxides of nitrogen, carbon monoxide and unburned hydrocarbon, their adverse effects, mechanisms governing their formation, and their abatement techniques will be discussed. Oxides of nitrogen are formed during high temperature burner operations with near stoichiometric combustion.

3.1 Oxides of Nitrogen

Oxides of nitrogen normally termed NO_x are produced mainly from high temperature combustion processes since nitrogen is the main composition of air. They are also produced from other natural processes such as bacterial action in the soil, lightning and volcanic eruptions [29]. Nitric oxide (NO) and nitrogen dioxide (NO_2) are considered the most important oxides of nitrogen due to their adverse effect towards animals and plants. Other oxides of nitrogen are nitrous oxide (N_2O), dinitrogen trioxide (N_2O_3), dinitrogen tetroxide (N_2O_4), dinitrogen pentoxide (N_2O_5), and nitric acid (HNO_3). Among these other oxides of nitrogen, nitrous oxide is considered as becoming a nuisance since it may be a significant contributor to global warming and to the possible destruction of the ozone layer and will be considered in the discussion.

3.1.1 Nitric Oxide (NO)

Nitric oxide (NO) is a colourless gas, deep blue when liquid and a bluish white snow when solid. NO is the direct precursor of NO_2 . NO is paramagnetic and contains an odd number of electrons [7].

Nitric oxide has some harmful effects on health but these effects are substantially less than those of an equivalent amount of NO_2 . Pure NO does not

irritate the lung, but forms methemoglobine when resorbed (via the respiration tract). *NO* is an endogenous modulator of the vascular pressure and is considered a “known substance” from the physiological-metabolic point of view [29].

Nitric oxide can be formed by three different mechanisms [21]:

1. Thermal *NO* is produced by oxidation of atmospheric nitrogen in the postflame gases.
2. Prompt *NO* is produced by high-speed reactions at the flame front.
3. Fuel *NO* is produced by oxidation of nitrogen contained in the fuel.

3.1.2 Nitrogen Dioxide

Nitrogen dioxide (NO_2) is a reddish brown gas with an irritating odour. Nitrogen dioxide has been known to have an adverse effect on the human respiratory system. It has an affinity for hemoglobin, which carries oxygen to body tissues. It thus deprives them of oxygen. It also form acid in the lungs and hence is much more toxic than *CO* for the same concentrations. Nitrogen dioxide also absorbs sunlight and initiates a series of photochemical reactions to form smog thus reduces visibility.

Nitrogen dioxide has an olfactory threshold as low as 0.1 ppm [4]. High concentrations, in the range of 1ppm, cause a whole range of reversible and irreversible effects arising from structural changes in cells of the respiratory system. There is also indication that, even at 0.5 ppm, the risk of bacterial infection of the lung increases. Epidemiological studies of exposure to the nitrogen dioxide released during cooking with gas indicate an increase frequency of respiratory illness among children [32].

3.1.3 Nitrous Oxide

Nitrous oxide (N_2O) has recently become a subject of intense research and debate

because of its increasing concentrations in the atmosphere and its known ability to deplete the ozone layer and also to contribute to the greenhouse effect. Nitrous oxide is a colourless, odourless nontoxic gas present in the natural atmosphere in relatively large concentrations (0.25 ppm) [25]. It is also known to every young student of chemistry as 'laughing gas'.

Nitrous oxide is formed mainly from biological activity of the soil and there are no significant anthropogenic sources. It has a low reactivity in the lower atmosphere and is generally not considered an air pollutant, thus not a potential health problem, nor does having a damaging effect on vegetation. The sources of nitrous oxide are given in Table 1.

It is evident from Table 1 that only 20% of the nitrous oxide produced is by fossil fuel combustion.

Table 1 The Relative Importance of N_2O Sources [30]

Source of Nitrous Oxide	Percentage of total emissions
Ocean/freshwater	14
Fossil fuel combustion	20
Natural land emissions	42
Increase in cultivated land	3
Biomass burning	10
Fertilizer	11

3.2 Carbon Monoxide Emissions

Carbon monoxide is colourless, odourless and tasteless. However, it has a strong affinity towards the hemoglobin of the bloodstream and is a dangerous asphyxiant. Carbon monoxide, when absorbed by the lung, reacts with proteins in the blood (primarily haemoglobin) to produce carboxyhaemoglobin ($COHb$). The amount of $COHb$ in the blood is dependent on the concentration of the gas, level of bodily

activity and the exposure time. *COHb* lowers the oxygen-carrying capacity of the blood and has the potential to starve the brain and other tissues of oxygen. Low to moderate concentrations of *COHb* impair thinking and perception, cause headaches, slow reflexes, reduce manual dexterity, decrease exercise capacity and cause drowsiness [13]. At high concentrations, death can result, as in the case of people committing suicide by inhaling vehicle exhaust fumes.

Carbon monoxide is a product of incomplete combustion of fossil fuels and the atmospheric oxidation of methane and other biogenic hydrocarbons. In fossil fuel combustion, carbon monoxide is formed when oxygen is scarce and fuel does not burn properly.

3.3 Unburned Hydrocarbon Emissions

Unburned hydrocarbon is also a product of incomplete combustion. It includes fuel that emerges at the combustor exit in the form of droplets or vapour, as well as the products of thermal degradation of the parent fuel into species of lower molecular weight such as methane and acetylene. Unburned hydrocarbon is usually formed due to poor atomization (for liquid fuel), inadequate burning rates, the chilling effects of burner walls air, or any combination of these.

Some hydrocarbon species are not considered to provide a serious health hazard, as these organic pollutants are almost entirely inert from the physiological point of view. Other hydrocarbons, such as the polynuclear organic compounds, have been shown to cause cancer when deposition is above certain threshold concentrations. Unburned hydrocarbons also act as a precursor to the formation of photochemical smog as has been discussed in the previous section.

4.0 DEVELOPMENT OF LOW NO_x BURNERS FOR DOMESTIC GAS APPLIANCES

Today natural gas is widely used for domestic and commercial heating systems. Although emissions of CO , SO_2 and unburned hydrocarbons are very low, the significant pollutant produced is NO_x . Typical NO_x emissions are in the order of 100ppm per combustion unit, and their combined effect results in a significant annual ground-level emission [14]. The continued trend in regulations will require lower NO_x emission levels than at present.

To meet increasingly stringent legislation, a variety of low- NO_x techniques can be employed. Most of these techniques minimise the production of thermal- NO_x , and in the main they are based on producing low flame or post-flame temperatures, or low residence times, thus leaving the formation of prompt- NO as the major source of NO_x [14].

Basically there are two methods of reducing NO_x . The first involved modifications to the combustion processes to prevent the formation of NO_x . The second method is to treat the combustion gas chemically, after the flame, to convert the NO_x to N_2 [3,9].

The first approach has been widely applied. The main aim is to reduce the flame temperature thus reducing the formation of NO_x . There are several ways of accomplishing this.

Water or steam injection has been shown to be a very effective technique to accomplish the above goal [28]. A typical NO_x reduction curve as a function of rate of water injection is shown in Figure 2 [8]. These data were obtained in an aeroderivative inductive gas turbine at full power. The same effect was also demonstrated by Fox and Schlein (1992) testing the FT8 gas turbine combustor in their final test run. The FT8 engine is an industrial/marine gas turbine engine which is a derivative of the widely used JT8D aircraft jet engine. However, to avoid detrimental effects on turbine durability the water has to be purified to a maximum of 2-5 ppm of dissolved solids [8,26]. Furthermore, there are other complications such as incorporating the water injection system to the combustor design. Another

disadvantage of water injection is the undesirable side effects of quenching CO burnout. These drawbacks caused water injection method to be unattractive for smaller gas turbines or where availability of sizeable water supply is difficult. However, it is a feasible technique for burner NO_x control in water heater or steam generator.

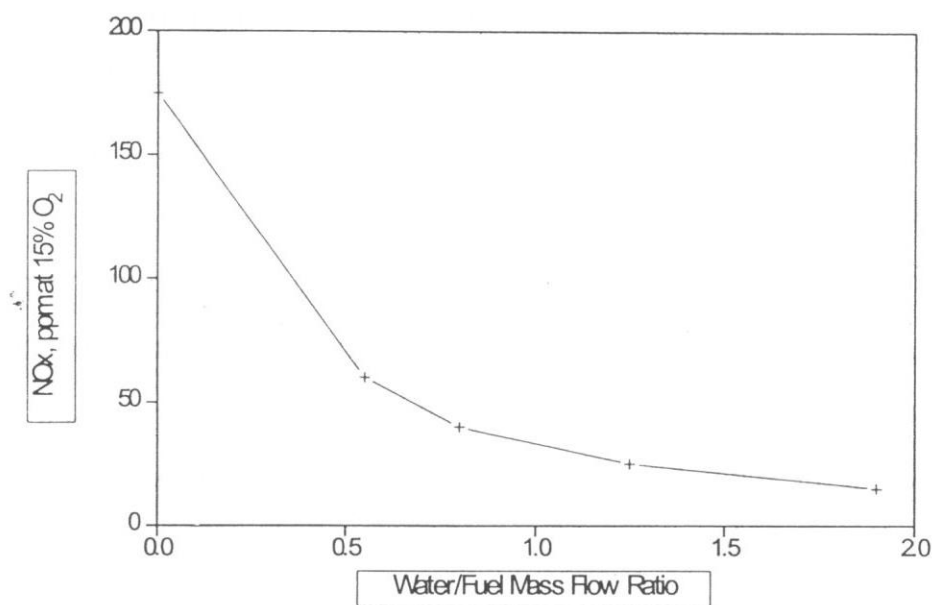


Fig.2 NO_x As A Function Of Water Addition In A Gas-Turbine Combustor Running On Natural Gas At A Pressure Ratio Of 30 (After Correa, 1991).

Another method of modifying the combustion processes is to either burn fuel-rich or fuel-lean. The operation of lean burning is to introduce additional air in order to reduce the flame temperature. This would generally cause a significant decrease in the production of NO_x , however, a reduction of the primary-zone flame temperature may increase the emissions of carbon monoxide and unburned hydrocarbon. On the other hand, the problem with fuel-rich combustion is the formation of soot and CO even though the stability margin is widen.

Other methods of NO_x control involve staged combustion, variable geometry

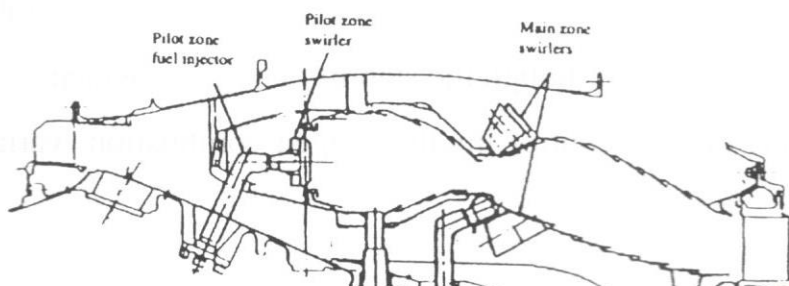
combustion, lean premixed prevaporised combustion and catalytic combustion. Staged combustor (Fig. 3) can incorporate either air-staging, fuel-staging, or both. Air-staging is being widely used to reduce the peak temperature of the flame.

By burning fuel-rich in the primary zone, nitric oxide production can be retarded. However, the hot product of combustion has to be rapidly cooled by injecting additional air downstream of the primary zone. Nimmo et al. (1991) studied the effect of air-staging using fuel-oil on a ceramic-lined tunnel furnace (0.8 m diameter x 3 m). They demonstrated in their study that addition of additional air downstream of the primary zone reduced the level of *NO* by about 45% at around a maximum of 40% secondary air. However, there was an increase in the level of carbon monoxide and nitrous oxide.

Fuel-staging is described by many workers as either a reburning process or thermal de NO_x process. This is the second method mentioned earlier of abating *NO* level, i.e., through post combustion treatment. In the reburning process, a secondary fuel is introduced in the second stage downstream of the lean primary zone to give a rich overall mixture. Tertiary air is then added further downstream to burn out *CO* and soot and give the required overall excess air. Many types of fuel can be used as the secondary fuel in order to convert *NO* back to molecular nitrogen, however, many workers in this field have agreed that natural gas is the best candidate. This method was first proposed by Wendt et al. (1973) and was demonstrated to be successful by Takahashi et al. (1983) by obtaining a *NO* reduction of at least 50%. The injection of natural gas in the reburn zone reduces *NO* to molecular nitrogen via the following reaction:



HCN participates in the shuffle reactions leading to the formation of a partially



equilibrated pool of NH_i species. The amine radicals either react with NO to produce molecular nitrogen or are oxidised to reform NO . In the present work an adoption of this reburn technique to low NO_x burner is investigated.

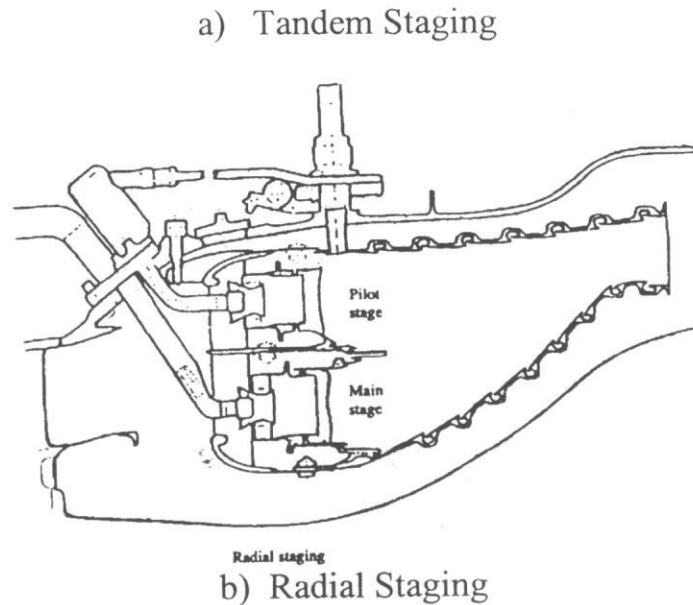


Fig.3 Staged combustors

In the thermal de NO_x process, nitrogen containing compound especially ammonia is injected as the secondary fuel [22,23]. These nitrogen containing compound will reduce NO in the post combustion product to molecular nitrogen. Reactions involved for the injection of ammonia are shown below:



or



However, the main drawback for this method is that the process works only in a narrow temperature window centred at approximately 1250K (roughly between 1100K and 1400K) in the absence of other additives [24]. Below 1000K, the

reaction is too slow to be of any use, whereas above 1500K, the addition of nitrogen containing compound will result in net increase in nitric oxide. This temperature window can be shifted to a lower temperature by adding hydrogen. However, the width of the window remained unchanged. Even then, a 90% reduction of *NO* level was reported when employing this technique.

A variable geometry combustor seems to be the ideal solution to overcome both the high power and low power operating range of burner. For minimum burner emissions if the thermal input is reduced from say 50 to 5 kW at constant excess air then the airflow is reduced by a factor of 10. For a fixed flow blockage burner this reduce the pressure loss by a factor of 100 which reduce the fuel and air mixing and the flow turbulence. If the burner flow area is reduced so that it acts as a flow control valve then the burner pressure loss is constant as the flow is reduced. Mechanical complexities involved seem to repulse burner designers from developing this technique. However, the considerable potential that variable geometry holds for emissions reduction has led to a renewal of interest in its application for both burner and gas turbine combustor [1,21]. In gas turbine combustor variable geometry is required to overcome flame stability problems in the lean low NO_x primary zones as the power is reduced by reducing the fuel flow at constant air flow.

Investigations by Saintsbury and Sampath (1974) on a simple variable geometry device by using variable baffle in the air stream surrounding an annular combustor has demonstrated excellent potential for reducing emissions from annular gas turbine combustors. It was demonstrated that combustion efficiency was improved and emissions were reduced especially carbon monoxide and unburned hydrocarbon. Nitric oxide emissions were also reduced at the high power condition.

Another attractive technique for reducing nitric oxide emissions is the lean-premixed or well mixed combustion or lean-premixed-prevaporised combustion for liquid fuel. The key factor here is to attain complete evaporation of the fuel and complete mixing of fuel and air prior to combustion. Avoiding droplet combustion

and operating the burner fuel-lean result in low reaction temperature and the elimination of hot spots in the combustion zone, and thus drastically reduced nitric oxide emissions [23]. However, there are some drawbacks for this technique. Sotheren, Pearce and Overton (1985) pointed out that the risks of autoignition and flashback in the premixing duct were perhaps the most severe problem. Other problem involved ensuring flame stability, stabiliser stability and combustion efficiencies. This lean burner technique is investigated in the present work but using rapid swirling jet shear layer mixing of fuel and air to achieve near premixed performance. In domestic central heating boiler an excess air of 30% is common for low CO control and this suitable for well mixed lean burn solution to low NO_x emissions.

Catalytic combustion involves the use of catalysts which allow fuel oxidation to take place at temperatures well below the lean flammability limit of the fuel. Due to this reason, the use of catalysts in gas turbine combustors to replace part of the thermal reaction zone allows stable combustion with peak temperatures that are about 1000K lower than those of conventional techniques. As nitric oxide emissions vary exponentially with reaction temperature, combustion at much lower temperatures can be expected to decrease the production of thermal NO . A schematic diagram of a catalytic combustor is shown in Figure 4 [19]. Burner system can also operate with catalytic combustion but only by using high excess air (>100%) to limit the catalyst temperature which gives a poor thermal efficiency.

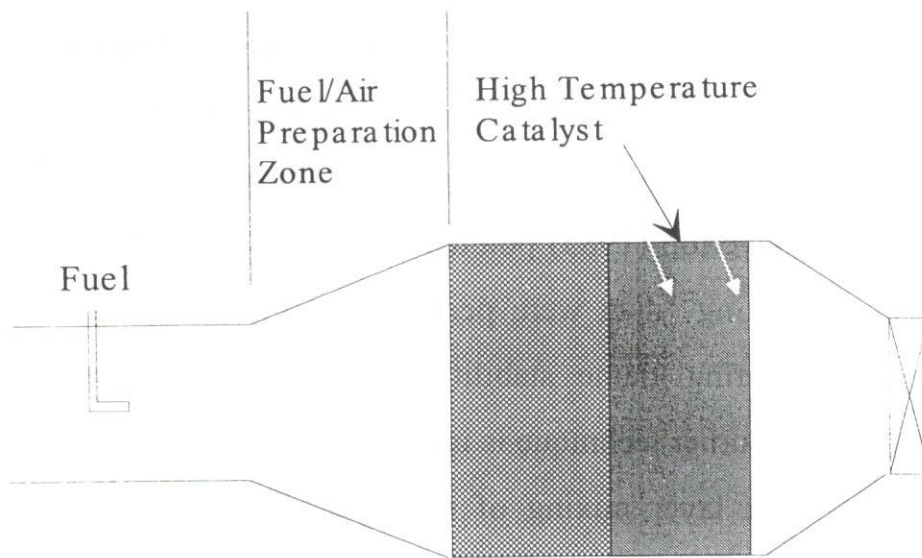


Fig.4 Catalytic Combustion [19]

Mularz, Gleason and Dodds (1978) investigated the use of a catalytic converter combustor and demonstrated emissions levels for carbon monoxide, unburned hydrocarbon and nitric oxide lower than the proposed standards of EPA 1981. The disadvantage of using catalytic combustor is the tendency of autoignition of the fuel upstream of the catalyst. Another disadvantage pointed by Mularz, Gleason and Dodds (1978) is that the temperature of the gases approaching the catalysts bed must be carefully controlled to prevent catalyst bed damage.

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