

**INHIBITION OF GASEOUS COMBUSTION MIXTURES USING
CERTAIN ADDITIVES**

By

Ruba Khaled Dabbas

Supervisor

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Professor Mohammed A. Hamdan

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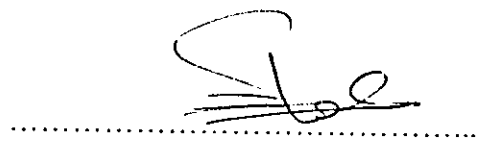

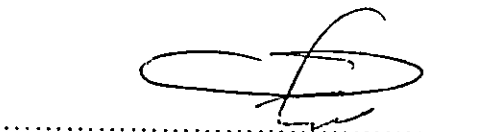
Faculty of Graduate Studies
University of Jordan

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COMMITTEE DECISION

This thesis was successfully defended and approved on:

Examination Committee	Signature
Prof. M. A. Hamdan, Chairman Professor	
Dr. Mahmoud A. Hammad, Member Associate Professor	
Dr. Ali Badran, Member Associate Professor	
Dr. Bilal Al-Okush, Member Assistant Professor	

DEDICATION

TO MY PARENTS & FAMILY,

FOR THEIR ENCOURAGEMENT AND SUPPORT

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ABSTRACT**INHIBITION OF GASEOUS COMBUSTION MIXTURES USING CERTAIN
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This thesis describes the experimental work to study the inhibitory effects of some solids on fuel-air flames, the fuel is LPG (Liquefied Petroleum Gas), which contains 70% butane and 30% propane. The flammable range for LPG-air mixture is found to range within 8.96% - 17.3%.

Inhibitors, which are used in this project, are calcium carbonate and stone, which is firestone (flint). Their inhibitory effects are studied on the lower flammability limits. Calcium carbonate shows an inhibitory effect superior by an order of magnitude. While Stone shows an inhibitory effect lower than calcium carbonate.

CHAPTER ONE

INTRODUCTION

1.1 GENERAL:

Due to its destruction of stratospheric ozone, the production of the widely used and efficient fire suppressant means such as Halon 1301 (CF_3Br) has been discontinued, and a number of alternate agents have been proposed. Since these agents are not as effective as CF_3Br and similar fire fighting material, there exists a need to understand the mechanism of inhibition and suppression of these proposed alternatives to help guide the search for more effective agents.

A large number of experimental and modelling studies have focused on the impact of halogenated inhibitors on hydrocarbon combustion, in either premixed or non-premixed flame. Such investigations have involved measurement of flammability limits, which are important to be studied in safety consideration especially in industrial operation. For example, in mines a flammable mixture may be easily build up, or in an aircraft during flight, where a shock wave preceding a turbulent flame can occur in a combustible mixture which is confined in space when the mixture composition falls within the appropriate range.

1.2 OBJECTIVES OF THE PRESENT STUDY:

The main objective of this study is to find a suitable inhibitor to replace Halon 1301. This will be carried out by studying the effect of addition of such inhibitor on the flammability limits of the combustible mixture.

1.3 THESIS LAYOUT:

This thesis consists of six chapters. The first of which is this introduction. The literature review of previous studies that have been carried out on flame inhibition is presented in chapter two. Chapter three is a background of the main points in this work, flammability limits and inhibitors. Chapter four reviews the theory. The experiment apparatus, procedures and observations are presented in chapter five. In chapter six, the results are obtained and discussed. Finally, the conclusions of the work and recommendations are given in chapter seven.

CHAPTER TWO

LITERATURE REVIEW

In this chapter a review of previous studies that were conducted in the field of flame inhibition will be discussed. Scientists have been concerned in studying inhibition and flammability limits since the beginning of the century; some of the recent studies will be introduced here.

2.1 PREVIOUS STUDIES:

The effect of a range of inhibitors and inorganic halogen compounds, was investigated on fuels such as H₂-Air and CH₄-Air Mixtures, (Hamdan, 1977). It was found that the degree of inhibition of some of these inhibitors is more effective on CH₄-Air than it is on hydrogen-Air.

It was shown that CF₃Br, known commercially as halon 1301, is more effective in inhibiting non-premixed flames of hydrocarbon fuels than thermal inhibitors such as N₂ and Ar due to its chemical inhibition effects, (Masri, 1993). When CF₃Br was mixed with the fuel, it decomposed in the very rich zone and yielded intermediate species like CH₃Br, CF₃H and CH₂CF₂. These species were not generated when the CF₃Br was mixed with the counterflow air. This was because the methyl radical, CH₃, which was the reason for the formation of these intermediates, existed only in rich mixtures and didn't interact with the Halon when the latter issued from the air side.

Further studies were done on CF_3Br and N_2 using non-premixed flames, (Trees et al., 1994). These two were compared to eleven gaseous agents; the results were based on mass and mole basis. CF_3Br was found to be the most effective in quenching the flames and the mass-based effectiveness of the other eleven agents was found to be nearly the same as that of nitrogen.

Numerical simulations of a structure of laminar counter-flow methane-air non-premixed flames with chemicals (chloromethane) and inert gas (nitrogen) added to the oxidizer stream, were performed using previous developed computer code, (Yang et al., 1994). When nitrogen was added, the flame temperatures were lowered, leading to a decrease in the concentrations of OH, O and H atoms. While chloromethane addition dramatically altered the flame structure, causing a decrease in concentrations of: OH, O and H atoms, and increase the flame temperatures.

Other studies were carried out to find how can inhibitors such as CF_3H and CF_2H_2 reduced the burning rate slightly more than if they were inert, (Linteris, 1994). Contrary to expectations, C_2F_6 reduced the burning rate more than a factor of two greater than if it were inert. The experimental and modeling results were studied on a premixed methane-air flame at inhibitor concentration reaching about 4%.

Further studies were concerned with the formation rate of HF (acid gas) in diffusion flames, which was strongly influenced by the mass flux of inhibitor into the flame sheet, (Linteris, 1995). Many of the agents (for example $\text{C}_2\text{H}_2\text{F}_4$, C_2HClF_4 , $\text{C}_3\text{H}_2\text{F}_6$, $\text{CH}_2\text{F}_2/\text{C}_2\text{HF}_5$, CF_3Br and CHClF_2) produced HF at rates within about 25% of that given by equilibrium thermodynamics in the diffusion flames tested. Most of the perfluorinated

agents tested (C_2F_6 , C_3F_8 , and C_4F_{10}) and the agents C_4F_8 , C_2HF_5 , and C_3HF_7 , produced 0 to 35% less than the equilibrium values. Co-flow diffusion flames with inhibitor added to the fuel stream show HF production rates ranging between 30 and 55% of the values given by equilibrium thermodynamics.

Extensive studies were carried out on diffusion flames by adding CF_3Br and CF_3H under normal gravity and micro-gravity conditions, (VanDerWege et al., 1995). Experiments showed that flames that couldn't be stabilised under normal gravity were quite stable under micro-gravity conditions. In addition, normal gravity experiments at reduced pressure did not reproduce the structure or stability limits of inhibited flames in micro-gravity.

Further tests on the reduction of burning rate were done numerically and experimentally for the inhibitors CF_3H , CH_2F_2 , and CF_4 in near-stoichiometric premixed methane-air flames, (Linteris, 1995). A decrease in the effectiveness of the inhibitors at higher concentrations was observed for all three agents both in the experiments and in the numerical calculations. When the inhibitor concentrations approached 8%, thermal decomposition reactions became more important, while below a concentration of about 5% inhibitor reactions involving fluorinated species cause a net increase in the consumption rate of H, O, and OH, while above 5% concentration, there is little additional radical consumption by these reactions.

Also the reduction in burning rate was determined experimentally and numerically for the fluorinated inhibitors C_2F_6 , C_2HF_5 , $C_2H_2F_4$, C_3F_8 and C_3HF_7 in near-stoichiometric premixed methane-air flames at inhibitor concentrations up to 8%, (Truett and Linteris,

1995). The calculations illustrated that these inhibitors were not inert in the present flames and show that the burning rate reductions are greater than could be accounted for by inhibitors acting as inert species.

An asymptotic analysis was performed to determine the influence of CF_3Br on the critical conditions of extinction of non-premixed methane-air flames, (Seshadri and Ilincic, 1995). The analysis considered addition of the inhibitor to the oxidiser stream of the non-premixed flame. Chemical reactions were presumed to occur in three distinct layers, which were the fuel consumption layer, the oxidation layer and the CF_3Br consumption layer. The asymptotic analysis predicted the value of the scalar dissipation rate at extinction to decrease and the corresponding value of the maximum flame to increase, with increasing concentration of CF_3Br in the oxidising stream. However the inhibiting effect of CF_3Br predicted by the asymptotic model was found to be weaker than that measured previously. The difference between the predictions of the asymptotic model and the measurements were attributed to the neglect of inhibition chemistry in the oxidation layer.

An experimental and numerical study were performed to characterise the critical conditions of extinction and the structure of counter-flow methane-air diffusion flame inhibited with CF_3Br and CF_3H , (Trees et al., 1995). It was found that the chemical kinetic mechanism employed is capable of predicting the critical conditions of the extinction methane-air flames inhibited with CF_3Br , especially if the agent was added to the oxidiser side. The results showed that the inhibition effect of CF_3Br is much stronger when added to the oxidiser stream of the counter-flow diffusion flame. When the

inhibiting effects of both CF_3Br and CF_3H were compared, it was found that the Br atom plays an important role in the mechanism of flame inhibition.

Experimental and numerical investigations were introduced using CF_3H as a fire suppressant with the oxidiser stream; to extinct the counter-flow $\text{CO}/\text{air}/\text{H}_2$ diffusion flame, (Fallon et al, 1996). In experiments, the extinction strain rate was evaluated by measuring the jet exit velocities at extinction and the jet separation distance. In numerical calculations the potential flow approximation was introduced to describe the outer flow field and the strain rate at extinction was determined from the axial velocity gradient on the oxidizer side.

The influence of CF_3Br , CF_3I , C_2F_6 , C_2HF_5 , CHF_3 , and CF_4 on a laminar flame propagation in air mixtures of CH_4 , CH_3OH , C_2H_6 , and C_2H_4 , was determined by numerical simulations, (Noto et al., 1996). Comparisons were made with experimental results and earlier calculations. For all organic fuels, the ranking of inhibition efficiency was CF_3Br , $\text{CF}_3\text{I} > \text{C}_2\text{F}_6 > \text{C}_2\text{HF}_5 > \text{CHF}_3 > \text{CF}_4$. The bromine and iodine compounds were clearly the most efficient flame suppressant.

The behavior of iron pentacarbonyl ($\text{Fe}(\text{CO})_5$) in methane/oxygen/nitrogen flames for diffusion and premixed flames was studied, (Reinelt and Linteris, 1996). In premixed flames, $\text{Fe}(\text{CO})_5$ at low concentrations reduced the burning velocities up to 100 times more effectively than CF_3Br and showed a strong dependence on fuel-air equivalence and oxygen mole fraction. However, at $\text{Fe}(\text{CO})_5$ mole fraction above 100 ppm, there was no additional reduction of the burning velocity. In diffusion flames, $\text{Fe}(\text{CO})_5$ was again shown to be more effective at the lowest concentrations, but the inhibition effect did not

level off at higher mole fractions as it did in premixed flames, the inhibition effect continued even above 500 ppm.

Further researches were done to find an alternative to the use of halogenated fire retardant, (Gilman et al., 1996). It was found that the flammability of a wide variety of polymers was dramatically reduced in the presence of relatively small concentrations of silica gel and potassium carbonate (K_2CO_3). The heat peak rate was reduced by 68% without significantly increasing the smoke or carbon monoxide levels during the combustion.

A numerical study of the inhibition effect of halogenated compounds was carried out for C_1 - C_2 hydrocarbon-air laminar premixed flames. Using CF_3Br , CF_3I , C_2HF_5 , C_2F_6 , CHF_3 , and CF_4 as inhibitors, (Noto et al, 1998). The burning velocity of C_1 - C_2 hydrocarbon decreased exponentially with increasing additive concentration over a wide range of additive concentration.

Further study was done on fluorinated inhibitors, numerically on fluoroethanes C_2F_6 , C_2HF_5 , and $C_2H_2F_7$, and experimentally for fluoropropanes C_3F_8 and C_3HF_7 , in near stoichiometric premixed methane air flames at inhibitor concentration 6%, (Linteris et al, 1998). The agents reduced the burning velocity of rich and stoichiometric flames primarily by raising the effective equivalence ratio and lowering the adiabatic flame temperature. For lean flames, the inhibition was primarily kinetic, since inhibitor reaction helped to maintain the final temperature.

Studies focused on the inhibition cycles, which regenerated the inhibitor, (Babushok et al., 1998). This led to the definition of an idealised or perfect cycle. It was demonstrated that for such an inhibitor in a stoichiometric methane-air flame, additive levels in 0.001-0.01 mole percent range would lead to a decrease in flame velocity of approximately 30%. This efficiency corresponded roughly to the behavior of metallic inhibitors such as $\text{Fe}(\text{CO})_5$, which is known to be 2 orders of magnitude more effective than currently used suppressants. This correspondence between the behavior of a perfect inhibitor and $\text{Fe}(\text{CO})_5$ led to the conclusion that the only gas-phase processes can account for its inhibitive power.

The flame inhibition mechanism of iron pentacarbonyl ($\text{Fe}(\text{CO})_5$) in premixed and counter-flow diffusion flames of methane, oxygen, and nitrogen was investigated, (Rumminger et al., 1999). For premixed flames numerical predictions of burning velocity were compared with experimental measurements, also for counter-flow diffusion flames, numerical prediction of extinction strain rate were compared with experimental results for addition of inhibitor to the air and fuel stream. The numerical predictions agreed reasonably well with experimental measurements at low inhibitor mole fraction, but at higher $\text{Fe}(\text{CO})_5$ mole fraction the simulation over predicted inhibition. The over prediction was suggested to be due to condensation of iron-containing compounds since calculated super-saturation ratios Fe and FeO are significantly higher than unity in some regions of the flame. These results led to the conclusion that the inhibition occurred primarily by homogenous gas-phase chemistry.

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It was noticed from these previous studies that most of the works were concentrated on gaseous inhibitors with little work that carried out using solid inhibitors.

In this work calcium carbonate and stone, which was firestone (flint), were used to inhibit diffusion premixed LPG-air mixture.

CHAPTER THREE

FLAMMABILITY LIMITS AND INHIBITORS

When discussing flame inhibition, one should discuss flammability limits and inhibitors. These limits are varied when inhibitors are added to combustible mixtures.

3.1 FLAMMABILITY LIMITS:

Combustible mixtures will only ignite and burn over a well-specified range of compositions, (Hamdan, 1977). The mixture will not burn when the composition is lower than the *lower flammable limit (LFL)*; the mixture is too lean for combustion to occur. The mixture will not burn when the composition is higher than the *upper flammable limit (UFL)*; the mixture is too rich for combustion to occur. A mixture is flammable only when the composition is between the LFL and the UFL. Typical units are volume percent fuels as a percentage of fuel and air. Lower explosive limit (LEL) and upper explosive limit (UEL) are used interchangeably with LFL and UFL. For example, a mixture of hydrogen and air containing 4% hydrogen will not ignite. As the proportion of hydrogen is gradually increased ignition will not happen until 5% hydrogen is reached, which is called the *Lower Flammable Limit* or the *Lean Limit*. If the proportion of hydrogen is further increased ignition become progressively easier and then once again harder until at 75% of hydrogen ignition will not again occur, which is called the *Upper Flammable limit* or the *Rich Limit*. As the mixture composition approaches either of these limits it is found that the burning velocity reduces. The burning velocities at both flammability limits are not necessary to be equal.

To illustrate the above, consider a plot of burning velocity against the composition of gaseous mixture on a volume basis v/v, Figure 3.1. The Figure illustrate that:

- Point A* Lower Flammable Limit
- Point B* Upper Flammable Limit
- Point C* Composition of mixture with maximum burning velocity
- Point D* Burning Velocity at Lean Limit
- Point E* Burning Velocity at Rich Limit
- Point F* Stoichiometric point for the mixture concerned

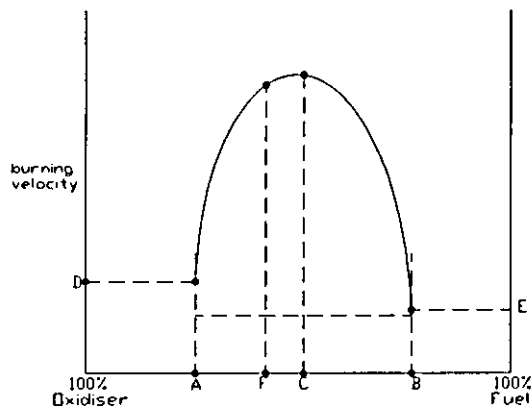


Figure 3.1, an illustrated drawing for flammability limits, (Hamdan, 1977).

Flammability limits have their importance in safety consideration, especially in industrial operation; in mines where one can easily get flammable mixture building up. The wider the two limits are the more a potential hazard is the mixture, (Hamdan, 1977). Table 3.1 shows flammability limits for most popular gases.

Flammable Mixture	Lower Limit (fuel %)	Upper Limit (fuel %)
Hydrogen/Air	4% (v/v)	75% (v/v)
Methane/Air	5.3% (v/v)	15% (v/v)
Ethyl Chloride/Air	4.25% (v/v)	14.3% (v/v)
Hydrogen/Oxygen	4% (v/v)	94% (v/v)
Methane/Oxygen	5.1% (v/v)	61% (v/v)
Ethyl Chloride/Oxygen	4% (v/v)	67.2% (v/v)

Table 3.1, flammability limits for most popular gases, (all limits are at a pressure of 1 atmosphere), (Hamdan, 1977).

Also flammability limits are important in many air-conditioning and refrigeration machines. There is now a halt on using chlorofluorocarbons (CFCs) because of their destruction of ozone layer, (Grosshandler et al., 1998). Hydrofluorocarbons (HFCs), such as R-32 (CH_2F_2), are one class of compounds that are being used as replacements. Unfortunately, more hydrogen on an HFC is associated with an increase in flammability.

The lower flammability limit is raised by an increase in static pressure above 1 atmospheric, (Hamdan, 1977), but the rich limit may be narrowed or widened. The effect being specific to the fuel, for example, for hydrogen and hydrocarbon – air mixtures the

range at upper limit is increased as the pressure is raised. However as pressure decreases the limits also tend to narrow and at very low pressures no mixture is able to burn because the two limits have merged together.

The flammability limits are experimentally determined data. The flammability limits in air depend on initial temperature and pressure, (Bjerketvedt et al., 1992). Standard test conditions are 25°C and 1 atm. Figure 3.2 shows the flammable range for some fuel-air mixtures.

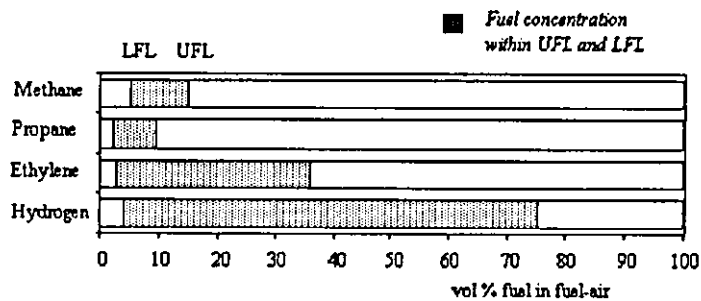


Figure 3.2 Flammable range for fuel-air mixtures at 1 atm. and 25°C, (Bjerketvedt et al., 1992).

The wide flammable range of hydrogen indicates that it is easy to get a flammable cloud of hydrogen in air. For propane and methane, the flammable range is much narrower, but an ignition source may "sit and wait" until the cloud can be ignited and explode. If the UFL has been passed, one has to go through the flammable concentration in the dilution process. It is good practice to operate safely below the LFL. As shown in Figure 3.3, the flammable range will widen when the initial temperature is increased.

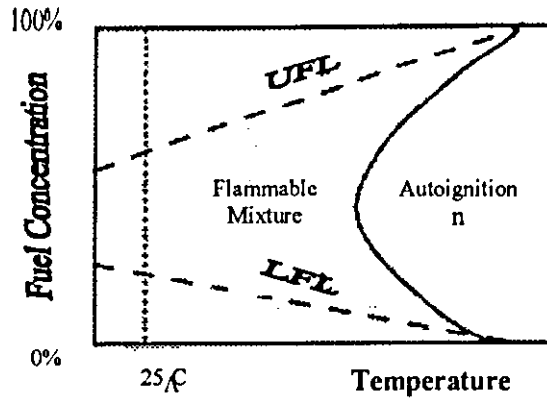


Figure 3.3 the effect of temperature on LFL and UFL, (Bjerketvedt et al., 1992).

Changes in initial pressure will, for hydrocarbons in air, not change the LFL significantly, but the UFL will increase. Flammability limits for fuel mixtures may be calculated by Le Chatelier's law:

$$\text{LFL}_{\text{Mix}} = \frac{100}{\frac{C_1}{\text{LFL}_1} + \frac{C_2}{\text{LFL}_2} + \dots + \frac{C_i}{\text{LFL}_i}} \quad (3.1)$$

Where, $C_1, C_2 \dots C_i$ [vol.%] is the proportion of each gas in the fuel mixture without air.

It was found that a good agreement existed between experiments and Le Chatelier's law for LFL at elevated temperature and pressure for fuel mixtures. It should be noted, however, that the formula does not work properly for H_2 and for unsaturated hydrocarbons. It is also only valid if the components are chemically similar.

3.2 INHIBITORS:

It has been found that the addition of certain inorganic or organic compounds to flammable mixtures will more efficiently decrease the range of flammability. Investigations into extinguishing fires using these types of compounds have been a topic of fire safety research for many years. The increase in inhibition is due to the chemical interaction of the inhibitor with the combustion process.

Organic halogen and halogen acids are known to be very good flame inhibitors. Their inhibitory effects on flames have been studied by means of measurements of flammability limits, burning velocity, and flame structure. The studies show that bromine and iodine compounds are very effective in inhibiting flame propagation, while the chlorine compounds are not so effective.

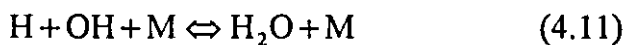
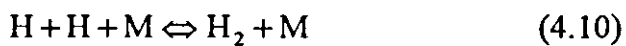
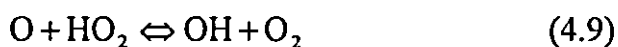
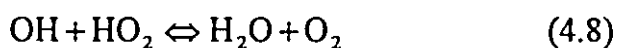
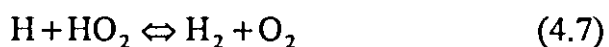
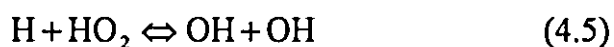
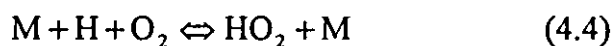
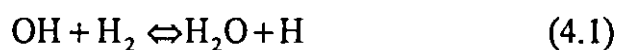
Metal compounds have been found to be up to several orders of magnitude more effective flame suppressants than the halogens. In particular, iron pentacarbonyl, was found to be one of the strongest inhibitors at reducing the burning velocity of premixed flame.

The reduction in the premixed laminar burning rate when adding inhibitors is useful for understanding the mechanism of chemical inhibition of fire since the diffusion flames often have a stabilization region which is premixed. A good correlation has been found between the reduction in burning rate and the concentration of inhibitors found to extinguish diffusion flames.

CHAPTER FOUR

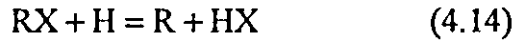
THEORY

In the reaction zone of a flame and due to the presence of the radicals H, OH, O the following reaction may take place, (Bulewicz et al, 1962):

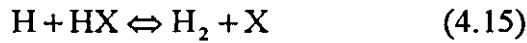


In fuel rich systems OH and O are present in much smaller concentrations than H₂ and the key reactions which consume molecules of oxygen are the reactions with H atoms. Inhibition may therefore occur by elimination of H; introducing chemical additives into the flammable mixture usually does this.

The most important inhibitors are the halogen compounds. When they are added to the rich fuel, the most important reaction is the formation of the halogen acid HX;

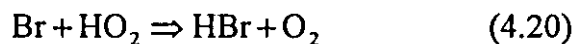
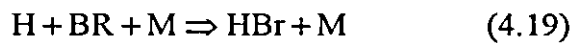
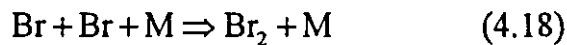
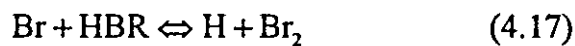
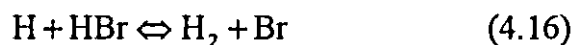


and in this case the essential role of the halogen atom will depend on the behavior of the halogen acid. The later probably enters the reversible reaction:



The position of equilibrium in reaction (4.15) depends on the halogen under consideration. For a less reactive halogen atom like Br and I the equilibrium may lie far over to the right in much of the flame and in principle this low reactivity itself may cause the inhibition by temporarily removing H atoms from the flame zone.

It was shown that this effect alone was insufficient by an order or magnitude to account for the observed inhibiting effects. These inhibiting effects could only be explained in terms of the series of reaction (4.16) to (4.20). Reaction (4.18) to (4.20) are three radical recombination steps, which control the radical pool size:



For realistic values of all the rate coefficient reaction (4.16) and perhaps (4.17) are effectively equilibrated in all but the early stages of the flame. The recombination steps then cause a new formation of HBr or Br₂, and the latter are recycled by reaction with H atoms in order to preserve the partial equilibrium. The HBr initially added thus in effect acts as an efficient homogeneous catalyst for the recombination of H atoms, since the equilibrium of (4.16) lies far to the right and equilibrium of (4.17) lies far to the left in most of the flame. Both the initial establishments of the partial equilibrium and the catalyzed recombination assist in reducing the concentration of H atoms available for the main flame reactions. In the general case the efficiency of the halogen acid, which acts as an inhibition, depends on:

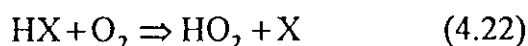
- a) The position of the partial equilibrium analogous to reactions (4.16) and (4.17).
- b) The rate coefficient of recombination steps like (4.18), (4.19) and (4.20).

A number of miscellaneous factors may reduce the effectiveness of halogen inhibition (4.19). Among these factors:

- a) The decomposition of the halogen acids:



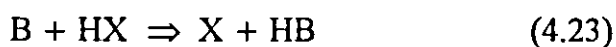
- b) The oxidation of the halogen acids:



- c) The failure of the carrier to decompose.

Factor (a) and (b) probably negligible in hydrocarbons-air flames will increase in magnitude from HF to HI and may in unusually hot flames become important. The

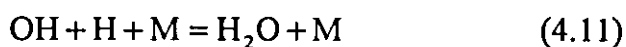
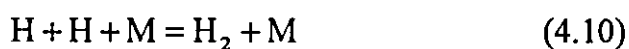
inclusion factor (a) underlines a critical factor in the mechanism the strength of the HX bond, if too strong the rate of reaction is correspondingly reduced, if it is too weak, HX will decompose rather than react.



Where,

B is such obstructing species as H, OH, CH₃ etc.

Less information about the mechanism of inorganic salts is available than for the halogen compounds. In the reaction zone of the rich-fuel flame the concentration of H atoms is usually in excess. In order to achieve thermal equilibrium then recombination of H atoms is to take place. Such recombination usually occurs in the post-reaction zone by collisions between hydrogen atoms and a third body (M), usually H₂, N₂ or H₂O, represented in the following reactions:



The addition of inorganic chemicals will catalyze these reactions since the additives will act as catalysts for the above reactions, hence the concentration of H atoms available for the main reactions in the flame will be reduced.

It was found that the mechanism of the inhibition using inorganic additives is both homogenous and heterogeneous, i.e. the inhibition is taking place on the surface of the inhibitor particles.

CHAPTER FIVE

EXPERIMENTAL APPARATUS, PROCEDURES AND OBSERVATIONS

5.1 EXPERIMENTAL APPARATUS:

The limits of flammability may be determined in a tall vertical tube of certain specified dimensions, (Hamdan, 1977), and under the variety of system and conditions, which are:

1. upward propagation, downward propagation
2. electric spark ignition, pilot flame ignition
3. atmospheric pressure, pressures above and below atmospheric

Variation in the dimension of the tube exerts some influence on the measurements. As a general rule flammability ranges widen as the tube diameter is made larger due to reduction in quenching effects, by the tube walls but the changes in limit for diameters above 5 cm diameter is very small, rarely exceeding a few tenth of 1%.

The apparatus used in this experiment is shown in figure 5.1, which is made from Pyrex glass. The apparatus consists of:

- A long cylindrical Pyrex vessel connects the parts of the apparatus, with inside diameter 20 mm and 1 m in length.
- Ignition tube with 7 cm in diameter where mixing and combustion occur.

- The inhibitors are introduced to the apparatus through the valve nears the ignition tube.
- A mercury manometer measures the gas mixtures, due to partial pressures.
- A vacuum pump is used to evacuate the apparatus, which introduced after a trap tube to prevent combustion remains and moisture from entering the pump.
- Two tubes are connected to the fuel and air cylinders
- To verify whether or not a mixture is able to propagate to the top of the ignition tube a thermocouple assembly is installed as shown so a deflection on a galvanometer indicated that a certain mixture has successfully propagated to the top.

5.2 EXPERIMENTAL PROCEDURES:

The following steps were followed during the present work:

- The apparatus was evacuated by the vacuum pump.
- The fuel and air were introduced to the apparatus, where the fuel was the LPG (Liquefied Petroleum Gas) that contained 70% butane and 30% propane.
- Then LPG-air mixtures were made up by partial pressure to 67.3 cm of Hg (atmospheric pressure) using the manometer, for example, LPG was introduced until the manometer reading was 10 cm of Hg, and air would fill the remain which was 55.3 cm of Hg.
- The mixture was introduced to ignition tube, without adding inhibitors to determine flammability limits, and then left for a period of 45 minutes to one hour to mix by diffusion before being ignited.
- Ignition was started by the removal of the greased ground glass plate from the bottom of the tube and then entering a lighted match.

- The limits of flammability were determined by trial and error technique, by introducing different gas mixtures until the right mixture was found.
- For each trial the apparatus was thoroughly evacuated, then a different gas mixture was introduced until the right limits were found.
- After determining the flammability limits, inhibitors were added to different gas mixture to find their inhibition effect also by trial and error technique.
- After each trial the connecting tubes were thoroughly flushed with each gas before use to remove the effect of the previous trial.

5.3 EXPERIMENTAL OBSERVATIONS:

During the experimental work, the following observations were noted:

A. Without the addition of the inhibitor:

1. The flame would:
 - a) Propagate all the way to the top of the tube with an “umbrella shape”.
 - b) Propagate to a certain distance, usually lower half of the tube before being extinguished, “ at flammability limits”.
 - c) Not ignite at all, and the flame, if produced it is at the mouth of the ignition tube.
2. Approaching both flammability limits ignition became progressively harder while ignition became easier away from flammability limits.
3. No ignition occurred before the lower flammability limit. While ignition occurred at the mouth of the tube after the upper flammability limit.

4. The flame color was blue. And the flame speed reduced approaching the flammability limits, (extinct quickly).
5. At the midpoint of gas mixtures, point C “composition of mixture with maximum burning velocity” in figure 3.1, the flame ignited with strong pressure wave and a loud noise.
6. It was found out that when the time for diffusion was less than 45 min, at 30 min for example, the ignition started outside the ignition tube then continued inside the tube.

B. With the addition of the inhibitor:

1. The flame color was totally orange.
2. The flame was weak and didn't reach the upper part of the ignition tube, until it was inhibited quickly. The noise of ignition was low.
3. Calcium carbonate absorbed moisture from the atmosphere, so when it was introduced to the apparatus, it would stick to the glass, while stone didn't stick to it.

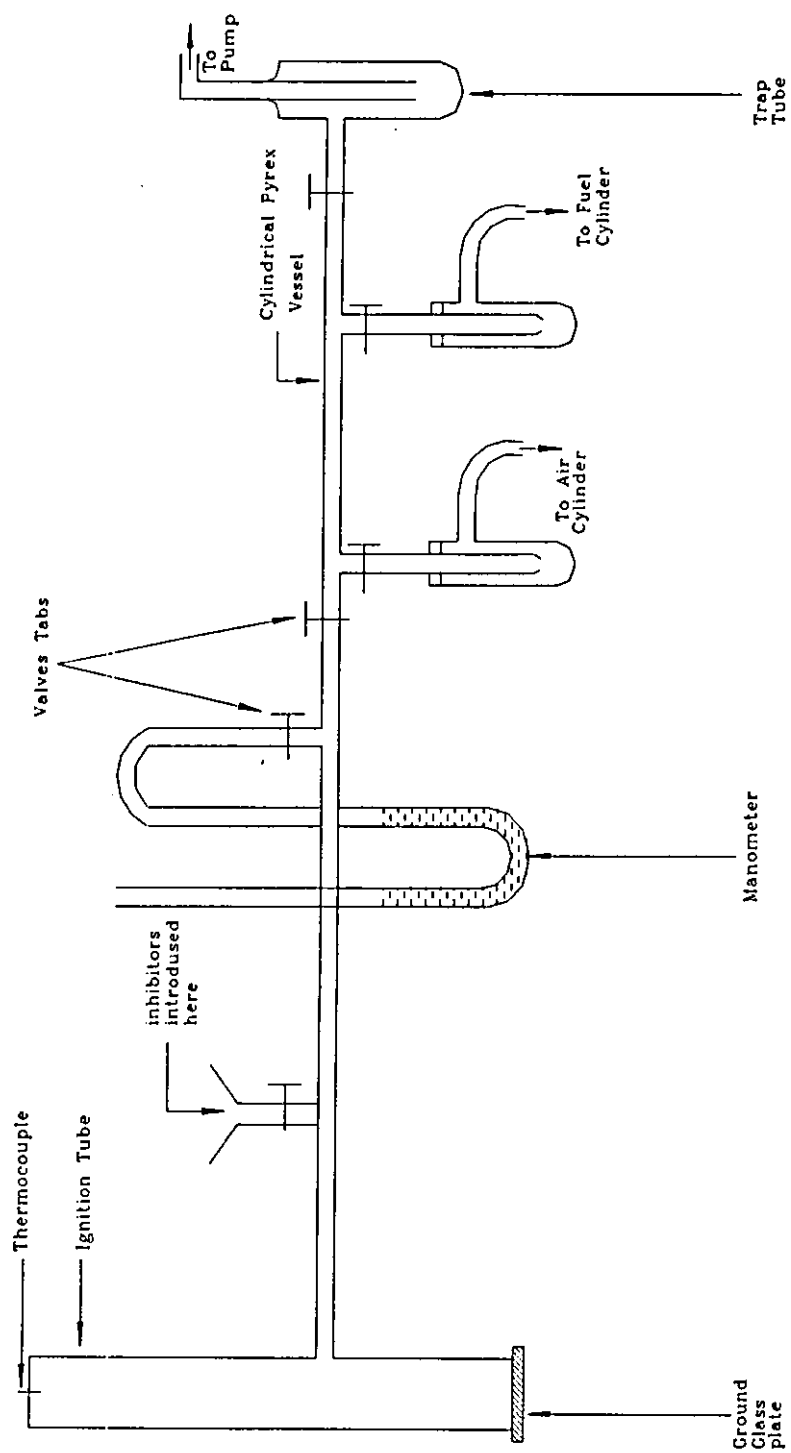


Figure 5.1 an illustrated drawing for the apparatus in the experiment

CHAPTER SIX

RESULTS AND DISCUSSION

6.1 RESULTS:

As mentioned earlier, the flammability limits were determined by trial and error method. It was found that the flammability limits for the LPG (butane 70%, propane 30%) are within (3 to 5.8 cm of Hg) 8.96% - 17.3% (at fuel percentage) and the flame colour was blue. At 17.3%, the UFL, the flame was strong with a loud noise inside the ignition tube, while at 8.96%, the LFL, the flame was weak and no noise was heard.

Inhibitors used in the experiment were calcium carbonate and stone, which was firestone (flint). Their effects on flammability limits are shown in tables 6.1 through 6.10.

6.2 DISCUSSION OF RESULTS:

The flammability limits for the LPG-air mixture is not wide because as mentioned earlier, air contains nitrogen and it has dilution effects, which agrees with Hamdan, (1977) and Bjerketvedt et al, (1992).

The results in tables 6.1 through 6.10 show the inhibitory effects of CaCO_3 and stone. The lean flammability limit is raised as the inhibitor amount increased. For example, the 10.4% are near the lean limit and no ignition occurs when adding 0.35g of CaCO_3 and at 0.55g of stone. This shows how both inhibitors raise the flammability limits.

Figures 6.1 and 6.2 show that both inhibitors affect the lower flammability limits and narrowed the flammable range. And when the inhibitor amount increases the LFL changes, for example, the combustible mixture at 10.4% will ignite when adding 0.3g of CaCO_3 , figure 6.1, while no ignition occurs when adding 0.5g, but it will ignite at 11.9%.

The same thing happens when adding stone, figure 6.2. Ignition for 10.4% is at 0.5g, while no ignition occurs when adding more stone the mixture will ignite again at 11.9%.

CaCO_3 is the inhibitor that showed the better inhibitory effect on the LPG; this is shown in tables 6.1 through 6.5. While the degree of inhibition of stone on LPG is low compared to CaCO_3 , as shown in tables 6.6 through 6.10. Figure 6.3 shows a comparison between both inhibitors, at fuel percentages. The figure shows that the amount of calcium carbonate needed to inhibit LPG-air flames is much lower than the amount of stone. For a 13.4%, when adding 1.0g of CaCO_3 , there is no ignition while the mixture will ignite when adding the same amount of stone.

Tables of results:

Using Calcium carbonate (CaCO_3):

- **Table 6.1**, ignition point at fuel percentage (10.4%, 3.5 cm of Hg):

CaCO_3	0.5 g	0.4 g	0.35 g	0.3 g
Status	No ignition	No ignition	No ignition	Ignition

- **Table 6.2**, ignition point at fuel percentage (11.9%, 4.0 cm of Hg):

CaCO_3	0.6 g	0.55 g	0.5 g
Status	No ignition	No ignition	Ignition

- **Table 6.3**, ignition point at fuel percentage (12.84%, 4.3 cm of Hg)

CaCO_3	0.9 g	0.8 g	0.75 g	0.7 g
Status	No ignition	No ignition	ignition	ignition

- **Table 6.4**, ignition point at fuel percentage (13.4%, 4.5 cm of Hg)

CaCO_3	1.0 g	0.95 g	0.9 g	0.8 g	0.7g
Status	No ignition	Ignition	Ignition	Ignition	Ignition

- Table 6.5, ignition point at fuel percentage (14.9%, 5.0 cm of Hg)

<i>CaCO₃</i>	<i>1.1 to 2.1 g</i>	<i>2.15 g</i>
Status	ignition	No ignition

Using stone:

- Table 6.6, ignition point at fuel percentage (10.4%, 3.5 cm of Hg)

<i>Stone</i>	<i>0.6 g</i>	<i>0.55 g</i>	<i>0.5 g</i>
Status	No ignition	No ignition	Ignition

- Table 6.7, ignition point at fuel percentage (11.9%, 4.0 cm of Hg)

<i>Stone</i>	<i>1.1 g</i>	<i>1.05 g</i>	<i>1.0 g</i>	<i>0.9 g</i>	<i>0.8 g</i>	<i>0.7 g</i>
Status	No ignition	Ignition	Ignition	Ignition	Ignition	Ignition

- Table 6.8, ignition point at fuel percentage (12.84%, 4.3 cm of Hg)

<i>Stone</i>	<i>1.6 g</i>	<i>1.55 g</i>	<i>1.5 to 1.2 g</i>
Status	No ignition	No ignition	Ignition

- Table 6.9, ignition point at fuel percentage (13.4%, 4.5 cm of Hg)

<i>Stone</i>	<i>1.9 g</i>	<i>1.85 g</i>	<i>1.8 g</i>	<i>1.7 g</i>
Status	No ignition	Ignition	Ignition	Ignition

- Table 6.10, ignition point at fuel percentage (14.9%, 5.0 cm of Hg)

<i>Stone</i>	<i>2.6 g</i>	<i>2.55 g</i>	<i>2.5 to 2.0 g</i>
Status	No ignition	Ignition	Ignition

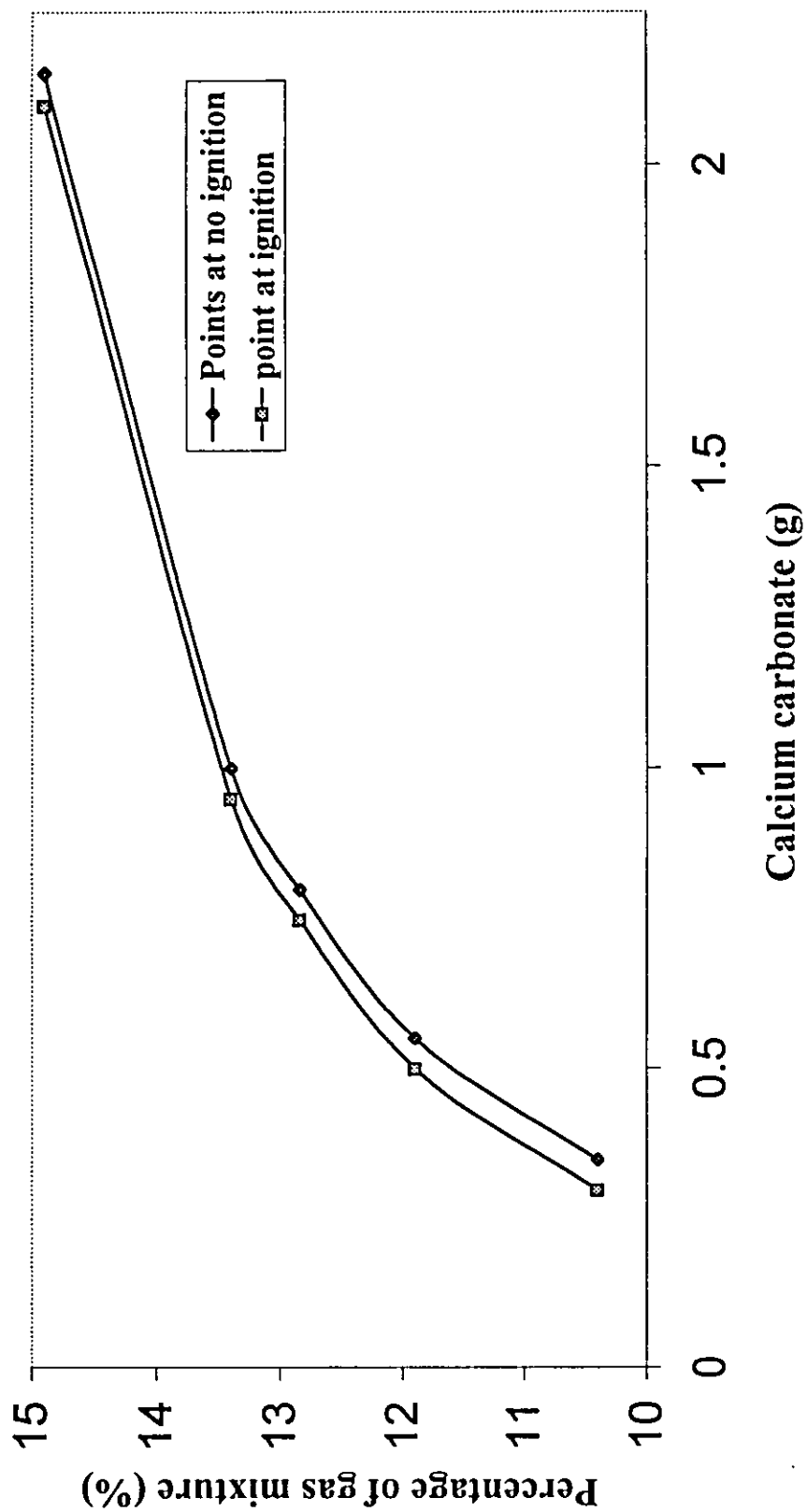


Figure 6.1. points of no ignition & ignition when using Calcium Carbonate

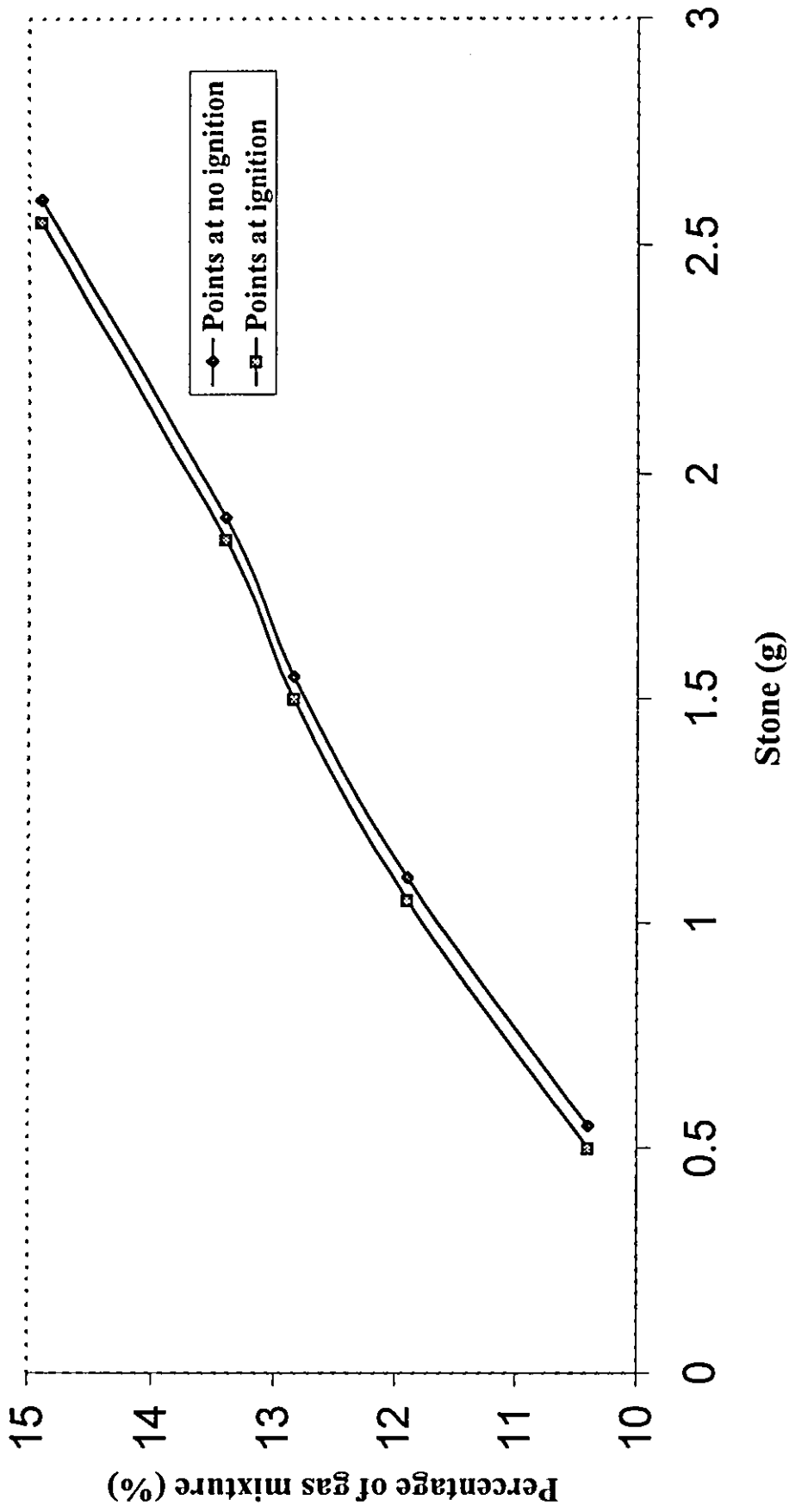


Figure 6.2. points of no ignition & no ignition when using stone

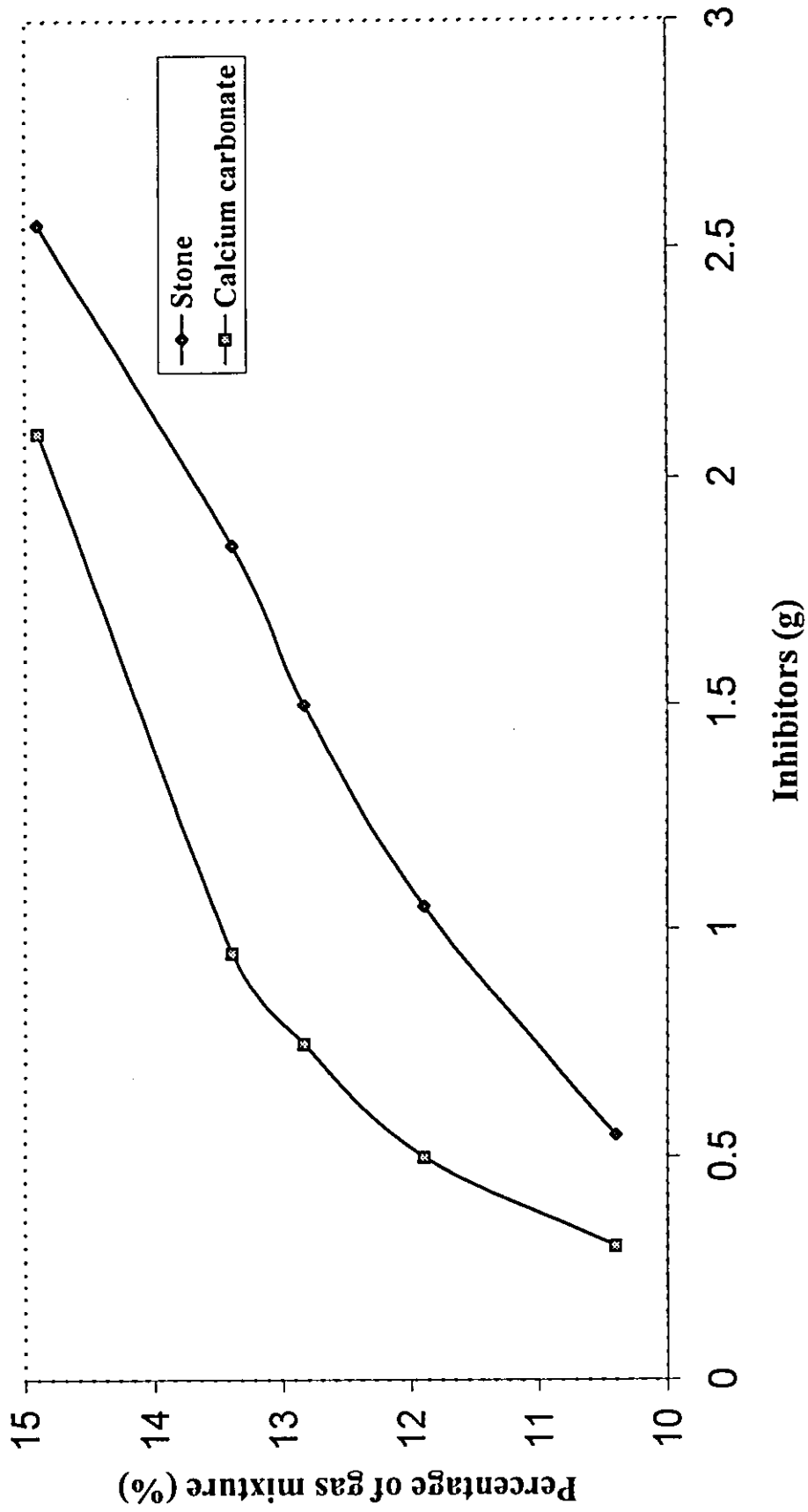


Figure 6.3, a comparison of inhibitory effects between Calcium carbonate & Stone

CHAPTER SEVEN

CONCLUSIONS AND RECOMMENDATIONS

7.1 CONCLUSIONS:

In an effort to construct a paradigm for flame inhibition by solid compounds, an experimental investigation was made for studying their inhibitory effects on premixed fuel- air flames. The fuel was LPG (Liquefied Petroleum Gas), which contained 70% butane and 30% propane.

The flammable range for LPG-air mixtures was found to range within 8.96%-17.3%, which resembled the flammable range for methane-air and propane-air mixtures in Hamdan (1977) and Bjerketvedt et al. (1992).

The solid inhibitors were calcium carbonate (CaCO_3) and stone, which was firestone (flint). The experiment showed that calcium carbonate acted as a better inhibitor than stone. Both inhibitors raised the lower flammability limit when added to the mixture, as the amount of inhibitor increased there was a distinct raise in the LFL.

7.2 RECOMMENDATIONS:

Suggestions for improvement in technique:

- The apparatus should be cleaned after the study of each trial, so that any combustion remains should be removed, which consume more time.

- Burning velocity and measurement of composition and temperature profiles should be studied, thus giving more information about the flame than already known, and hence a more complete knowledge of the mechanism of inhibition.
- More studies should be done on various types of solid inhibitors other than calcium carbonate and stone to get a better inhibitor such as clay.

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ملخص

إخماد مزيج غازي قابل للاحتراق باستخدام مواد مضافة

إعداد

ربي خالد أحمد الدباس

المشرف

أ.د. / محمد أحمد حمدان

تصف هذه الرسالة التجربة العملية التي أجريت بفرض دراسة تأثير بعض المواد الصلبة على إخماد لمب مزيج غاز- الهواء،

الغاز المستعمل هو غاز البترول السائل المكون من 70% بيوتين 30% بروين. وقد وجدت حدود اللهب لهذا المزيج أنها تتراوح ما بين

8.96% - 17.3%.

المواد المضافة التي استعملت في إخماد اللهب هي كربونات الكالسيوم والحجر، وهو حجر الصوان. وقد درس تأثيرهما على الحد

الأدنى للهب. كربونات الكالسيوم أظهرت تأثيرا عاليا عندما استعملت في إخماد اللهب، في حين أن الحجر أظهر تأثيرا أقل من كربونات

الكالسيوم.