DECOMPOSITION OF HYDROGEN SULFIDE IN AN ALTERNATING CURRENT, FREQUENCY TUNED PLASMA REACTOR

By

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CHAPTER I

INTRODUCTION

Hydrogen sulfide (H_2S) is a well known naturally occurring substance. It is commonly found in natural gas. It is also often present in refinery "off gas" streams and in exit gas streams from hydrodesulfurizers.

Most hydrocarbon materials whether gas, liquid or solid, occur in natural mixtures with varying sulfur contents. For various reasons this sulfur content must be lowered. The removal of sulfur from crude oil is achieved by hydrodesulphurization giving hydrogen sulfide. Sweetening of sour gas also produces large volumes of hydrogen sulfide. The volumes of hydrogen sulfide requiring processing will continue to increase. Clearly, if the hydrogen fraction of the H_2S is recovered it could be recycled back to hydrodesulfurization units to achieve further sulfur removal from the feed stock.

The Claus process is probably the only commercially successful method for processing hydrogen sulfide. While this is an effective technique for processing hydrogen sulfide, one valuable part of the molecule, the hydrogen is lost to water during the oxidation and reduction steps. This thesis investigates the use of a new technology, frequency tuned capacitive discharge reactors, to dissociate hydrogen sulfide into elemental sulfur and hydrogen.

Hydrogen sulfide is environmentally undesirable and is expensive to remove from waste gases. The Clean Air Act requires that H_2S be controlled to concentrations of less than 50 ppm. The findings of this research could well provide a simple alternative to conventional methods of processing H_2S from oil refinery " sour gases." The practical and economic implications could be far reaching.

A decomposition process yielding sulfur and hydrogen under moderate reaction conditions shows considerable commercial promise for a variety of applications in the hydrocarbon industry. The economic potential for such a process is staggering. Each year the United States produces almost seven million tons of waste H_2S. This represents a loss of more than 125 billion standard cubic feet of hydrogen per year accounting for an annual cost of over $225 million [11].

Circumventing the unfavorable thermodynamic requirements of the overall decomposition is the underlying key to using H_2S as a hydrogen source. It is important to note that this does not imply a violation of fundamental physical laws. It is however, possible to optimize product yields by carefully choosing various reaction parameters and process conditions without violating fundamental chemical concepts.

The decomposition of H_2S has already been accomplished in a thermal plasma reactor. However, the process conditions are extremely severe and require a very high input of power. According to recent reports, efforts at Kurchatov Institute, Moscow, Russia have moved on to an industrial scale with a 1.0MW installation at a natural gas processing facility [14]. This thesis is an attempt to achieve the decomposition of H_2S at atmospheric pressure and temperature and at low power consumptions.

Research on alternating current, silent glow discharge reactor (SGDR) began at Oklahoma State University (OSU) in 1987 as a cooperative effort with the Naval Research Center. To date, most of the research on these reactors has been done at the Naval Research Center and OSU. Work at Oklahoma State University [20,32] has shown that a SGDR can be fine tuned by varying the frequency of the voltage applied to the reactor. Much of the research reported in the literature came before the existence of an optimum frequency was known. Thus, many of the earlier experiments reported were carried out at a fixed frequency, which is generally much lower than optimum frequency. This single fact is the major reason why past investigators did not fully exploit and develop plasma reactors using alternating current [11].

An efficient glow discharge reactor has many potential industrial applications, such as:

- purification of stack gases from factories,
- emergency air purification in building and hazardous materials storage areas,
- destruction of hazardous waste gases ; and
- use as a chemical synthesis reactor.

The following advantages can be realized by destructing H_2S in a SGDR.

- The operating temperature would be very low.
- There will be no requirement of catalyst.
- The reaction can be carried out at or near atmospheric pressure.
- An extra salable product, namely hydrogen, can be obtained.
- The reactor itself can be used as a pollution control device for very low H_2S concentrations.

The objectives of this research are as follows.

- To build the experimental apparatus and have it operate properly and safely.
- To experimentally determine the key operational and design parameters for the reactor.
- To determine the degree of H_2S conversion.
- To assess the potential of the new technology and make further recommendation for continued research.

The preceding paragraphs should be sufficient to provide a strong justification for carrying out this study from an applied point of view. Also the thought of developing a simple and economic method itself would seem to provide additional motivation to undertake such a study.

A review of the literature on destruction of hydrogen sulfide is presented in Chapter 2. Chapter 3 describes the experimental set up and the safety factors considered in building the experimental apparatus. The experiments conducted are discussed in Chapters 4 and 5.

Results are presented in graphical manner and where necessary in tabular form. Chapter 6 gives a brief dicussion on the thermodynamic consideration. Finally a general discussion of all the relevant data obtained and their implications are discussed in Chapter 7.

CHAPTER II

LITERATURE REVIEW

Background

The Claus process involves the partial oxidation of hydrogen sulfide to SO_2 and subsequent reaction of H_2S and SO_2 to form water and sulfur. Overall this is a highly exothermic reaction. An alternative to the Claus process may well be based on non-oxidative decomposition of H_2S to H_2 and S. The advantage of having an extra salable product is counterbalanced by the fact that this reaction is endothermic. The decomposition of molecules into smaller molecular species can be brought about by various means. Essentially these processes require a sufficient energy supply to rupture the chemical bonds. A brief description of the plasma concept is given below. This chapter is further divided into sections that deal with different methods of H_2S destruction.

Plasma Origin

Plasma is defined as the state of ionized gases. It was described by Crookes in 1879 as "a world where matter may exist in a fourth state." In recent times this has attained an important and a crucial place in research and industry worldwide [5].

Sir William Crookes used a tube and evacuated it to a low pressure under which conditions electrical current can be forced through the gas. When excited by the electrical energy, the rarefied gas showed a glow. This tube then came to be known as the so called "glow discharge tube" and has become a basic tool of researchers in plasma studies. The "fourth state of matter" was given the name "Plasma" by Irving Langmuir in 1928 [5].

General Mechanism of Plasma Reactions [11, 30]

Gases are normally good electrical insulators. However, under the influence of an applied electric field of sufficient strength gas molecules can be ionized. Electric conduction then takes place and an electrical discharge occurs. Given the right current conditions, gas breakdown can occur and a discharge results. It actually fills the gap between the electrodes with a soft glow.

The alternating current silent glow discharge reactor (SGDR), also known as the alternating current corona reactor, glow discharge reactor, or alternating current silent discharge plasma reactor, utilizes electrical energy to create a relatively low temperature plasma (electric discharge) in a reactor cavity. When organic or inorganic materials flow in the plasma, their chemical bonds are broken by absorbing the electrical energy of the plasma. Elemental atoms and free radicals result, which then recombine to form simpler reaction products.

There are two types of common plasmas, thermal induced and electrically induced. An important difference exists between these two types of plasmas. Thermally induced plasmas are in thermal equilibrium, and the temperature of the neutrals and charged species are in equilibrium (isothermal). However, in electrically induced plasmas, the temperatures of the charged and neutral species can be quite different.

In glow discharges, induced by alternating current, the ions and molecules and free radicals are roughly at ambient temperatures, while the electron temperature is two to three orders of magnitude greater. Reactions in plasmas are fundamentally different than normal combustion reactions. In plasmas the initial step is believed to be rupture of the chemical bonds by electrons, in contrast to standard kinetic models, where the initial step in combustion reactions involves collision of the reactant molecules.

Plasma Decomposition of H_2S

Electrical discharge reactors have been used in the study of chemical reactions for many years. Different types of electrical discharge reactors have been used to study a wide range of phenomena. The earliest work began around 1927 and concentrated on direct current reactors.

The influence of the silent electrical discharge upon hydrogen sulfide was studied by Schwarz and Kunzer [23] in 1929. They used a Siemen tube inside a furnace and passed pure hydrogen sulfide at various velocities and temperatures. It was reported that dissociation took place to a certain extent and then was overcome by recombination caused by activation of the sulfur by the discharge. Their results showed that H_2S can be dissociated by an electric discharge and marked the beginning of research in this field.

In 1965, a technology that involved decomposition of H_2S into its constituent elements by electric discharge was patented by a Japanese company [15]. A glow discharge was carried out with a current of 0.264 amperes at 825 volts and 218 mm Hg pressure in a cylindrical electrode made of stainless steel. It was reported that after a discharge of 1 minute and with 1.63 liters of H_2S, 0.53 grams of S (99.6% pure) and 0.38 liter of H_2 were produced. The remaining gas consisted of unreacted H_2S. This technology is no longer in use presumably, due to being expensive and requiring low pressure.

Recent work by Soviet scientists reported the dissociation of H_2S in a non-equilibrium microwave generated plasma [2, 26]. The reported conversion ranged from 40% to 90%. However , no information is available regarding the specific reaction conditions.

Rusonov et al., [26] have studied the effect of spatial non-equilibrium in the decomposition of H_2S in a plasma. They showed that the centrifugal forces present in the discharge lead to spatial separation of the substances and as a consequence disturbed the chemical equilibrium, thereby slowing down the reverse reaction and increasing the conversion. The achievement of the centrifugal effect is believed to have reduced the energy consumption considerably.

Researchers at the Argonne National Laboratory [12] have reported that H_2S conversions in excess of 99% can be achieved by recycling unconverted H_2S back to a microwave plasma reactor. It was also reported that this process is more economical than the conventional waste treatment process. The reactor used in this research was reported similar to the one used by the Soviets.

Many theoretical and experimental studies have been devoted to the dissociation of H_2S in "thermal plasmas." The mechanism for this process and its output and energy characteristics have been investigated [2,21]. The rate of dissociation of H2S in a thermal plasma has been reported to be determined by a limiting step, $H_2S + M \rightarrow H + HS + M$.

Nester et al. [18] proposed addition of small amounts of oxygen in order to reduce the limiting significance of the above reaction. Potapkin and coworkers [21] have determined that the primary mechanism for dissociation of H_2S is a function of the amount of oxygen in the mixture. They concluded that the energy costs of obtaining the products is significantly reduced with the presence of oxygen in the initial mixture.

The dissociation of hydrogen sulfide mixed with propane in a thermal plasma was studied by Balebanov et al. [3]. They showed that hydrocarbons hinder making sulfur by H_2S decomposition in a thermal plasma. Also the energy consumed in making hydrogen decreases as the propane level rises. It was also reported that the yield characteristics improve when oxygen is added, at the start.

Vastola and Stacy [29] studied the effect of hydrocarbons on the plasma induced dissociation of H_2S. Results shown indicate that H_2S can be completely dissociated by an electric discharge to produce molecular hydrogen and carbon disulfide. These experiments were conducted using a power source of 1.2 kW and 120 MHz RF induction heater.

Reaction Mechanism of H_2S in a Thermal Plasma

Potapkin et al.,[21] have investigated numerically the dissociation process in thermal plasma. The main elementary stages of the process are established. The main reaction that determines the dissociation kinetics was found to be $H_2S \rightarrow H + HS$. The atomic hydrogen that was formed in this case entered into reaction with H_2S; $H + H_2S \rightarrow HS + H_2$. In their turn, the HS radicals reacted with each other to form molecular hydrogen and disulfur; $HS + HS \rightarrow H_2 + S_2$.

The following gives the complete mechanism proposed by Potapkin et al.

$$H_2S + M \Leftrightarrow H + HS + M$$

$$H + H_2S \Leftrightarrow H_2 + HS$$

$$HS + HS \Leftrightarrow H_2S + S$$

$$S + H_2S \Leftrightarrow H_2 + S_2$$

$$S + HS \Leftrightarrow H + S_2$$

$$H + HS \Leftrightarrow H_2 + S$$

$$HS + HS \Leftrightarrow H_2 + S_2$$

$$S + S + M \Leftrightarrow S_2 + M$$

$$S_2 + S_2 + M \Leftrightarrow S_4 + M$$

The rate constants for the forward reactions were reported to be taken from literature data; and the rate constants of the reverse reactions calculated via the equilibrium constants. This mechanism is a development of earlier kinetics proposed by Balebanov et al. [2]. The kinetic curves obtained by these two mechanisms have been reported to show agreement in the range of 3- 5% [21].The critical scheme proposed in reference 2 is given below. The mechanism used here is a very simplified one, where the effect of reverse reactions was not taken into account.

$$H_2S \rightarrow H + HS$$

$$H + H_2S \rightarrow H_2 + HS$$

$$HS + HS \rightarrow H_2S + S$$

$$S + H_2S \rightarrow H_2 + S_2$$

Temurova et al.[27] investigated the dissociation process in a barrier discharge plasma. They conducted an analysis of the mechanism reported in reference 2 and concluded that a mechanism involving ions and free radicals does not explain the experimental results. They based their conclusions on the account of energy consumption for the H_2 molecule, which was significantly higher according to the mechanism than that found experimentally in their research. They put forward the following mechanism:

$$(H_2S)^+ + n\,H_2S \rightarrow (H_2S)_n^+$$

$$(H_2S)_n^+ + e \rightarrow nH_2 + n/2S_2$$

While these are some significant reports on kinetics of H_2S decomposition in a thermal plasma, inconsistencies do seem to exist, suggesting the need for further investigation. Boenig Herman in his book [5], says "it is typical of plasma processes that, for a given molecular feed, a process can proceed chiefly via ionic reactions under one set of plasma parameters and chiefly by a free radical mechanism under a different set of plasma parameters".

Thermal Decomposition

Raymont in his authoritative work on the thermal decomposition of H_2S has reported that direct decomposition is a very unlikely contender for a viable process to produce hydrogen from H_2S. In his work [16], he suggested alternate techniques such as upset equilibrium and thermodynamic loop system as attractive routes to achieve hydrogen production.

Catalytic Decomposition

Hydrogen sulfide reacts with a wide variety of inorganic substances, particularly transition metals and oxides to give hydrogen and a sulfide. Reactions of this type have been extensively studied.

The use of Ni [13] and NiO [32] has been investigated in surface chemisorption of H_2S. In both cases H_2S decomposes at the surface of the solid resulting in the liberation of hydrogen, but the sulfur atoms are irreversibly bound either by strong chemisorption or by formation of molecular sulfide by reaction with the surface atoms of the solid.

The use of iron and nickel [6] for adsorption of H_2S has been studied. The interaction of H_2S with metals and non-metallic elements has been reported to result in the formation of sulfides from which sulfur cannot be recovered. Also, most of these processes require high temperature and high vacuum.

Radiolytic Decomposition

The alpha and gamma radiolysis of H_2S has been the subject of research in the recent past. However very little information is available and there is need for further investigation of this mode of decomposition. Torrey [25] dissociated H_2S into its elemental components by radiolysis. He deduced a mechanism involving high energy H_2 atoms and SH radicals. Huyton and Woodward [9] found that hydrogen and sulfur were produced with gamma particle radiation.

Photochemical Decomposition

The effect of light at various wavelengths on H_2S has been studied by several investigators. Darwent and Roberts [1] made an extensive study on the photochemical decomposition at various temperatures and pressures using cadmium and mercury lines as the energy source. They deduced a free radical mechanism for the photochemical reaction. They observed that the yield of H_2 was independent of H_2S at pressures above 200 Torr. However, temperature dependence of the yield has been reported to vary with wavelength of light used.

Very recently Gratzel [17] showed that H_2S can be split catalytically into its components by visible light. The use of an aqueous transparent suspension of colloidal cadmium sulfide particles loaded with ruthenium dioxide is believed to be the key to the process. This process has been patented in Switzerland.

Other Decomposition Methods

Ultrasonic irradiation has been shown to produce colloidal deposits of sulfur from aqueous hydrogen sulfide solutions [19]. Electrochemical oxidation has been reported to have brought about the decomposition and similar process have been patented both in Germany and USA.

As reported in reference [14] the petrochemical industry has been looking at several alternatives to the CLAUS/SCOT process in order to recover hydrogen, but none have developed beyond bench scale testing. The H_2S treatment alternatives apart from those discussed above include

- Chemically promoted thermal decomposition,
- Combination of electrolysis and chemical decomposition,
- Liquid-metal conversion;

These approaches produced hydrogen instead of water, but all were abandoned because of poor yield, high energy cost, or some other process limitations.

One process being studied at the Amoco Research Center involves a complex series of reactions, including an indirect electrochemical reaction followed by a purification step to recover sulfur [14]. The sulfur produced in this process was reported to be a sticky, gummy mass that proved very difficult to recover and purify.

Recently Shell and Dow have announced chelate-based H_2S waste treatment processes. The process is claimed to have better economics than the conventional Claus/Scot plant. However ,the process does not recover hydrogen, which still ends up as water.

In conclusion, it appears that the decomposition of hydrogen sulfide can only be brought about under relatively severe conditions and even then, the yields are poor as dictated by the thermodynamics.

CHAPTER III

EXPERIMENTAL APPARATUS AND SAFETY

The first phase of the experimental effort involved non-destructive runs using compressed air. This was mainly done to obtain the electrical characteristics of the plasma. The second phase of the experimental effort involved the destructive tests. The apparatus included a gas handling system, a reactor, the energy supply system (AC power source, oscillator, and transformer), exhaust scrubbers and auxiliary instrumentation. A schematic diagram of this apparatus is presented in Figure 1. Since hydrogen sulfide is a lethal gas, all destructive experiments were conducted at the OSU Hazardous Reactions Laboratory. The cylinders and the reactor system were housed in a very well ventillated blowout bays located outside the building.

Experimental Materials and Equipment

Materials

a. Compressed Gases :

H_2S - Linde Specialty Gases, 99.5% pure. Used without further purification or testing.

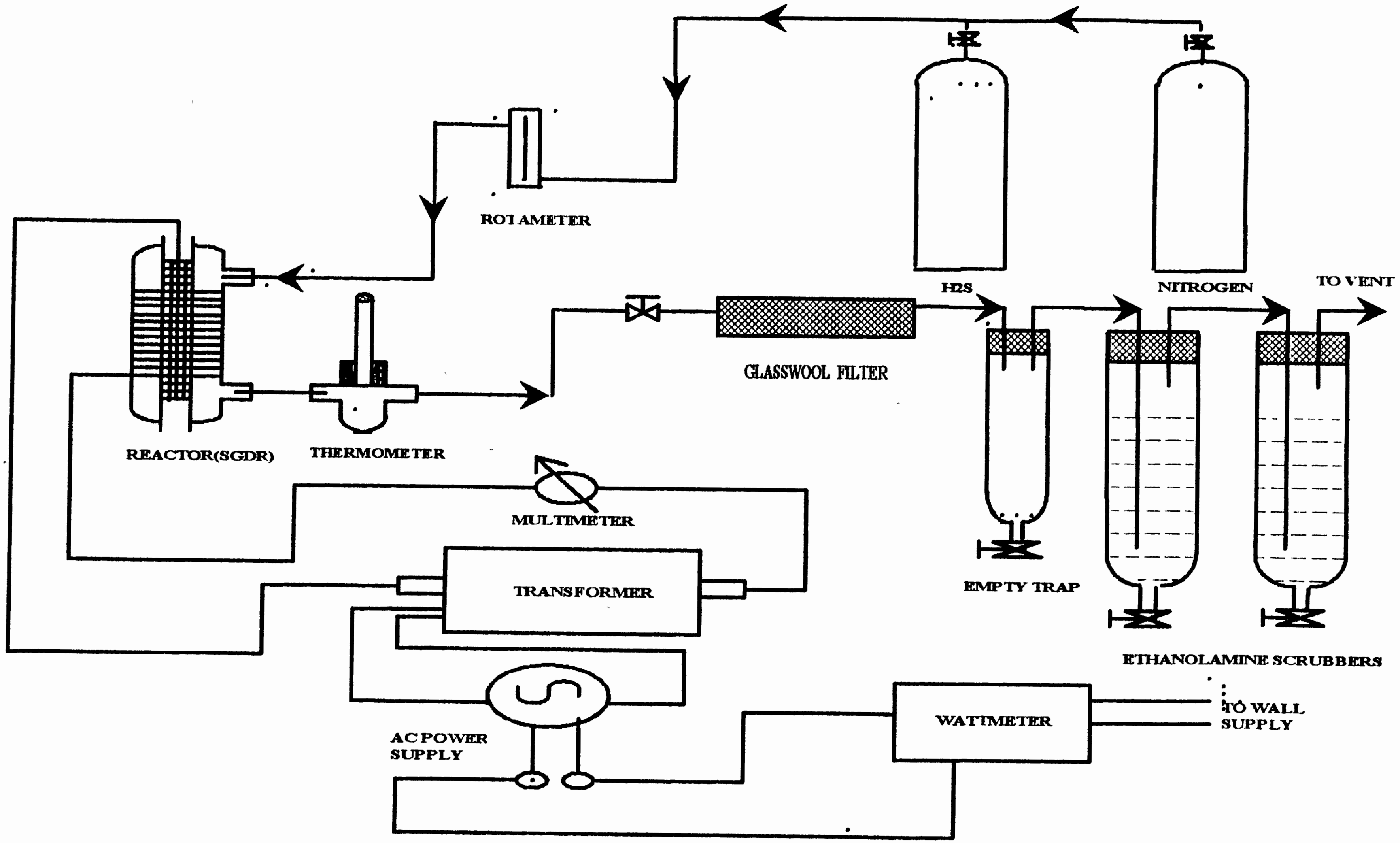

Figure 1 . Schematic of Experimental Apparatus

Air - Linde Specialty Gases, zero grade

N_2 - Linde Specialty Gases, dry grade 99.9% .

b. Solvents :

Ethanol amine - Fisher Scientific

Carbon disulfide - Aldrich Chemical

Equipment

The following Equipment was used in the experiments.

a. AC Power Supply - California Instruments, Model 1001TC

b. Oscillator-California Instruments, Model 850 T

c. Flow metering - Rotameter from Fischer and Porter Co., Model No. 10A6\32N, Tube Specifications-FP-1\8 -16-G-5/ 448 D009U01.

d. Watt meter - General Electric, Schenectady, NY., Model No. 3720341, Amp - 5/10, Volts-100/200, watts 500/1000/2000.

e. Multimeter - John Fluke Mfg. Co., inc., Everett, Washington, Model 8050A.

f. High Voltage Probe - John Fluke Mfg. Co., inc., Everett, Washington, Model 80K6

g. High Voltage Wiring - Anixter Chicago National , 8mm Silicone resistor core, Taylor Prowire.

h. Transformer - Jefferson Luminous Tube Transformer, Cat. No.721-411, Primary 120V, 60 Hz ,Secondary 15000V, 60 MA, Mid Point Grounded.

Description of Experimental Apparatus

Gas Handling System

Just one experimental gas (hydrogen sulfide) and nitrogen were piped into the system. However, the gas feed system can be used with slight modification to handle additional gases if necessary. The cylinders were located in a very well ventilated cell located outside the building for safety. The feed rate of the hydrogen sulfide stream was controlled with a simple metering valve on a rotameter.

Exhaust System

The gas streams flowed downward through the quartz reactor, where the gases dissociated when conditions were right for plasma formation. The main flow was passed through a glass wool filter and two bubblers of ethanol amine to remove unconverted hydrogen sulfide before being vented to the atmosphere. The solid sulfur deposited on the inside walls of the reactor was removed by dissolving in carbon disulfide. The solution was then left overnight in a hood for carbon disulfide to evaporate, thus recovering sulfur.

Reactor System

The SGDR used in this research is basically a combination of a capacitive and inductive device. It consists of two concentric glass cylinders, which form an annulus for gas flow. Silver paint or an inductive coil is wrapped around the outer glass cylinder. A second electrode is positioned inside the inner cylinder. When an electric potential is applied, the glass walls serve as as a dielectric causing the current to diffuse into the plasma or glow in the annulus. Gases flowing in the annulus pass through the plasma where plasma energy causes breakage of chemical bonds and subsequent reactions. The electrodes never come in contact with the gases during the process.

Initially the reactor was constructed of pyrex tubing. To provide leak proof connection between the steel piping and the glass reactor inlet and outlet ports, Teflon swage locks were used. During the first phase of experiments some runs were carried out with the reactor under pressure. The reactor made of Pyrex suffered some cracks due to localized arcing and subsequent overheating. After this failure, the reactor was made of quartz tubing for thermal stability. In the destructive tests Copper mesh was used as an inner electrode. Copper coil was wound around the outside of the reactor, which acted as the outer electrode. Specifications of different reactors used are given in table 1 below.

TABLE 1
SPECIFICATIONS OF REACTORS

Reactor	Outer dia of inner tube,cm	Inner dia of outer tube,cm	Annular gap cm	Length cms
1	1.50	2.26	0.380	25
2	1.70	2.85	0.575	24
3	1.80	2.78	0.490	61
Quartz used in destructive test	1.80	2.78	0.490	28

Energy System

The components of this system included an AC power supply, an oscillator for frequency tuning and a transformer to step up the voltage. A watt meter was used for power measurements. The watt meter was installed between the AC power supply equipment and the 110V wall supply. This watt meter measures the total power consumed by the whole set up. The power source and other electrical measurement equipment was inside the building.

Auxiliary Instrumentation

Additional equipment included a multimeter to record the secondary voltage applied to the reactor, a watt meter to record the power consumption and a rotameter to control the gas flow. The flow meter was calibrated for hydrogen sulfide under ambient pressure and temperature conditions. This was done experimentally with the help of a "bubble flow meter". The time taken for the bubble to flow the length of a 100 ml burette was clocked, thus giving the actual flow rate corresponding to the rotameter scale. A relation was then established by regressing the data points obtained.

Safety

Safety was a prime concern in the hydrogen sulfide experimental setup. Hydrogen sulfide is a colorless, highly flammable gas with an offensive odor. It is a toxic gas and is extremely poisonous in very small quantities. Concentration of 700 ppm by volume and above can result in acute poisoning. The odor of hydrogen sulfide in higher concentrations is objectionable. It will rapidly deaden the sense of smell, making it impossible to sense dangerous concentrations.

The complete experimental system, excluding the AC power source, the watt meter and multimeter was set up in the bay outside the building. The electrical equipment was connected to the reactor system by long cables running through the ports that connected the main building and the experimental cell outside. This enabled the experimental operation to be controlled from a remote place. This also ensured that the operator was not in danger of coming in contact with the high voltage wires.

The effluent from the plasma reactor went to the gas scrubber. The gas scrubber consisted of two bubblers containing ethanol amine, which reacted with hydrogen sulfide to produce a salt. The gases coming out of the scrubber were vented to the atmosphere.

CHAPTER IV

NON-DESTRUCTIVE TESTS

One of the objectives of this thesis was to study the physical and electrical characteristics of a SGDR. This was done using air as the test gas. By conducting these experiments key variables could be identified before performing the destructive tests.

Independent variables tested were primary voltage, frequency, pressure and electrode material. The dependent variable was secondary voltage. The experiments were conducted in a safety hood. Previous research [20,32] showed that for a given set of conditions, an optimum frequency existed which would give maximum power input to the reactor. Research presented in this chapter shows how this optimum condition is affected under different conditions. It is worthwhile to mention that variables like pressure and the type of electrode material used in this research were not subjects of previous work .

Procedure

An outline of the procedure followed in these experiments is given below,

1. After ensuring that all electrical connections were properly made, the inlet valve to the reactor was opened and the gas was allowed to flow through the reactor.
2. The power supply was turned on and the primary voltage was set at a desired value.

3. Frequency was varied from 60 Hz to 1000 Hz in steps of 10 Hz. Sufficient time was allowed (about 5 minutes)for the reading to stabilize before it was recorded.
4. Steps 2 and 3 were repeated for the same reactor at different primary voltages.
5. The whole procedure was repeated for several different reactors.

The graphs presented in figures 2 and 3 show only those points between which the plasma was visible. To ensure proper visibility all experiments were carried out in the dark.

Results and Discussion

The results are presented in a graphical form and discussed in the following sections. Experimental data with all fixed conditions are tabulated in Appendix B.

Secondary Voltage Dependence on Frequency and Primary voltage

Figures 2 and 3 show the relationship between secondary voltage and frequency at different primary voltages and for a particular reactor size. All tests were conducted with dry air. Each figure represents a constant primary voltage applied to the transformer. The secondary voltage initially increases with an increase in frequency. At a certain value of frequency for a particular primary voltage, secondary voltage goes through a maximum and then begins to decrease with increasing frequency. The peak is called the optimum condition.

The reason for this optimum condition is believed to be a result of the secondary electrical circuit loading on the transformer. Transformers are basically inductive devices

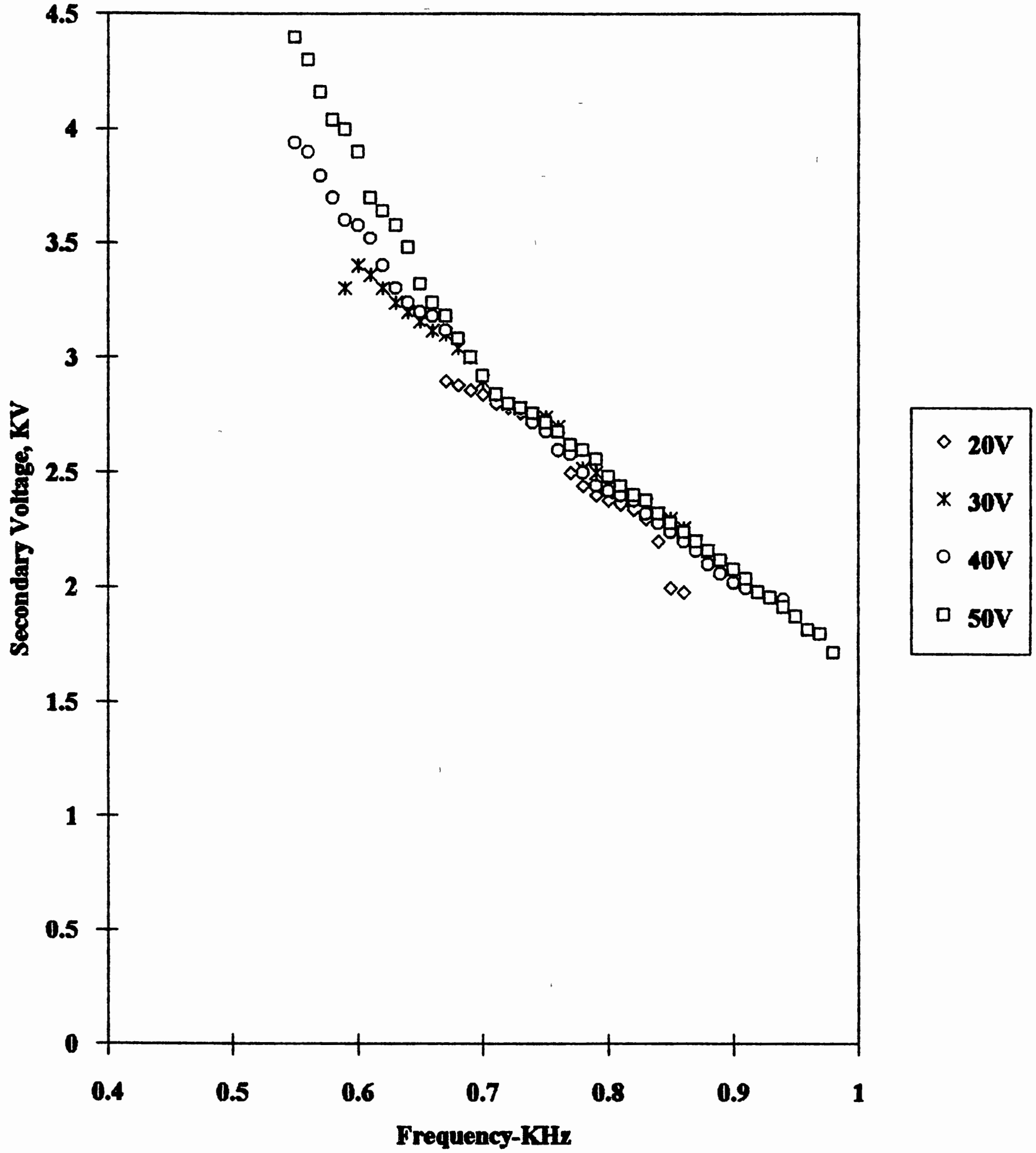

Figure2 . Dependence of Secondary Voltage on Frequency for Air at Fixed Primary Voltages for Reactor 1

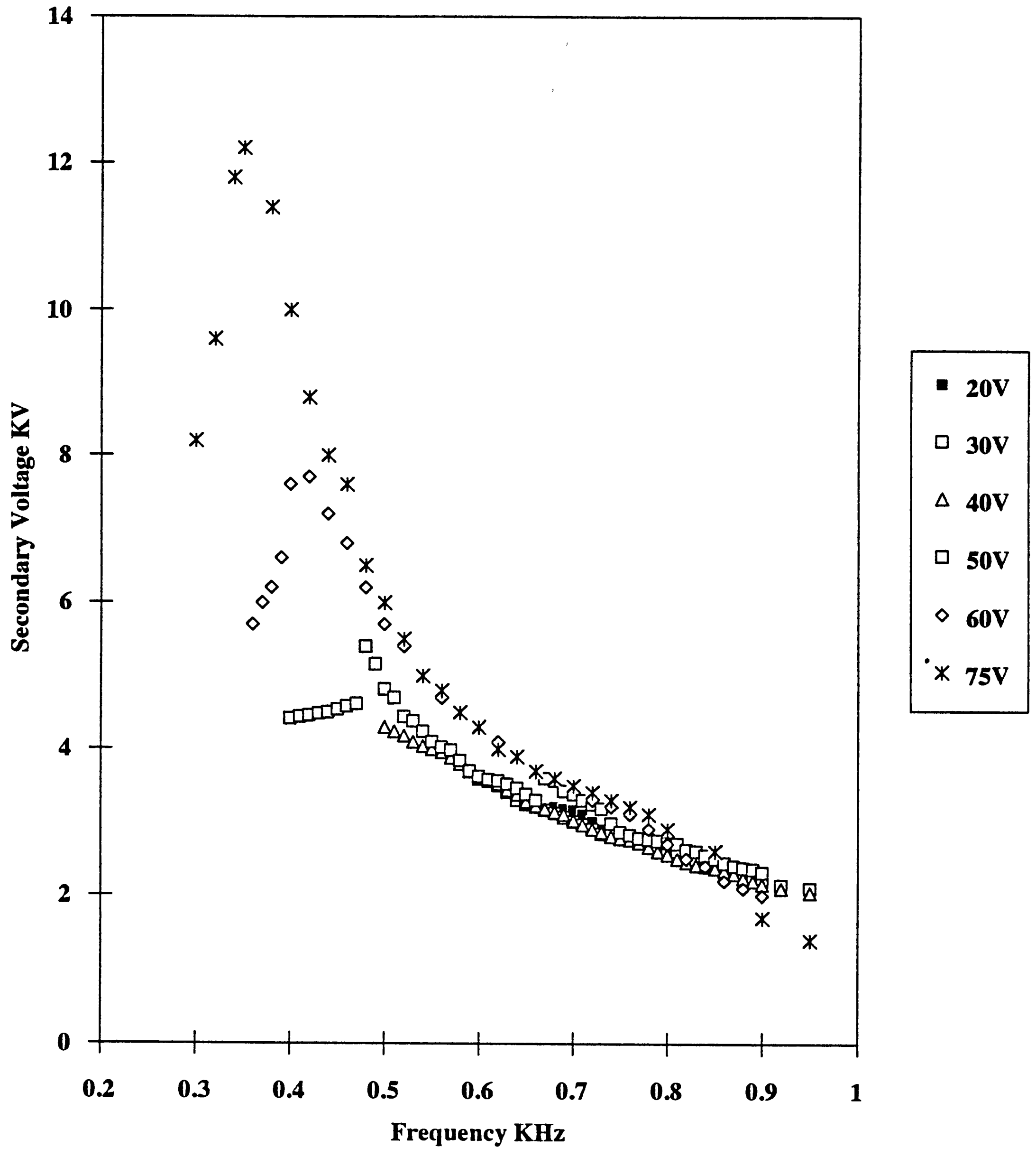

Figure 3 . Dependence of Secondary Voltage on Frequency for Air at Fixed Primary Voltages for Reactor 2

and consists of two sets of windings. When a capacitance generating circuit is connected to the secondary side of a transformer, a frequency exists where the capacitance of the circuit is equal to the inductance of the transformer. This condition is called resonance, and at this point secondary voltage goes through a maximum [4].

It was observed that as primary voltage increased, the value of maximum secondary voltage increased. The value of optimum frequency also decreases as primary voltage decreases. At the same time the range between which the plasma occurs also increases. Thus optimum frequency reveals the frequency range to be expected for operation of a particular capacitive discharge reactor system.

Visual observations revealed that the glow was more intense for reactor 1 than for reactor 2. Also, the jump in secondary voltage at the start of the plasma was large for reactor 1. It is likely that this is because of a smaller gas gap. Work presented in reference [30] shows that there is a critical diameter ratio, above which a direct discharge results. This ratio was reported to be 1.46. Reactor 2 used in this research has a diameter ratio slightly above 1.46.

It can be seen from the graphs, the value of secondary voltage at optimum frequency increases with increasing primary voltage. The same behavior was noticed with both types of reactors. These runs were performed at primary voltages ranging from 20V to 75V. The trend basically remains the same for all runs.

Effect of Changing Electrode Material and Reactor Size

Figure 4 shows the dependence of secondary voltage on frequency at a fixed primary voltage for a scaled up reactor with a silver nitrate inner and outer coating. This figure shows a rather unusual phenomenon. The secondary voltage is maximum at lower frequencies where the plasma is not existent. The frequency at which plasma becomes visible is at a higher value and not at the maximum secondary voltage. The reason for this is not clear at this time. The same phenomenon was observed at a different primary voltage (50V). The hysterisis was followed this time and the results are as shown in Figure 5. However, this observation is not believed to be caused by experimental error. The hysterisis study was done by changing the frequency in the reverse fashion once the forward cycle was completed. It was observed that the hysterysis does not retrace the path followed by the forward cycle.

A visual observation of plasma revealed that it was brighter and more uniform along the length of the reactor than that formed in the previous runs, where the reactors were coated with micro paint. A possible explanation to this could be that silver nitrate is a better conductor of electricity. Also the secondary voltages were higher at the same onditions when compared to the previous reactors. This could be because of bigger size of the reactor, which increased the electrode surface area and eventually lead to more consumption of power.

Secondary Voltage Dependence on Pressure , Primary Voltage and Frequency

Secondary voltage dependence on pressure and frequency for a fixed primary voltage is presented in Figure 6. The optimum frequency is attained at approximately the

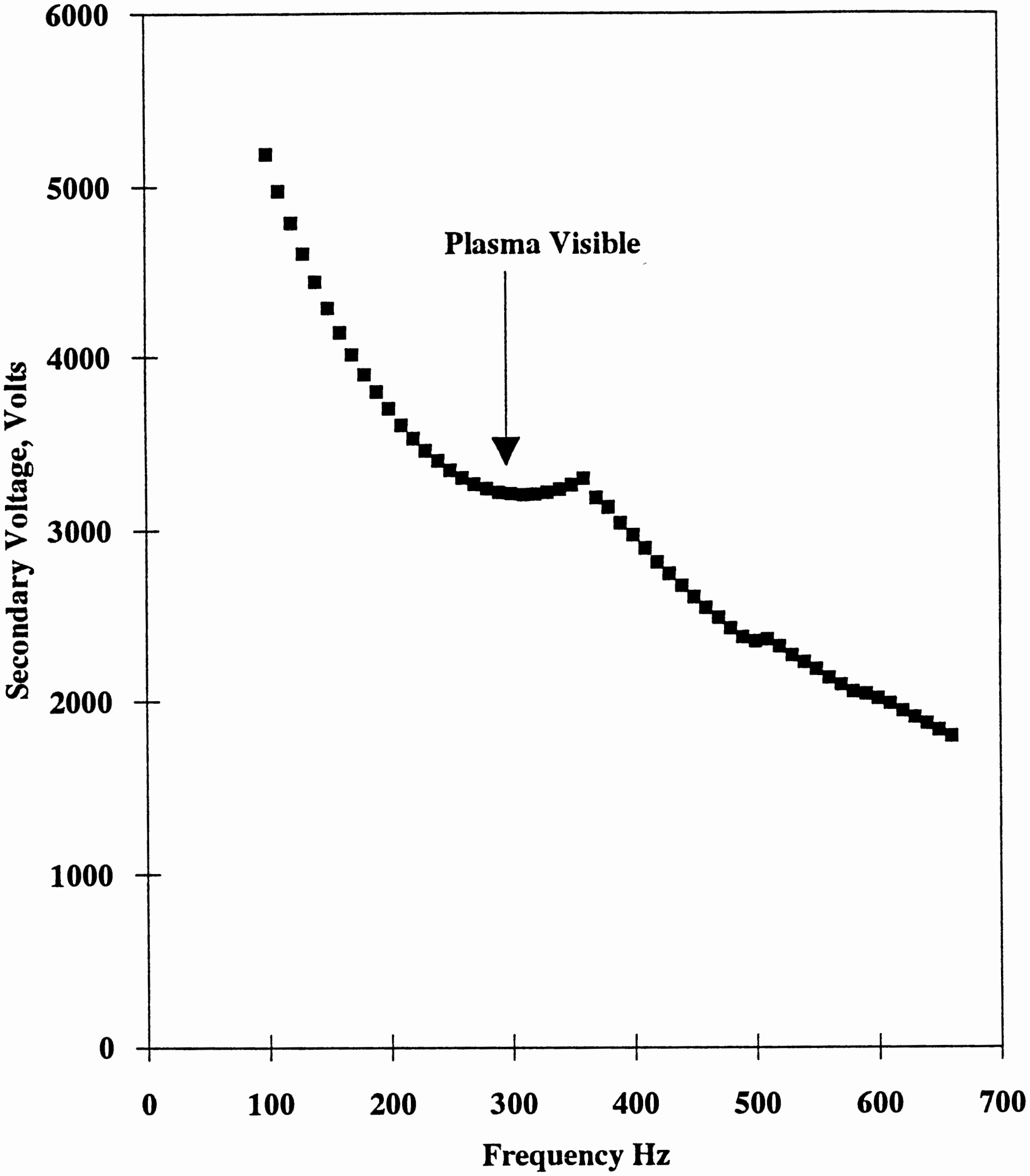

Figure 4 . Effect of Frequency on Secondary Voltage for Air at a Primary Voltage of 45 Volts for a Scaled Up Reactor (Reactor 3) Coated with Silver Nitrate

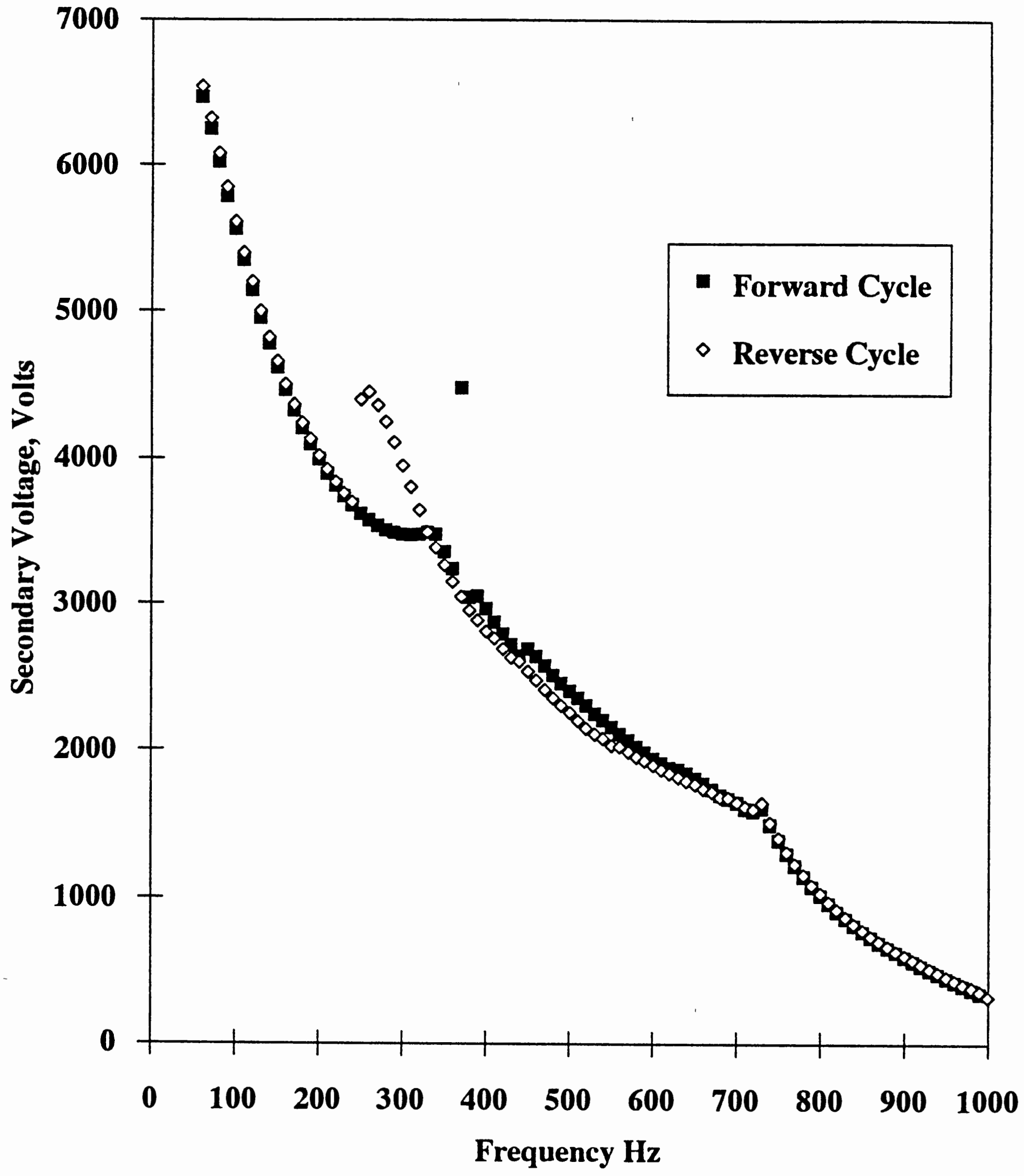

Figure 5 . Hysterysis Curve for Reactor 3 at a Primary Voltage of 50Volts, Using Air

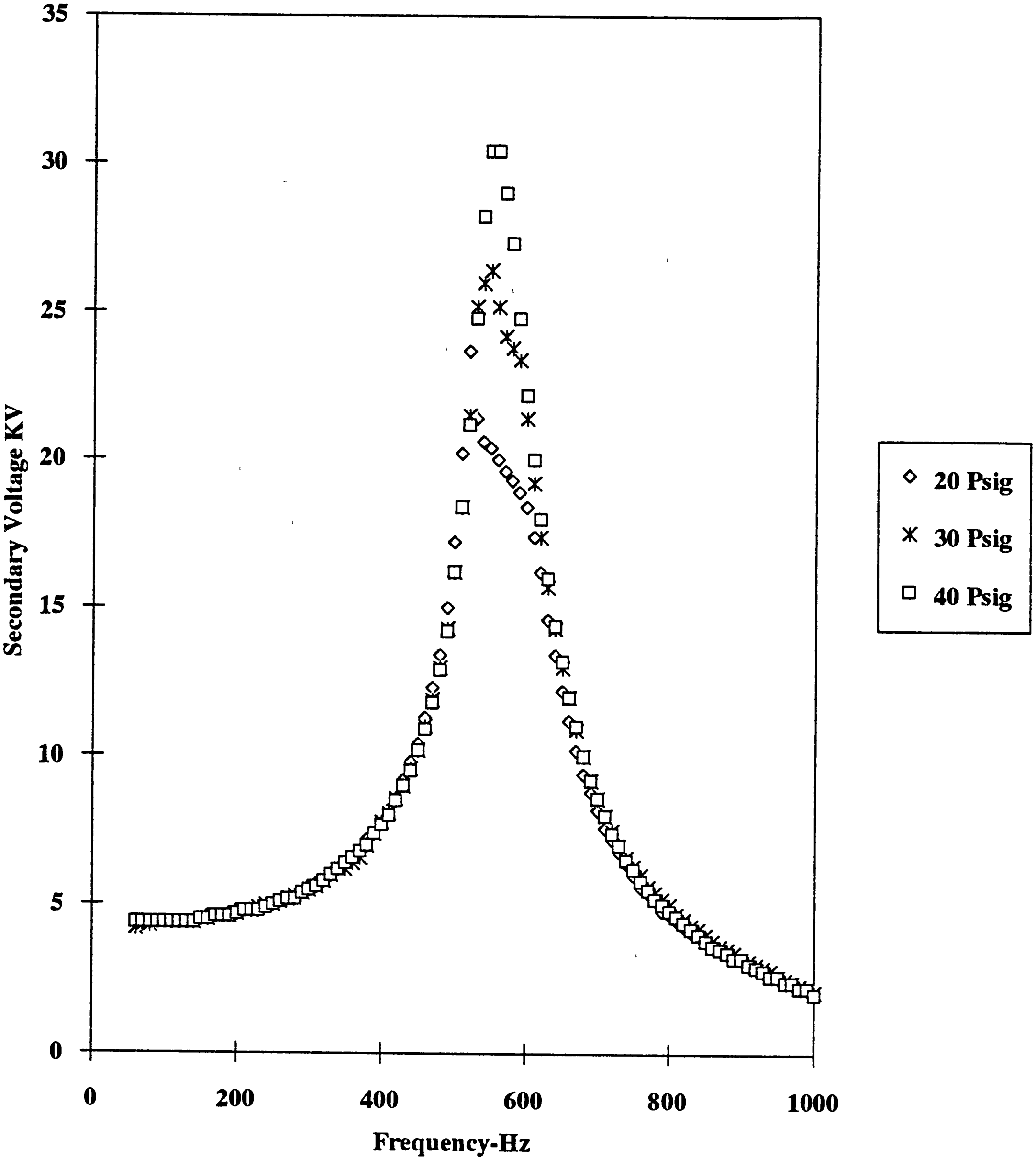

Figure 6 . Dependence of Secondary Voltage on Pressure and Frequency for Air at a Primary Voltage of 30 Volts for Reactor 3

same point for a given primary voltage, but the value of secondary voltage at this optimum frequency increases with an increase in pressure. An attempt to conduct these experiments at higher primary voltages was made but was not successful. Strong localized arcing was observed with subsequent overheating of the reactor. On removal of outer electrode coating and careful inspection of the surface of the reactor, cracks were observed on the outer wall. Also, the inner electrode coating was badly corroded.

Error Analysis

Experimental error of the non-destructive tests was determined using duplicate data runs. This was achieved by retaking several data points after a non -destructive run. Maximum deviation from the original reading was ±20 volts on the secondary voltage. Optimum frequency error was ±10 Hz.

CHAPTER V

DESTRUCTIVE TESTS

The purpose of these experiments was to determine the degree of conversion of hydrogen sulfide and how it is affected by primary voltage, frequency and flow rate. The experimental apparatus is shown in Figure 1.

The experimental procedure was divided into two parts. In the first part, for a fixed flow rate and primary voltage, the frequency at which deposition starts was noted and a range of frequency was selected to carry out specific destructive tests. The second part included the specific destructive tests.

Preliminary Destructive Tests

The procedure for this part is outlined below,

1. All electrical connections were checked for proper wiring.
2. Nitrogen was passed through the reactor for 10 minutes as a purge gas.
3. With nitrogen still flowing the reactor was switched on .With the primary voltage set at a desired value the reactor was run for at least 10 minutes so as to stabilize the plasma.
4. Nitrogen was turned off and H_2S was bled into the reactor slowly at a desired flow rate.

5. Frequency was now varied in steps of 10 Hz . The point at which deposition of sulfur would start was visually observed and noted . From here on , the experiment was continued untill sufficient data points were obtained. The variables involved in this data collection were secondary voltage and power. A constant visual monitoring was observed throughout the length of experiments.
6. The sulfur deposited was dissolved using carbon disulfide after the experiment was completed. The reactor was rinsed with acetone and then washed with water and dried before being used for the next run.
7. The above steps were repeated for different primary voltages at a fixed flow rate.
8. The flow rate was then changed and the whole procedure was repeated.

The data for this is tabulated in appendix C and the results are discussed below. Figures 7-10 show the variations of secondary voltage with frequency for a particular flow rate and for different primary voltages.

The basic characteristics remain the same as was found in our preliminary non-destructive tests. However, the optimum frequency appears to be spread over a wide range rather than occurring at a single point. This also gives an indication of the dependence of optimum frequency on the gas composition and hence the degree of ionization of a gas.

Table 2 gives an idea of the effect of flow rate and primary voltage variation on frequency at which destruction of hydrogen sulfide occurs. It appears that the change in flow rate at one primary voltage does not have any significant effect on the frequency at which destruction occurs. However, change in primary voltage at a particular flow rate seems to shift the frequency at which deposition of sulfur occurs.

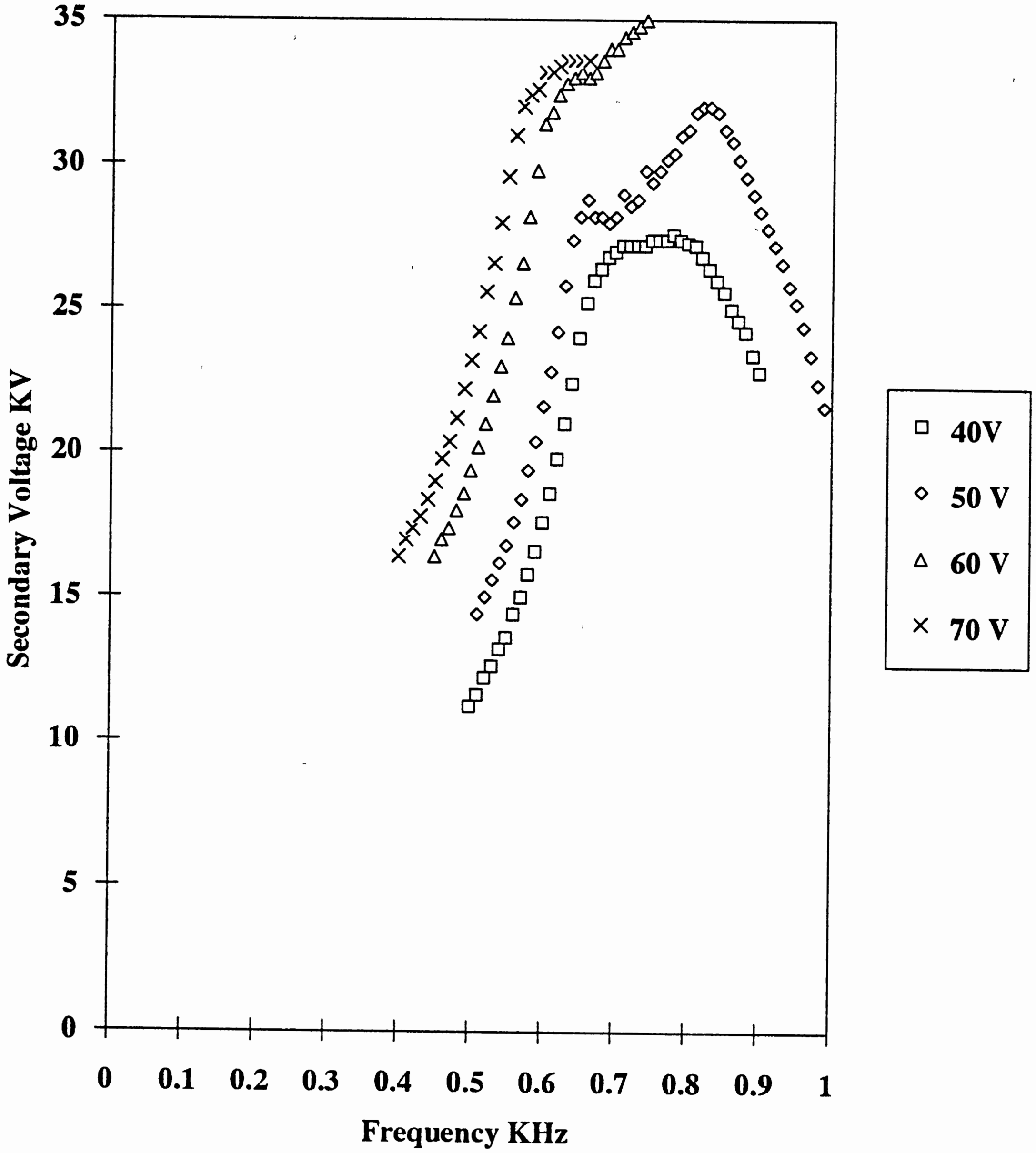

Figure 7 . Dependence of Secondary Voltage on Frequency for H_2S at Different Primary Voltages and a Flow Rate of 147 CC/MIN

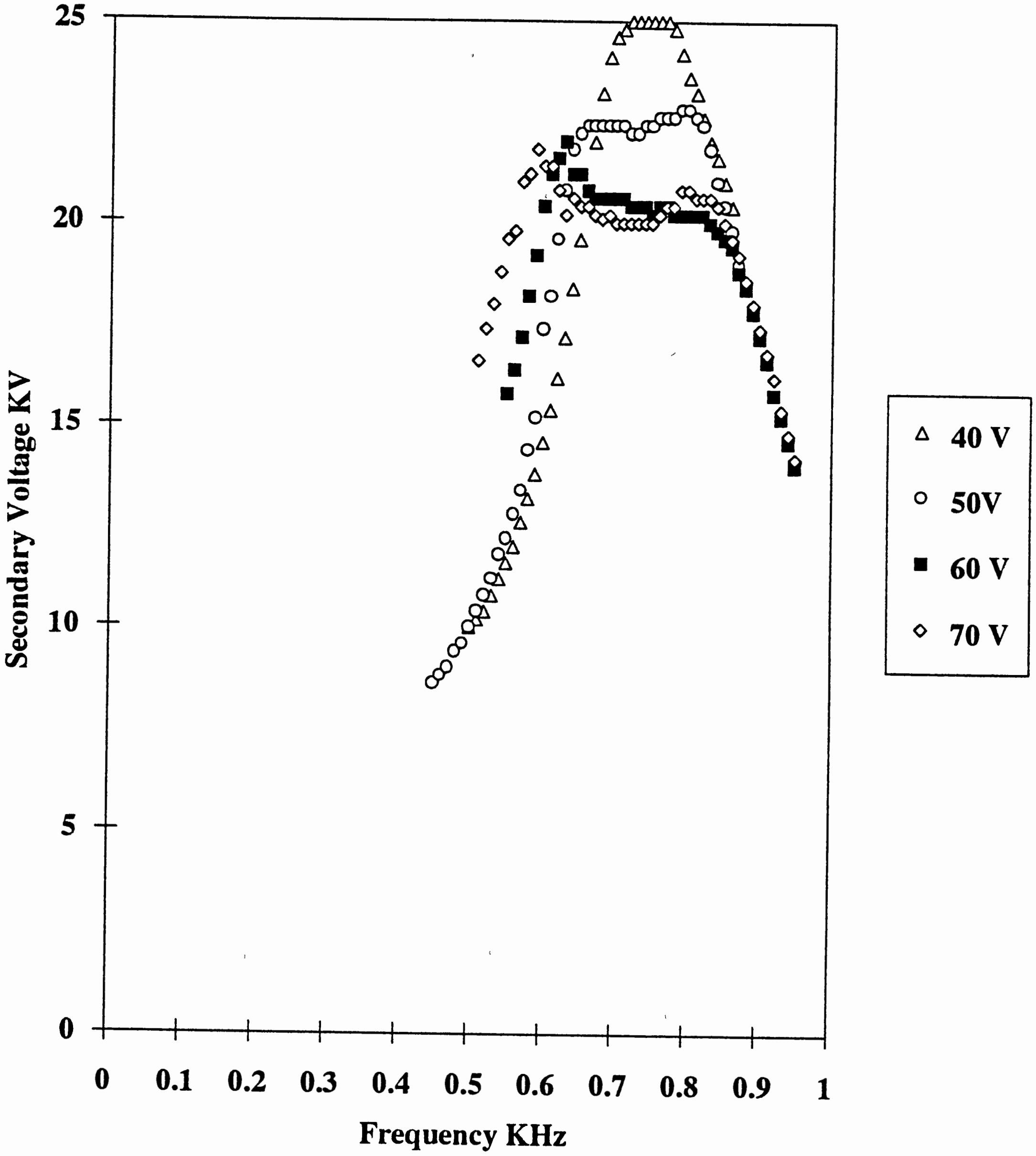

Figure 8 . Dependence of Secondary Voltage on Frequency for H_2S at Different Primary Voltages and a Flow Rate of 256 CC/MIN

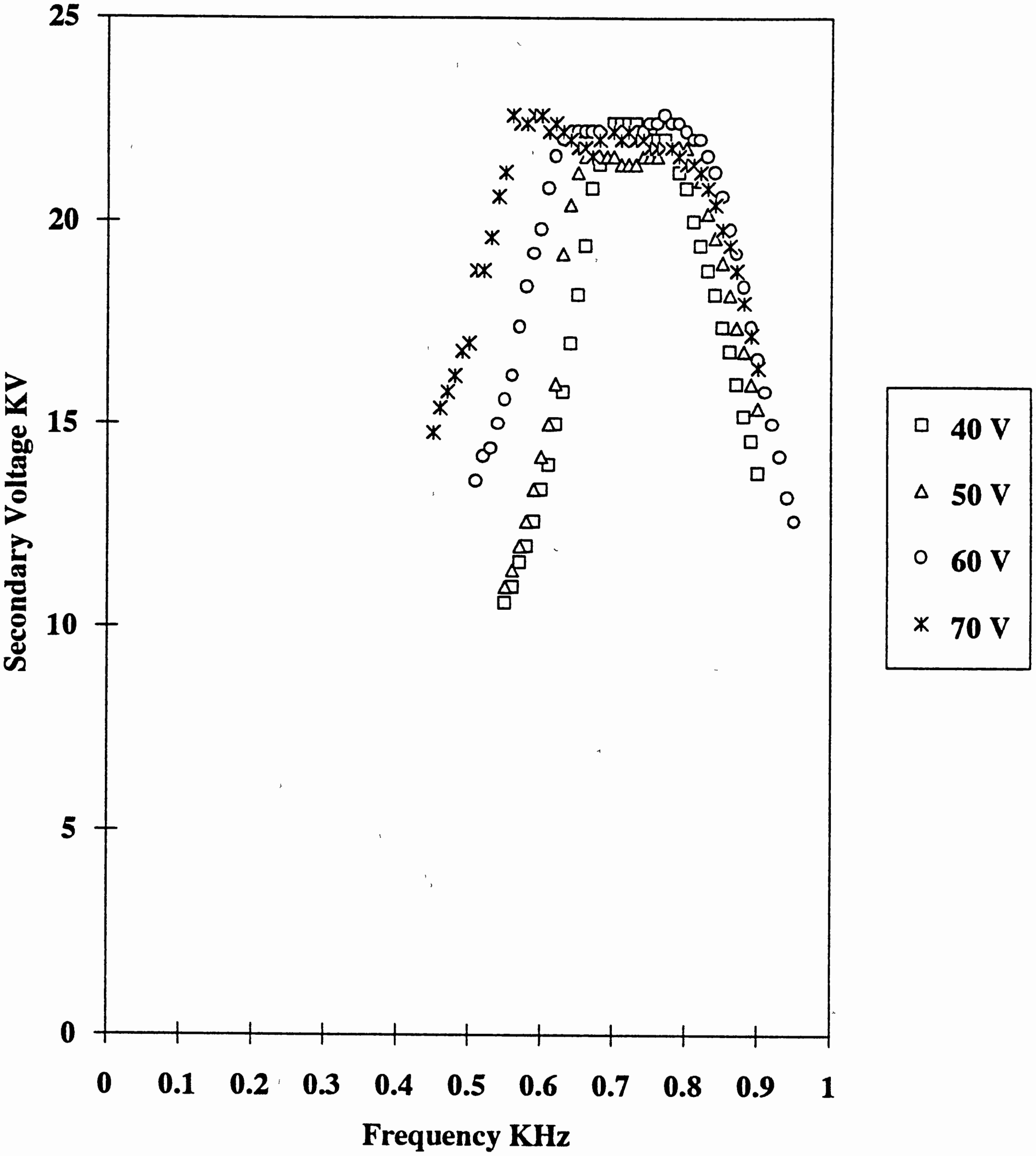

Figure 9 . Dependence of Secondary Voltage on Frequency for H_2S at Different Primary Voltages and a Flow Rate of 367 CC/MIN

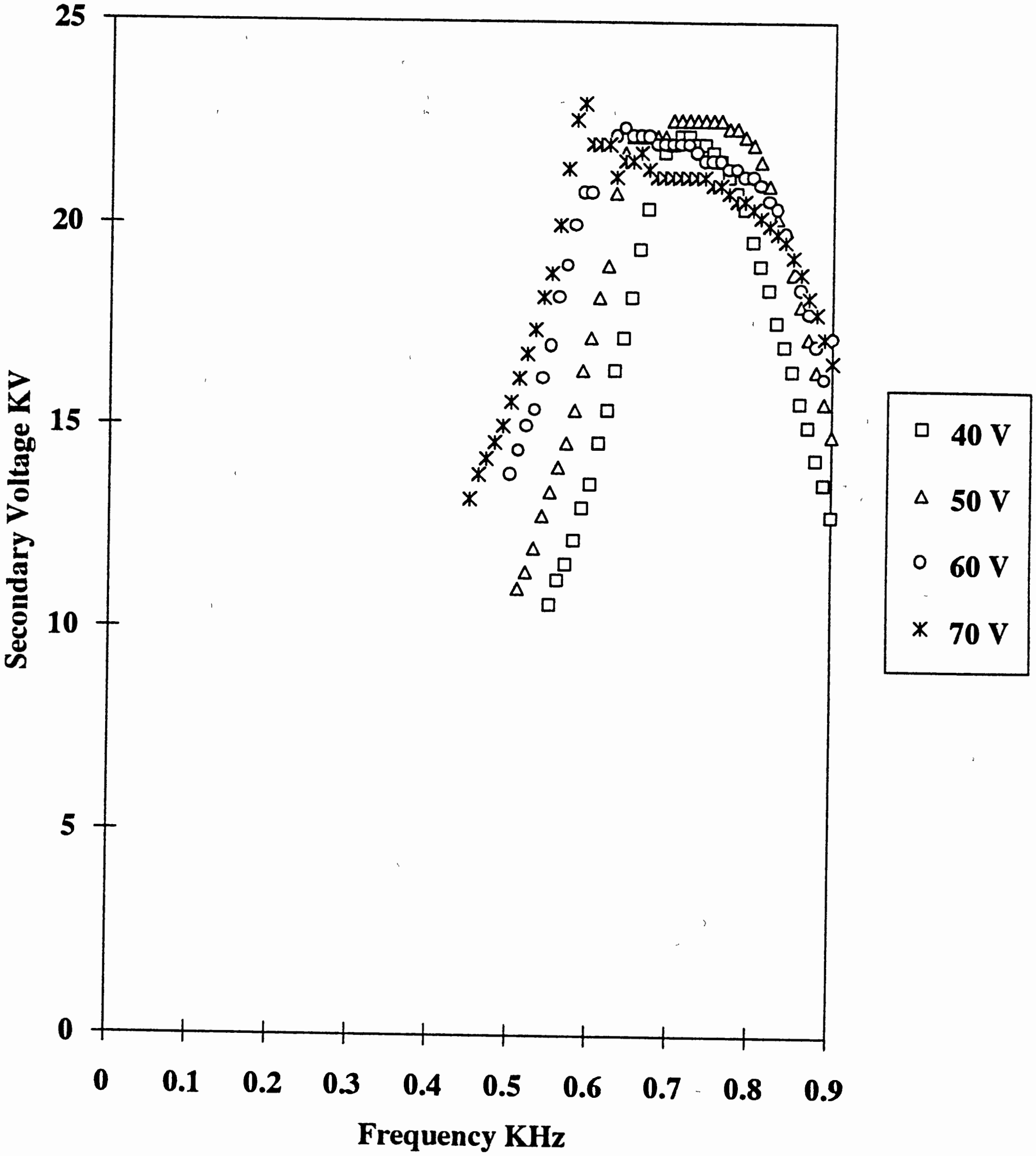

Figure 10 . Dependence of Secondary Voltage on Frequency for H_2S at Different Primary Voltages and a Flow Rate of 481 CC/MIN

TABLE 2

EFFECT OF FLOW RATE AND PRIMARY VOLTAGE ON FREQUENCY AT WHICH DEPOSITION OCCURS

	Frequency at which deposition occurred (Hz)			
	Flow rate (cc/min)			
Primary Voltage, volts	147.2	252.95	367.19	481.15
40	670	690	680	670
50	640	640	640	640
60	580	590	590	590
70	540	550	550	540

Actual Destructive Tests

The second part of the procedure involved the actual destructive tests and is outlined below,

1. The reactor was thoroughly cleaned and dried. Steps 1 to 4 of the previous procedure was carried out.
2. Depending on the flow rate some period of time was allowed to lapse for the nitrogen to pass out of the reactor before clocking the destructive run. Note that this involved visual observation of the plasma and the judgment of the author.
3. Toward the end of the run (10 minutes), secondary voltage, power input and atmospheric temperature were noted.
4. Exactly at the end of the desired period of time the reactor was switched off .
5. H_2S was shut off and nitrogen passed through in order to purge the reactor of H_2S.
6. The reactor was then dismantled and the sulfur deposits on the inside of the reactor and on the tubing were dissolved in carbon disulfide and carefully collected in previously weighed bottles.
7. The solution was then set in a hood for the solvent to evaporate leaving sulfur behind.
8. The reactor was washed clean with water and dried before the start of next run.
9. The above procedure was repeated for different frequencies for a particular flowrate and primary voltage.
10. The whole procedure was repeated for a different primary voltage.

The experimental data are tabulated in Appendix C. In the tables shown the first column gives the run numbers. Some run numbers are omitted as a result of failure of a particular run usually caused by leaking of the flow system or electrical problems. The second column gives the frequency at which the test was run and the next three columns give the process conditions. The last two columns give the amount of sulfur recovered and the calculated value of percentage H_2S conversion, respectively. The results of these experiments are presented in a graphical manner and are discussed in the sections below.

The maximum conversion achieved in all these experiments was 92%. The range of conversion obtained under various conditions was very wide (1% to 92%). The calculation of H_2S conversion was based on the ideal gas assumption. The amount of sulfur recovered is a key factor in these calculations. The details of the calculations are shown in Appendix A.

Dependence of H_2S Conversion on Frequency

All previous work reported has been at one particular frequency. Results of this section are presented in Figures 11-16. Fixed conditions were flow rate and primary voltage. The data points typified in these graphs appear to be scattered over a small range of conversion. It should be noted that from our preliminary studies on destructive tests, it was found that the optimum frequency was observed over a range and not at a single point. The findings here seem complimentary to that observed previously.

Dependence of H_2S conversion On Primary Voltage

Figure 17 shows a plot of percentage conversion versus primary voltage. Percent Conversion was taken as the average of the conversions obtained at different frequencies for a particular primary voltage. Average conversions obtained were higher at higher primary voltages. The data presented are for experiments conducted up to 100 V. Runs

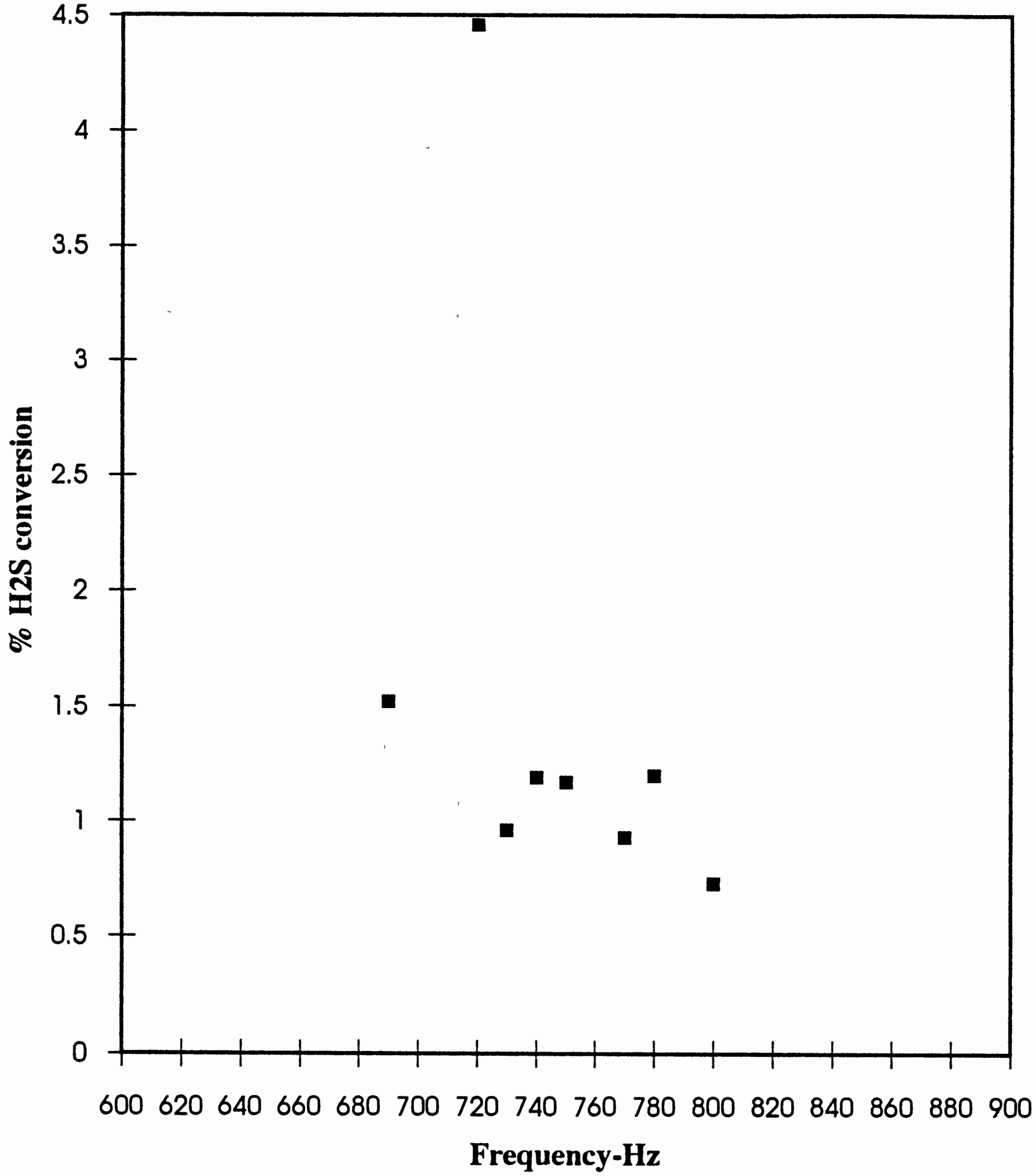

Figure 11 . Dependence of H_2S Conversion on Frequency at a Primary Voltage of 40 Volts and a Flow Rate of 147 CC/MIN

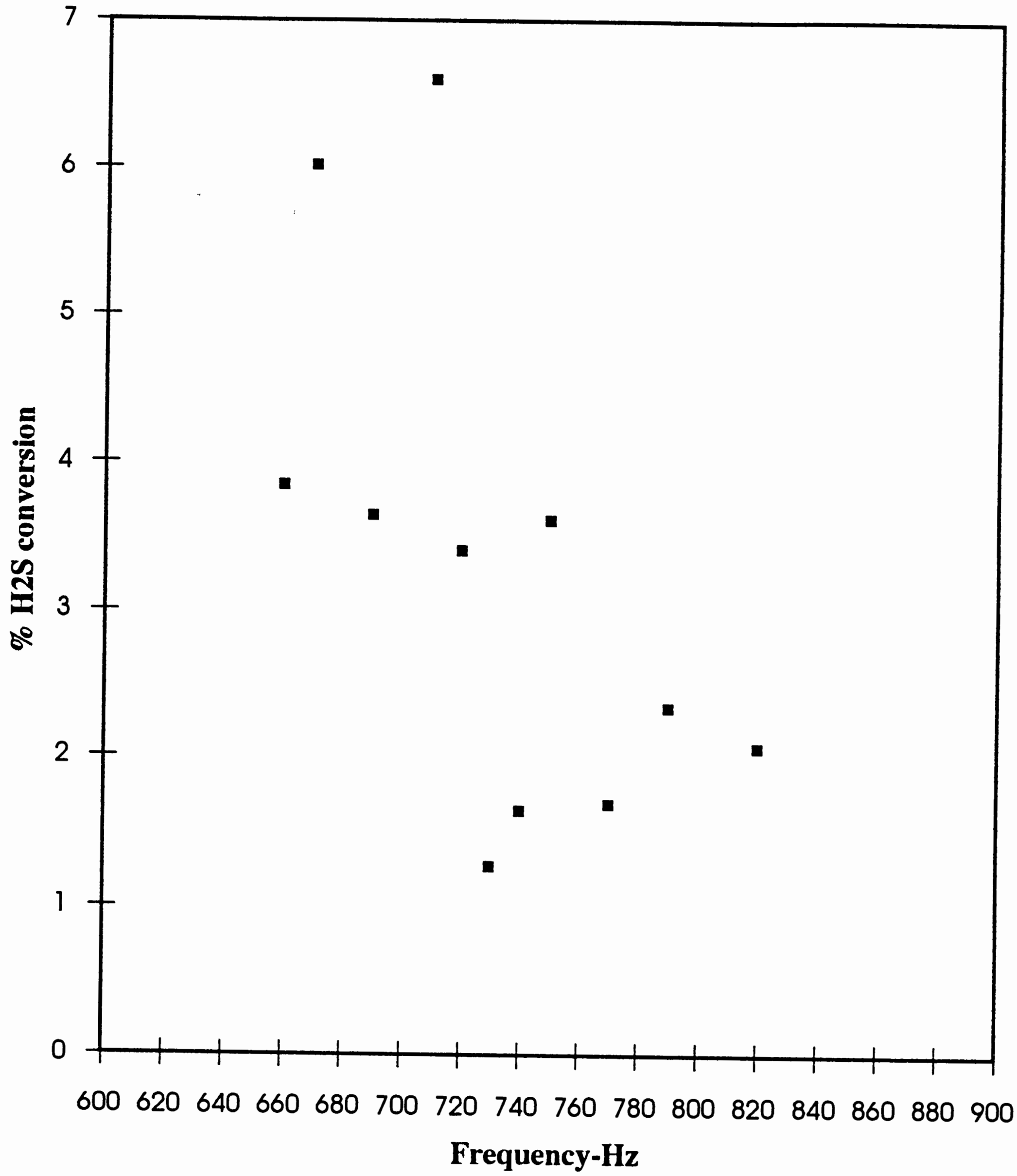

Figure 12 . Dependence of H_2S Conversion on Frequency at a Primary Voltage of 50 Volts and a Flow Rate of 147 CC/MIN

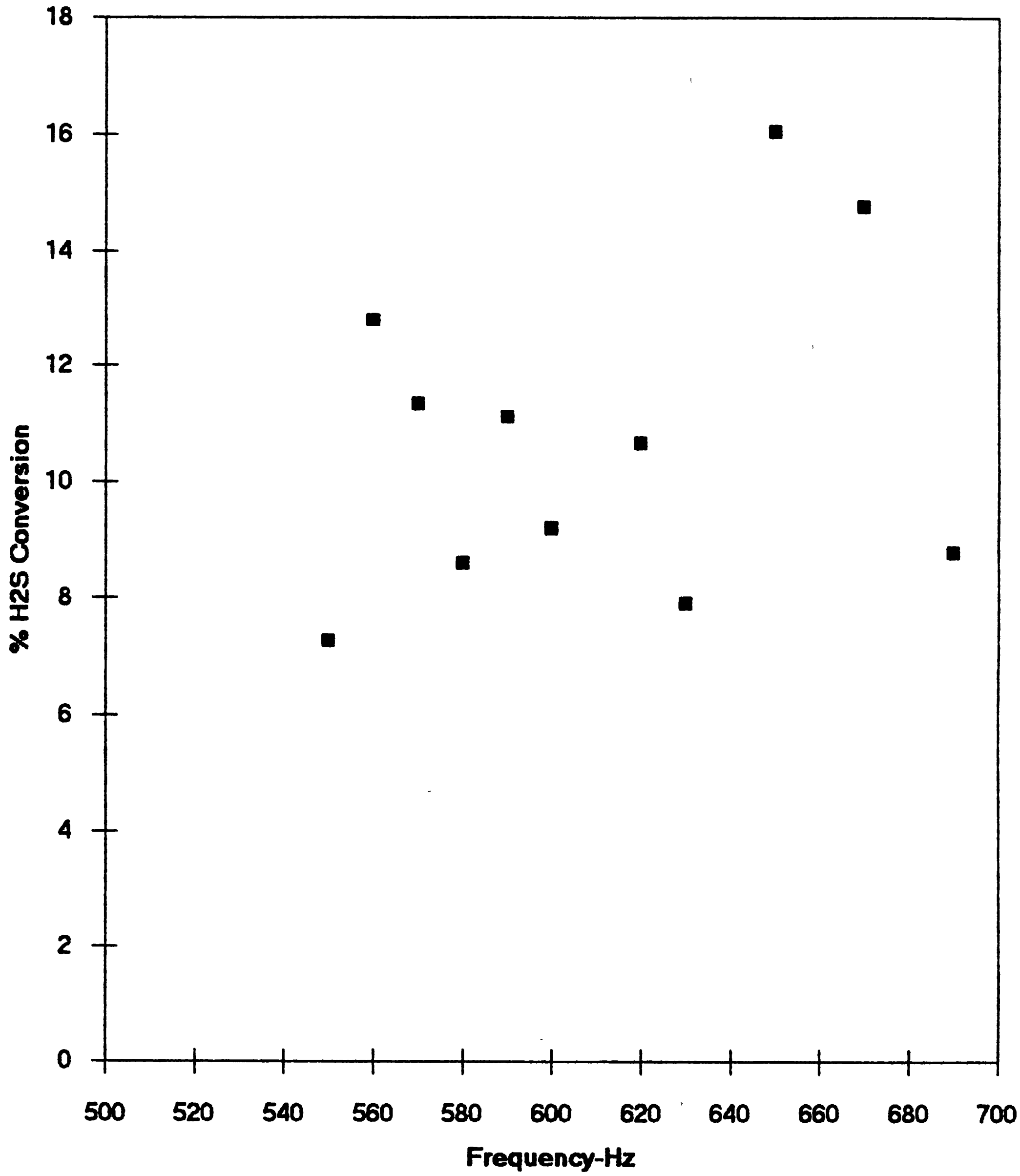

Figure 14 . Dependence of H_2S Conversion on Frequency at a Primary Voltage of 70 Volts and a Flow Rate of 147 CC/MIN

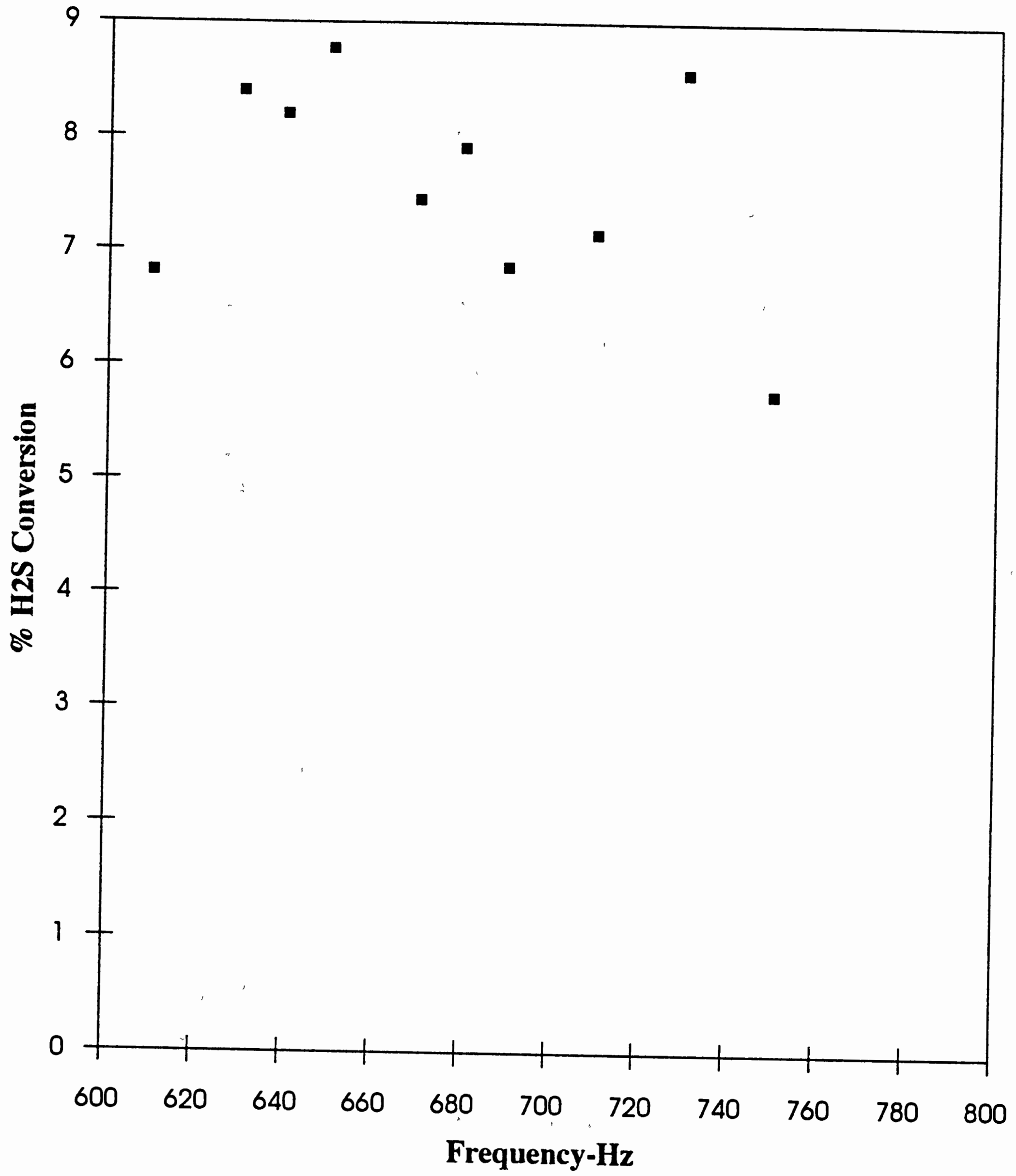

Figure 13 . Dependence of H_2S Conversion on Frequency at a Primary Voltage of 60 Volts and a Flow Rate of 147 CC/MIN

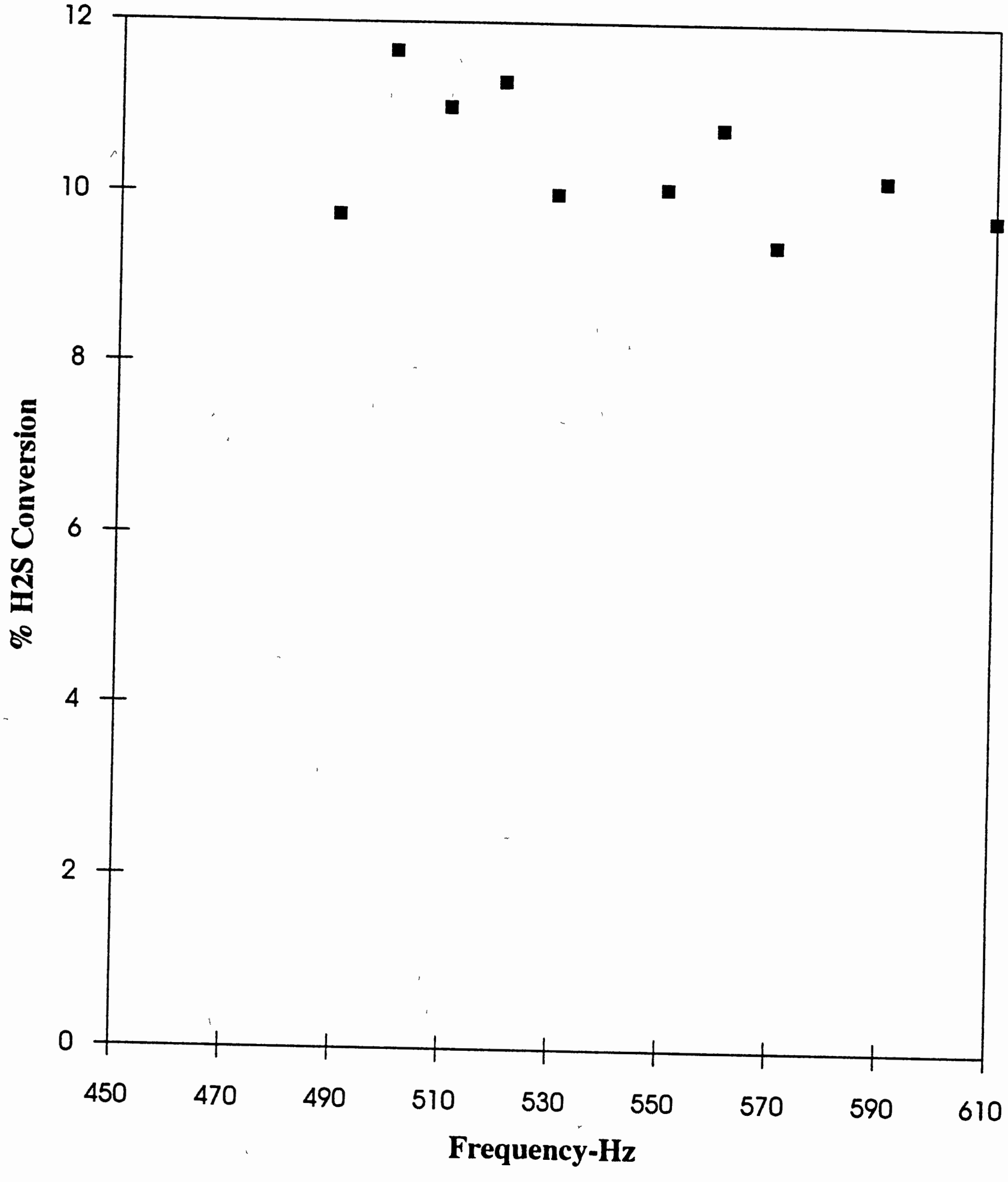

Figure 15 . Dependence of H_2S Conversion on Frequency at a Primary Voltage of 90 Volts and a Flow Rate of 147 CC/MIN

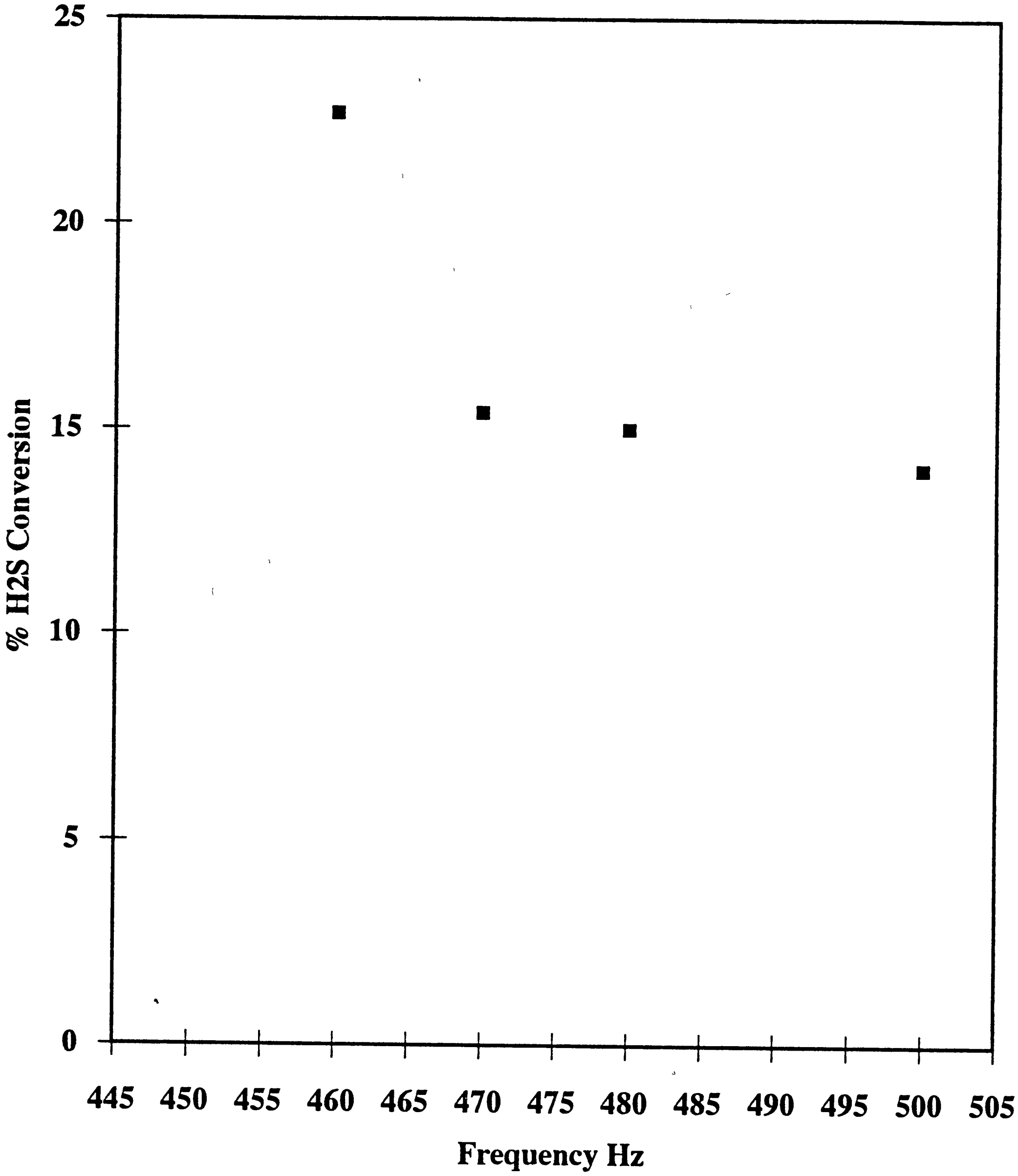

Figure 16 . Dependence of H_2S Conversion on Frequency at a Primary Voltage of 100 Volts and a Flow Rate of 147 CC/MIN

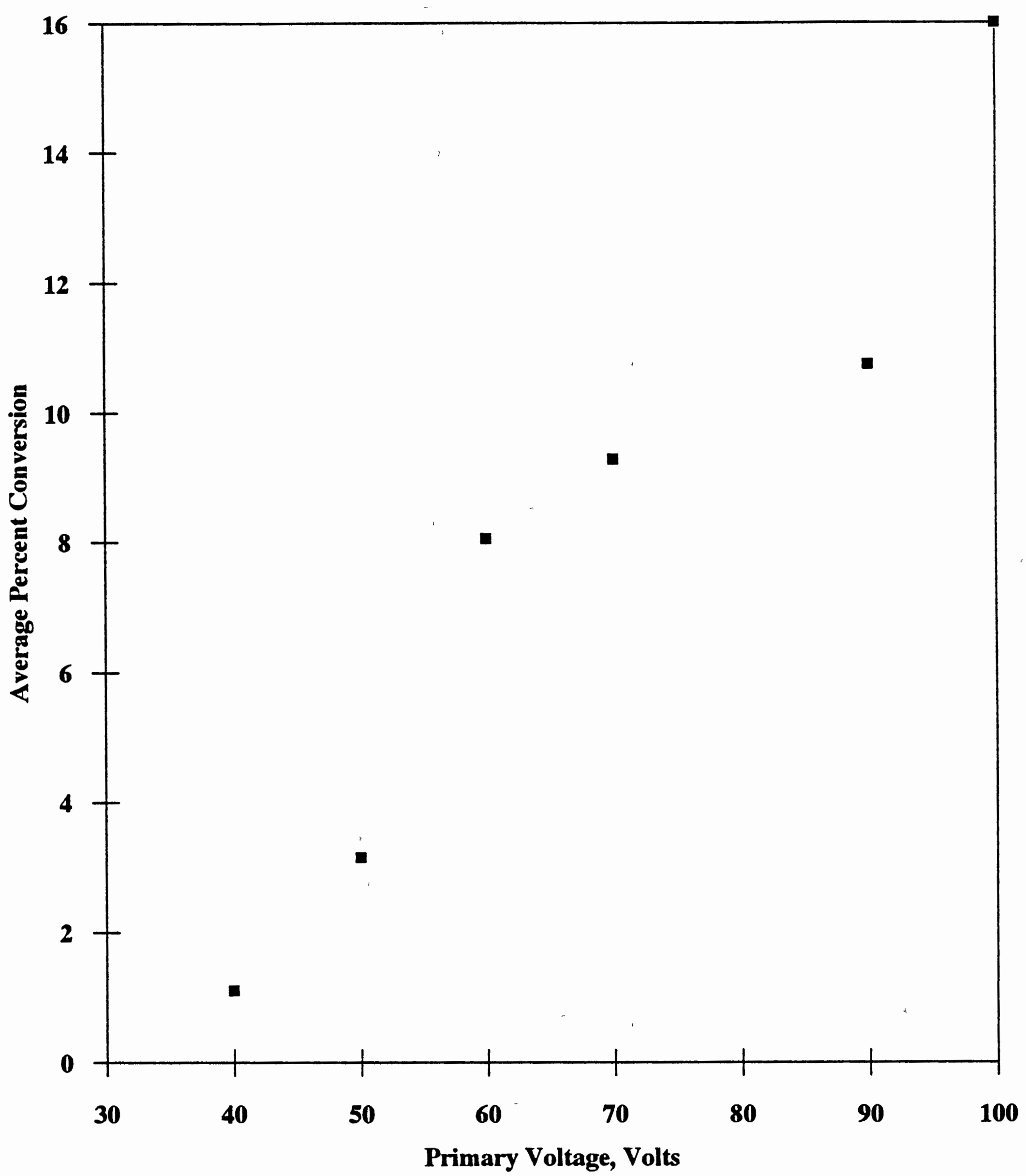

Figure 17 . Effect of Primary Voltage on H_2S Conversion at a Flow rate of 147.2cc/min

at even higher voltages were tried but were unsuccessful. At higher voltages the transformer output limit was overshot and the transformer tripped. Persistent tries lead to damage of the transformer. Subsequent experiments were conducted only at a primary voltage of 70 volts.

Van Drumpt [28] observed a similar phenomenon in his investigation on oxidation of hydrogen chloride. He concluded that conversion increased almost linearly with the applied voltage. However his experiments were conducted at a frequency of 50 Hz alone.

Dependence of H_2S Conversion on Flow Rate

Figure 18 shows the variation of H_2S conversion at different flowrates for a fixed frequency and a fixed voltage. Flow rate can have a very profound impact on H_2S destruction. Higher conversions were obtained at lower flow rates. Piatt [20] in his work on methane destruction observed a similar phenomenon. This trend was also observed by researchers at Argonne National Laboratory, where they studied H_2S destruction in a microwave plasma [12]. This phenomena is intriguing, especially because the general belief is that plasma reactions are instantaneous. However, these results seem to suggest that more time is required in the reaction zone for the larger molecules to break.

Reference [5], gives the suitable gas flow condition in glow discharge plasmas to be 10 cc/min. However the condition may not apply to all reactions. It should be noted that H_2S conversion of more than 90 % was obtained at a flow of about 9cc/min. It is not clear at this time, if the conversion is affected by the flow rate alone or in general just by increasing the residence time. Further investigation is necessary to shed some light on this.

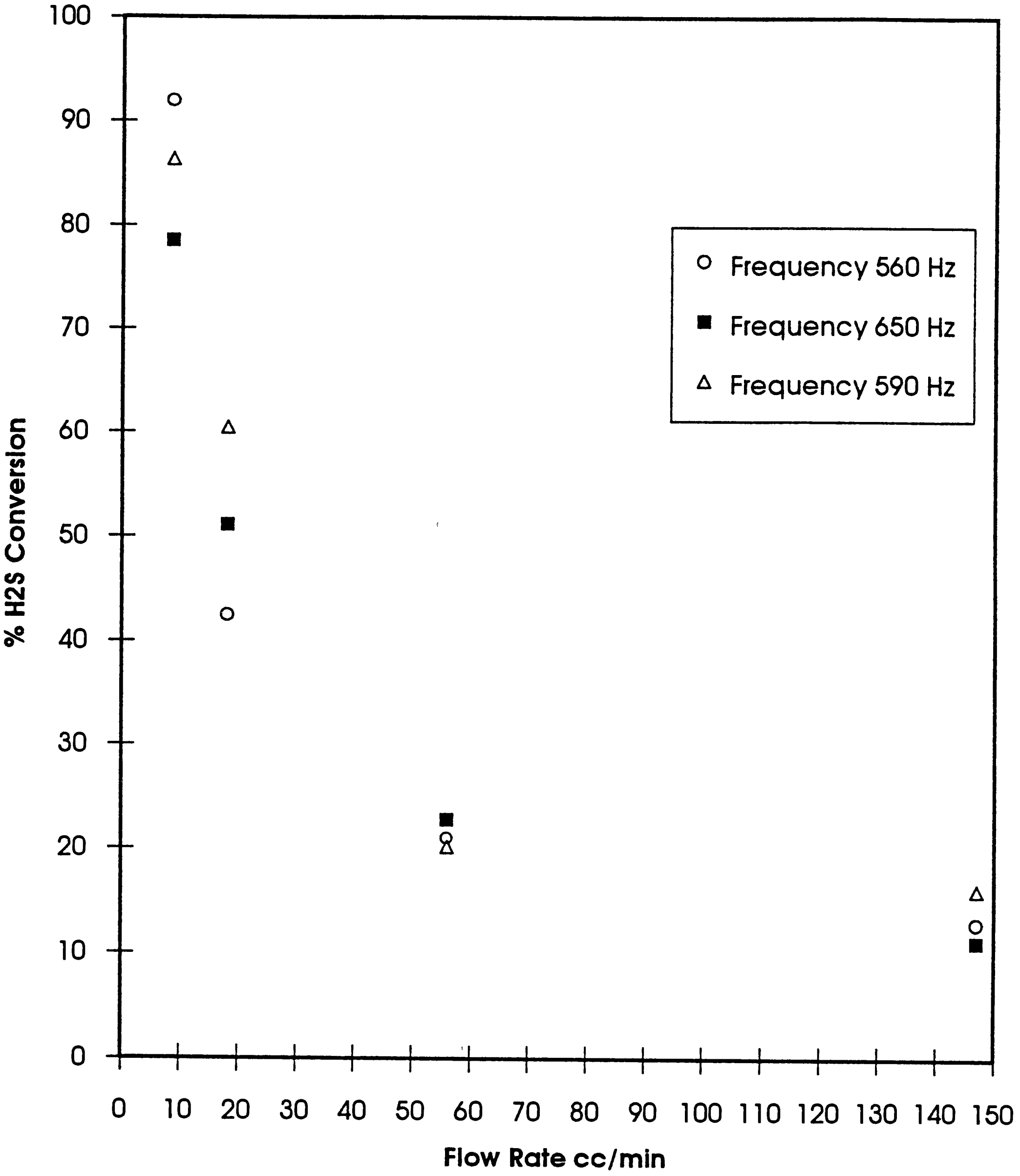

Figure 18 . Variation of H_2S Conversion on Flow Rate at a Fixed Primary Voltage of 70 Volts and at Different Fixed Frequency

Dependence of H_2S conversion on Duration of Experiment

The curve plotted in Figure 19 shows that conversion of H_2S is slightly larger for shorter duration of run than those at higher duration runs. This could be because of recombination of the sulfur formed over a period of time. The degree of recombination is probably smaller for runs of shorter runs.

Effect of Using Two Reactors In Series on H_2S Conversion.

The effect of using two reactors in series was studied by arranging two reactors in series with parallel electrical connections. The results of this run are tabulated below. Conversions obtained were about the same as that obtained for a single reactor for the same operating conditions. A possible explanation to this could be the division of power. Since the reactors were connected in parallel, the transformer would divide the output power equally between the two reactors and consequently conversions in both reactors would be less compared to a single reactor.

It should be noted that the two reactors used were of similar dimensions but of different material (quartz and Pyrex). The Pyrex reactor developed cracks during the second run itself and further investigation on this was abandoned due to the non-availability of a second quartz reactor.

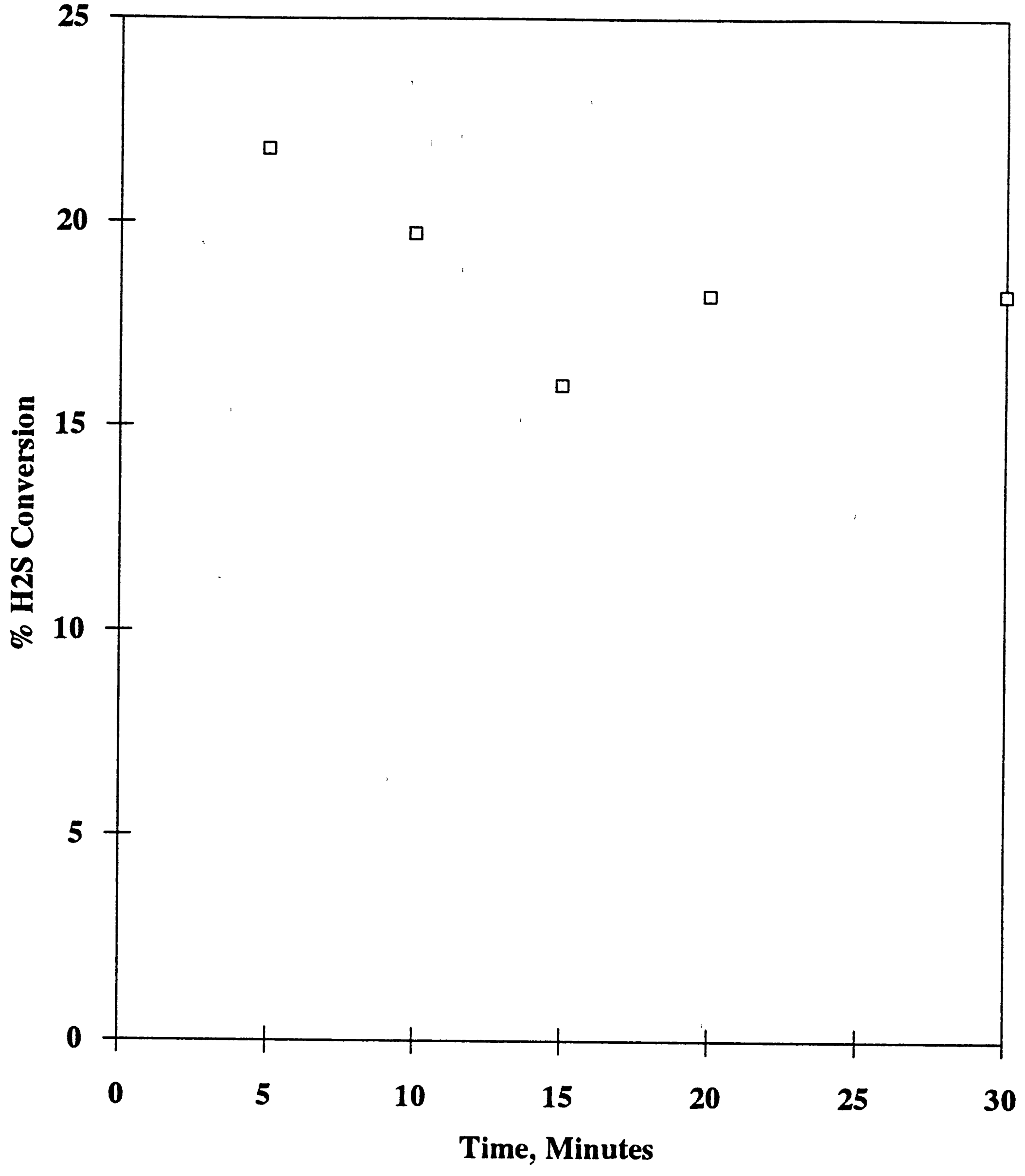

Figure 19 . Dependence of H_2S Conversion on Duration of Experiment at a primary Voltage of 70 Volts, a Flow Rate of 56 CC/MIN and a fixed Frequency of 590 Hz

TABLE 3

DATA OF H_2S CONVERSION USING TWO REACTORS IN SERIES

Reactors Used	Run #	Frequency Hz	Secondary **Voltage KV**	Power **Watts**	Temperature C	Amount of Sulfur	Percentage **Conversion**
2	98	560	6 3	101	30	0 1203	17
1	78	560	7 2	102	28	0 1273	17 5

Reproducibility

Determination of experimental error was accomplished with duplicate or triplicate data runs. The reproducibility data is presented in table 4 below.

TABLE 4.

REPRODUCIBILITY DATA

Run #	Frequency Hz	Primary voltage,volts	Flow Rate cc/min	Duration of run, mins	Percentage Conversion	Maximum Percentage Error
100	560	70	8.7	10	91.9	
103	560	70	8.7	10	88.7	3.5
101	590	70	8.7	10	82.3	
104	590	70	8.7	10	77.1	
107	590	70	8.7	10	78.47	5.6
99	650	70	8.7	10	86.4	
102	650	70	8.7	10	79	8.56
87	590	70	56	20	18.2	
95	590	70	56	20	17.1	6.6
88	590	70	56	30	18.24	
94	590	70	56	30	19.15	4.75
80	590	70	56	10	19.6	
85	590	70	56	10	19.73	0.65
92	650	70	18.18	5	18.2	
93	650	70	18.18	5	18.4	1.1

The above range of error seems to be realistic, subject to the experimental conditions. It should be noted that the reactor was located outside the building and drastic changes in weather conditions did prevail during the course of these experiments. Also, the electrical behavior of equipment like AC power supply, oscillator and more importantly the transformer play a important role. The state of the reactor walls due to charges existing because of previous runs is also believed to affect the electrical characteristics of the system. In view of the above, the author believes that a maximum relative error of + or - 10 % is very realistic for the equipment and analysis method used in these experiments.

Analysis of the Product material, Sulfur

Sulfur collected in the reactor appeared as a bright yellow, solid or film on the glassware. All of the material formed would dissolve in carbon disulfide instantly. Some of the recovered sulfur was subjected to X-ray diffraction studies at the Chemistry Department at OSU by Dr. Elizebeth. M. Holts. The study revealed that the sulfur formed was crystalline and of orthorhombic form. It was also inferred that the elemental sulfur formed was in the S_8 form.

CHAPTER VI

THERMODYNAMIC CONSIDERATIONS

Objective

A program developed by the NASA Center at Lewis was used to calculate equilibrium distribution for decomposition of hydrogen sulfide at various temperatures and atmospheric pressure. This was done initially to get an idea of the maximum attainable conversion at equilibrium and to be aware of the conditions at which it was possible. This eventually lead to the conception of effective temperature in the plasma.

Introduction

The principles applied to solution of all problems involving chemical equilibrium has been stated by Oliver et al. [24] as " that composition that satisfies the mass balance and total pressure specifications and also satisfies all the many simultaneous equilibria involved.

The suitability of modern computers for iterative procedures has accelerated the development of a number of practical techniques for calculation of complex product distributions. Chemical equilibrium is usually described by either of two equivalent formulations - equilibrium constants or minimization of free energy. The free energy minization technique is the one used by NASA scientist and is discussed in detail in Appendix E.

Inference

Figure 20, shows the variation of mole fraction of the pertinent species with respect to temperature at equilibrium. Complete decomposition of H_2S is achieved at a temperature of about 5100 K. A conversion of 91.9 %, which was the maximum attained in our project corresponds to a temperature of at least 3000 K. This only helps in telling that the effective temperature of the plasma was 3000K. More than anything else, it points out one of the important aspects of plasma technology. As already stated the thermal energy is substituted by the electrical energy and the idea of achieving the destruction at atmospheric pressure and temperature is shown to be possible.

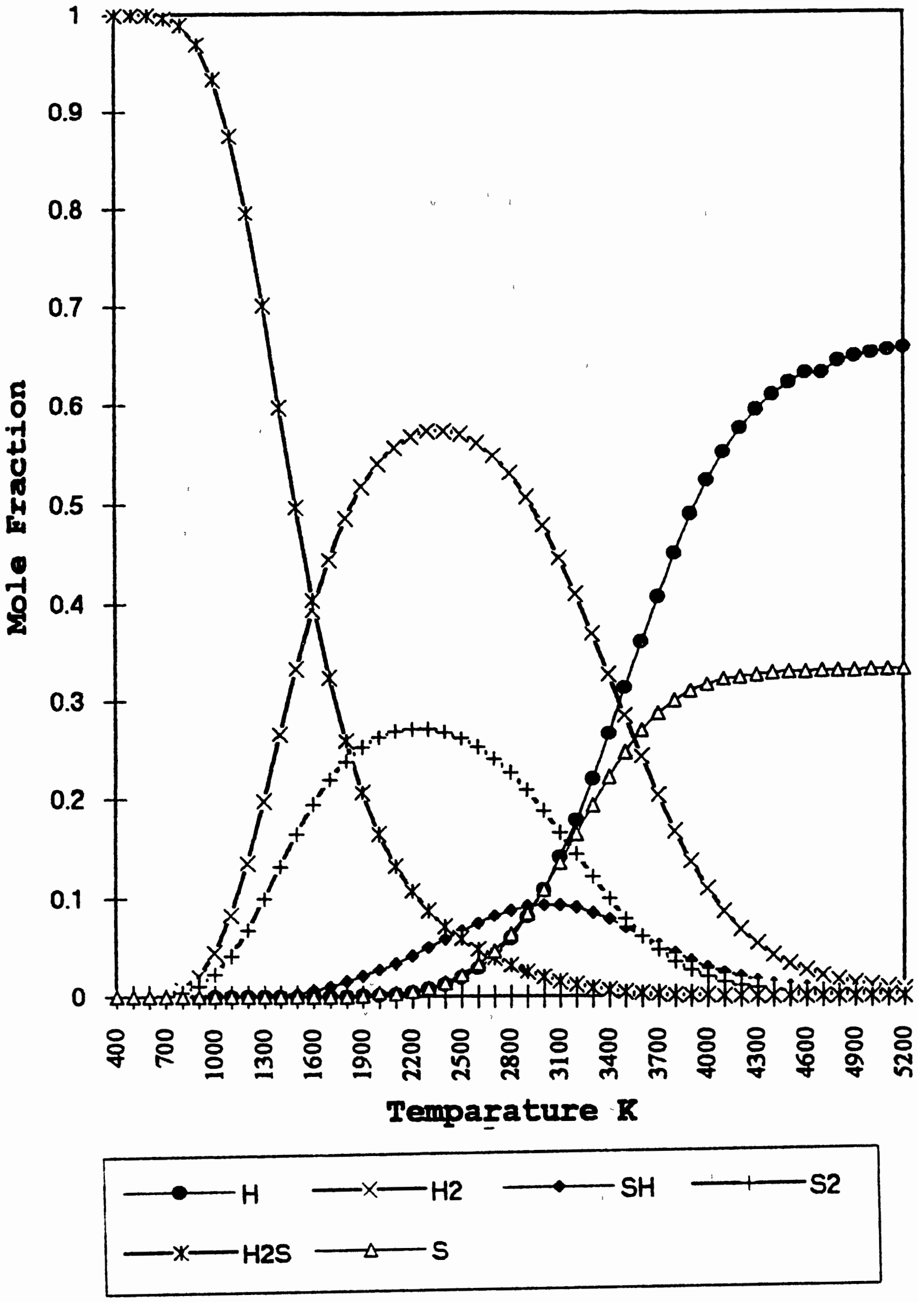

Figure 20 . Product Distribution for H_2S Decomposition Process at Various Temperatures

CHAPTER VII

CONCLUSIONS AND RECOMMENDATIONS

Research presented in this thesis was primarily exploratory and used to determine the feasibility of H_2S destruction in a Silent Glow Discharge Reactor (SGDR). Experiments were conducted to identify critical variables affecting the destruction process and to recommend areas for further investigation. Preliminary experiments were also conducted to learn the electrical characteristics of the equipment. These preliminary experiments were an expansion of previous work and basically conducted to get a better understanding of the plasma environment and to provide a good foundation for later work.

Conclusions

The conclusions from these experiments are as follows

1. An optimum frequency exists which yields a maximum secondary voltage and therefore maximum power input to the reactor. The optimum frequency is a function of primary voltage, gas composition, reactor size, electrode material and pressure condition.
2. Higher primary voltages gave higher secondary voltages and lower values of optimum frequency. The range of frequency over which a plasma occurs increases with increasing primary voltage.

3. Silver nitrate as an electrode seems to give a good plasma environment (based on visual observation), but is not wear resistant and at the same time costly. In the author's view, the use of coated surfaces is a disadvantage for the type of reactors used in this research. In addition any damage to the reactor walls or corrosion of inner electrode is not detectable immediately. Use of an inductive coil on the outside helps to avoid such handicaps and at the same time provides a visual observation of the process going on in the reactor.
4. The use of copper mesh as an inner electrode and a copper coil as an outside electrode seems to work very well. The reasons are,
    - No corrosion problems at higher voltages and temperatures,
    - Cheaper,
    - Plasma formed was uniform and intensity of glow was good on visual observation.
5. Study on scale up of a reactor was not definitive. However, it seems that the secondary voltage increases with increasing length of the reactor. From the studies of H_2S conversion dependence on flow rate, one can deduce that the use of a scaled up reactor would pave the way for achieving higher conversions at higher flow rates.
6. Reactors made of Pyrex glass are not suitable to be used as plasma reactors. This was evident by the fact that every reactor used in this research developed cracks due to thermal instability. It should be noted that the walls of the reactor heat up during the operation of a plasma.
7. With increase in pressure at a particular primary voltage, the value of optimum secondary voltage increases while the optimum frequency shifts only slightly.
8. For a particular primary voltage, the change in flow rate does change the frequency at which H_2S destruction starts. However, at a particular flow rate, for change in primary voltages, the frequency at which H_2S destruction starts, shifts. In effect it appears that the destruction starts only after a certain level of input power is reached.

9. For a set of fixed conditions, H_2S destruction is achieved over a wide range. However at frequencies over a certain range the degree of variation on the overall conversion achieved is not large.
10. H_2S conversion increases with decrease in flow rate
11. Finally, the use of Alternating Current Frequency Tuned Reactor in the destruction of hydrogen sulfide, under the right conditions seems feasible. Feasible at least to the extent of 92% conversion !

Recommendations

1. The experimental apparatus used in this research need refinement. Discussions with the manufacturers indicated that accuracy of the electrical measurement devices wanes when not at 60 Hz. Electrical equipment and measuring devices (transformer, watt meter, high voltage probe, multi meter etc) that can function efficiently at higher frequencies are required.
2. The decomposition reaction considered in this research involved only pure H_2S. In practice, of course, H_2S sources available for processing contain other components such as carbon dioxide, ammonia, carbonyl sulfide, nitrogen etc. The effect of these other components on the process requires careful consideration.
3. The effect of residence time on the reaction requires further investigation.
4. Research to provide insight into the kinetics and mechanism of the process is certainly warranted.
5. Research is needed to determine energy requirements for scale up of plasma reactors.

6. Intensive research is needed in developing a reactor design equation and a numerical computer simulation model that can be used to optimize and predict hydrogen production and to design large scale commercial reactors.
7. All plasma processes at the moment are operated on empirical knowledge. Development of models to describe the phenomena at hand are required to understand and quantify the process. Many models have been developed which include kinetic expression, fluid and concentration profiles etc. However, they are system dependent i.e. the parameters determined from experiments and the model fit each other for that particular reactor geometry. What is needed, are models which relate process variables to process parameters and conditions and which can phenomenologically describe the process taking place for any reactor geometry, temperature and pressure conditions. To do this it probably requires the identification of dominant mechanisms.
8. With the necessary models at hand one can consider on line controls of the plasma process as a potential research topic.
9. Finally research will be needed in the direction of establishing a possible commercial process for decomposition of hydrogen sulfide into hydrogen and sulfur in an alternating current, frequency tuned plasma reactor.

BIBLIOGRAPHY

1. Darwent B. and Roberts R. , The Photochemical and Thermal Decompositions of Hydrogen Sulfide. Proc. Roy. Soc. (London); 1953; 216A: 344

2. Balebanov, A.V.; Butylin, B. A.; Zhivotov, V.K.; Krokvenko, V.I.; Matolich, R.M.; Macheret, S. S.; Novikov, G. I.; Potapkin, B. V.; Rusanov, V.D., USSR. Dissociation of Hydrogen Sulfide in Plasma. Dokl. Akad. Nauk SSSR; 1985; 283: 657-60 [Phys. Chem.] 76.

3. Balebanov, A.V.; Zhivotov, V.K.; Krasheninnikov, E. G.; Nester, S. A.; Nester, S. A.; Potapkin, B. V.; Rusanov, V. D.; Samarin, A. E.; Fridman, A. A; Shulakova, E.V., USSR. Hydrogen Sulfide Dissociation in the Mixture with Propane in Microwave Discharge. Khim. Vys. Energ; 1989; 23: 440-3 67.

4. Bedell, F., The Principles of the Transformer. New York: Macmillan Company; 1986.

5. Boenig Herman V. Fundamentals of Plasma Chemistry and Technology: Technomic Publication Company; 1988.

6. Blyholder C. D. and Cagle G.W. Environ., Infrared Spectra of H_2S, CS_2, SO_2, CH_3SH, C_2H_5SH Adsorbed on Feand Ni. Sci. and Tech.; 1971; 5(2) 158

7. Computer Program for Calculation of Complex Chemical Equilibrium Compositions and Applicatiopns. Sanford Gordon, Bonnie J. McBride and Frank J. Zeleznik: Lewis Research Center; October 1984

8. Czernichowski, A.; Jorgenson, P.; Lesueur, H.; Chapelle, J.; Meguernes, K. Complete Decomposition and Vaporization of Hydrogen Sulfide by a Plasma - chemical Procedure. Colloq. Phys.; 1990; 51.

9. Huyton D. W. and Woodward T. W., The Effect of Elecron Scanvengers on the Gama Radiolysis of Gaseous Hydrogen Sulfide. Can. J. Chem.; 1970; 48: 2300.

10. Zelezenik F.J. and Gordon S. Calculation of Complex Chemical Equilibria. Ind. Eng. Chem.; 1968; 60(6): 27.

11. Johannes A.H. (Oklahoma State University). Personal Communication.

12. Harkness, John B.L.; Gorski, Anthony J.; Daniels, Edward J., Argonne, IL 60439, USA. Hydrogen Sulfide Waste Treatment by Microwave Plasma Dissociation. Proc. Intersoc. Energy Convers. Eng. Conf; 1990; 25th: 197-202 49, 51.

13. Besten I. E. and Selwood P. W., The Chemisorption of Hydrogen Sulfide, Methyl Sulfide and Cyclohexene on Supported Nickel Catalysts. Journal of catalysis; 1962; 1: 93.

14. Gorski A. J., Daniels E.J. and Harkness J.B.L., Treatment of Hydrogen Sulfide Waste Gas. Energy systems Divisions, Argonne National Laboratory; 1990

15. Maruzen Oil Company Limited, Japan; July 9. (1965); 14413.

16. Raymont, M. E. D., The Thermal Decomposition of Hydrogen Sulfide. Calgary, Alberta: The University of Calgary; 1974.

17. Gratzel M., Visible Light Cleaves Hydrogen Sulfide. Chem Engg. News; July 27, 1981; 59(30): 40.

18. Nester, S. A.; Rusanov, V. D.; Fridman, A. A., USSR. Dissociation of Hydrogen Sulfide in Plasma with Low Oxygen Content, Khim. Vys. Energ; 1988; 22: 461-4 52

19.Schmitt P.D., Oxidation Promoted by Ultrasonic Radiation. J. Am. Chem. Soc.; 1929; 51: 370.

20. Piatt, M. A., Methane Destruction in an Alternating Current Plasma Reactor, Oklahoma State University, Stillwater: 1988.

21. Potapkin, B. V.; Rusanov, V. D.; Strelkova, M. I.; Fridman,A. A., USSR.Hydrogen Sulfide Dissociation Kinetics in Thermal Plasma. Khim. Vys. Energ; 1988; 22: 537-40 76.

22. . Potapkin, B. V.; Rusanov, V. D.; Strelkova, M. I.; Fridman,A. A., USSR. Influence of Oxygen Additions on the Kinetics of Hydrogen Sulfide Dissociation in Thermal Plasma. Khim. Vys. Energ; 1990 24: 156-61.

23. Schwarz R. and Kunzer W., Influence of the Silent Electrical Discharge Upon Hydrogen Sulfide. Z. Anorg. Allgem. Chem.; 1929; 183: 287

24. Oliver R.C., Stephanou S. E. and Baier R. W., Calculating Free Energy Minimization. Chem. Eng. Eng.; Feb 19 (1962): P.121.

25. Torrey, R. M. P., Diss. Abs. Int. B.; 1970; 30(8): 3599.

26. Rusanov, V. D.; Fridman, A. A.; Macheret, S. O., Moscow, USSR. Effect of Spatial Non-equilibrium Behaviour During the Dissociation of Hydrogen Sulfide in a Nonuniform Plasma.Doll. Akad. Nauk SSSR; 1985; 283: 590-4 [Phys.].

27. Teimurova, F. A.; Rasulov, A. M.; Klimov, N. T., USSR. Dissociation of Hydrogen Sulfide in Barrier Discharge Plasma. Khim. Vys. Energ; 1991; 25: 378-9 67.

28.Van Drumpt, J. D., Oxidation of Hydrogen Chloride with Molecular Oxygen in a Silent Electrical Discharge. Ind. Eng. Chem Fundam.; 1972; 11: 594-5

29. Vastola, F. J.; Stacy W. O., University Park, Pa., USA. The Plasma-induced Reaction of Hydrogen Sulfide with Hydrocarbons. Am. Chem. Soc., Div. Fuel Chem., Prepr; 1967; 11: 234-7.

30. Tsai, Ven Yen., Conceptual Design and Performance Analysis of Frequency Tuned Capacitive Discharge Reactors. Oklahoma State University, Stillwater: 1990

31. White, W. B.; Johnson S. E. and Dantzig, G. B., Chemical Equilibrium in Complex Mixtures. J. Chem. Phys.; 1958; 28(5): 751.

32. Thomas W. J. and Ullam U. Chemisorption of Hydrogen Sulfide and Carbon Disulfide on Sulfided Nickel Oxide and Vanadium Pentoxide. J.Catalysis; 1969; 15(4): 342.

APPENDIX A

SAMPLE CALCULATION

Sample calculations for run 100 are as follows:

Experimental Data :

Amount of sulphur collected = 0.1043 grams
Flow Rate = 8.7 cc/min
Duration of Run = 10 minutes
Pressure = 1 atm
Temperature = 299 K

Assumptions :

1. Ideal gas. Therefore the equation PV=nRT, applies.
2. overall stoichiometric equation is

$$H_2S \text{---}> H_2 + S$$

R, the gas constant = 0.0821 (atm-liter)/(K-Mole)

H_2S going in = 0.0087 liters/minute

$$\text{therefore, moles of } H_2S \text{ going in} = \frac{1 \text{ atm} \times 0.0087 \text{ liters/min} \times 10 \text{ mins}}{0.0821 \text{ atm-liter/mole-K} \times 299 \text{ K}}$$

$$= 0.00354 \text{ moles}$$

Moles of sulphur collected = (0.1043 grams) / (32.02 grams/mole)

= 0.00325 moles

H_2S going out would be = 0.00354 - 0.00325

= 0.00029 moles

$$\text{Percentage Conversion} = \frac{\text{moles } H_2S_{\text{in}} - \text{moles } H_2S \text{ out}}{\text{moles } H_2S_{\text{in}}} \times 100$$

$$= (0.00354 - 0.00029) / (0.00354) \times 100$$

$$= 91.90\ \%$$

APPENDIX-B

NON-DESTRUCTIVE TEST DATA

TABLE 5

NON-DESTRUCTIVE TEST DATA CORRESPONDING TO FIGURE 2
RUN 1

Reactor :1 Electrode : Silver paint

Primary Voltage : 20 V

Type of Gas : Air

Frequency - Hz	Secondary Voltage - KV	Frequency - Hz	Secondary Voltage - KV
670	2.90	770	2.50
680	2.88	780	2.44
690	2.86	790	2.40
700	2.84	800	2.38
710	2.80	810	2.36
720	2.78	820	2.34
730	2.76	830	2.30
740	2.74	840	2.20
750	2.70	850	2.00
760	2.60	860	1.98

TABLE 6

NON-DESTRUCTIVE TEST DATA CORRESPONDING TO FIGURE 2
RUN 2

Reactor : 1 Electrode : Silver Paint

Primary Voltage : 30V

Type of Gas : Air

Frequency - Hz	Secondary Voltage - KV	Frequency - Hz	Secondary Voltage - KV
600	3.40	680	3.04
610	3.36	690	3.00
620	3.30	700	2.90
630	3.24	710	2.84
640	3.20	720	2.80
650	3.16	730	2.78
660	3.12	740	2.78
670	3.10	750	2.76

TABLE 6 (CONTINUED)

Frequency - Hz	Secondary Voltage - KV	Frequency - Hz	Secondary Voltage - KV
760	2.70	830	2.36
770	2.60	840	2.32
780	2.52	850	2.30
790	2.50	860	2.26
800	2.44	860	2.20
810	2.40	880	2.16
820	2.38	890	2.10

TABLE 7

NON-DESTRUCTIVE TEST DATA CORRESPONDING TO FIGURE 2
RUN 3

Reactor : 1 Electrode : Silver Paint

Primary Voltage : 40 V

Type of Gas : Air

Frequency - Hz	Secondary Voltage - KV	Frequency - Hz	Secondary Voltage - KV
520	4.00	740	2.72
550	3.98	750	2.68
560	3.96	760	2.60
570	3.80	770	2.58
580	3.70	780	2.50
590	3.60	790	2.44
600	3.58	800	2.42
610	3.52	810	2.40
620	3.40	820	2.38
630	3.30	830	2.32
640	3.24	840	2.28
650	3.20	850	2.24
660	3.18	860	2.20
670	3.12	870	2.16
680	3.08	880	2.10
690	3.00	890	2.06
700	2.92	900	2.02
710	2.84	910	2.00
720	2.80	920	1.98
730	2.78		

TABLE 8

NON-DESTRUCTIVE TEST DATA CORRESPONDING TO FIGURE 2
RUN 4

Reactor : 1 Electrode : Silver Paint

Primary Voltage : 50V

Type of Gas : Air

Frequency - Hz	Secondary Voltage	Frequency - Hz	Secondary Voltage
430	5.20	630	3.58
440	5.36	640	3.48
450	5.58	650	3.32
460	5.58	660	3.24
470	5.52	670	3.18
480	5.28	680	3.14
490	5.20	690	3.00
500	5.12	700	2.92
510	4.90	710	2.84
520	4.80	720	2.80
530	4.64	730	2.78
540	4.50	740	2.76
550	4.40	750	2.72
560	4.30	760	2.68
570	4.16	770	2.62
580	4.04	780	2.60
590	4.00	790	2.56
600	3.90	800	2.48
610	3.70	850	2.48
620	3.64	900	2.08

TABLE 9

NON-DESTRUCTIVE TEST DATA CORRESPONDING TO FIGURE 3
RUN 5

Reactor : 2 Electrode : Silver Paint

Primary Voltage : 20V

Gas : Air

Frequency - Hz	Secondary Voltage - KV	Frequency - Hz	Secondary Voltage - KV
670	3.16	760	2.76
680	3.20	770	2.72
690	3.18	780	2.66
700	3.16	790	2.60
710	3.10	800	2.56
720	3.00	810	2.52
730	2.90	820	2.46
740	2.84	830	2.42
750	2.80	840	2.40

TABLE 10

NON DESTRUCTIVE TESTS CORRESPONDING TO FIGURE 3
RUN 6

Reactor : 2 Electrode : Silver Paint

Primary Voltage : 30V

Gas : Air

Frequency - KHz	Secondary Voltage - KV	Frequency - KHz	Secondary Voltage - KV
600	3.60	740	2.82
610	3.56	750	2.80
620	3.50	760	2.78
630	3.40	770	2.76
640	3.30	780	2.72
650	3.24	790	2.66
660	3.20	800	2.62
670	3.16	810	2.60
680	3.12	820	2.56
690	3.06	830	2.50
700	3.00	840	2.46
710	2.96	850	2.42

TABLE 11

NON DESTRUCTIVE TESTS CORRESPONDING TO FIGURE 3
RUN 7

Reactor : 2

Electrode : Silver Paint

Primary Voltage : 40V

Gas : Air

Frequency - KHz	Secondary Voltage - KV	Frequency - KHz	Secondary Voltage - KV
500	4.30	730	2.86
510	4.21	740	2.80
520	4.18	750	2.78
530	4.10	760	2.76
540	4.04	770	2.72
550	4.00	780	2.66
560	3.96	790	2.60
570	3.88	800	2.56
580	3.80	810	2.50
590	3.70	820	2.46
600	3.60	830	2.42
610	3.56	840	2.40
620	3.52	850	2.38
630	3.44	860	2.34
640	3.38	870	2.30
650	3.28	880	2.24
660	3.22	890	2.20
670	3.18	900	2.16
680	3.14	910	2.10
690	3.10	920	2.04
700	3.02	930	2.00
710	2.96	940	3.96
720	2.90	950	1.9

TABLE 12

NON-DESTRUCTIVE TESTS CORRESPONDING TO FIGURE 3
RUN 8

Reactor : 2 Electrode : Silver Paint

Primary Voltage : 50V

Gas : Air

Frequency - Hz	Secondary Voltage - KV	Frequency - Hz	Secondary Voltage - KV
400	4.42	660	3.30
410	4.44	670	3.60
420	4.46	680	3.56
430	4.48	690	3.42
440	4.50	700	3.38
450	4.34	710	3.30
460	4.58	720	3.24
470	4.62	730	3.18
480	5.40	740	2.98
490	5.16	750	2.86
500	4.82	760	2.82
510	4.70	770	2.78
520	4.44	780	2.76
530	4.38	790	2.74
540	4.24	800	2.72
550	4.10	810	2.70
560	4.02	820	2.62
570	3.98	830	2.60
580	3.84	840	2.54
590	3.70	850	2.50
600	3.62	860	2.44
610	3.58	870	2.40
620	3.56	880	2.38
630	3.52	890	2.36
640	3.46	900	2.32
650	3.38		

TABLE 13

NON-DESTRUCTIVE TESTS CORRESPONDING TO FIGURE 3
RUN 9

Reactor : 2 Electrode : Silver Paint

Primary Voltage : 60V

Gas : Air

Frequency - Hz	Secondary Voltage - KV	Frequency - Hz	Secondary Voltage - KV
360	5.70	640	3.90
370	6.00	660	3.70
380	6.20	680	3.60
390	6.60	700	3.50
400	7.60	720	3.30
420	7.70	740	3.20
440	7.20	780	3.10
460	6.80	800	2.90
480	6.20	820	2.70
500	5.70	840	2.50
520	5.40	860	2.40
540	5.00	880	2.20
560	6.70	900	2.10
580	4.50	920	2.00
600	4.30	999	1.40
620	4.10		

TABLE 14

NON-DESTRUCTIVE TESTS CORRESPONDING TO FIGURE 3
RUN 10

Reactor : 2 Electrode : Silver Paint

Primary Voltage : 75V

Gas : Air

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
100	8.60	560	4.80
150	8.20	580	4.50
200	7.70	600	4.30
250	7.60	620	4.00
300	8.20	640	3.90
320	9.60	660	3.70
340	11.80	680	3.60
350	12.20	700	3.50
380	11.40	720	3.40
400	10.00	740	3.30
420	8.80	760	3.20
440	8.00	780	3.10
460	7.60	800	2.90
480	6.50	850	2.60
500	6.00	900	2.20
520	5.50	950	1.70
540	5.00	999	1.40

TABLE 15

NON-DESTRUCTIVE TESTS CORRESPONDING TO FIGURE 4
RUN 12

Reactor : 3 Electrode : Silver Nitrate

Primary voltage : 45V

Gas : Air

Frequency - Hz	Secondary Voltage - KV	Frequency - Hz	Secondary Voltage - KV
100	5.18	150	4.30
110	4.97	160	4.14
120	4.80	170	4.02
130	4.61	180	3.90

TABLE 15 (Continued)

Frequency - Hz	Secondary Voltage - KV	Frequency - Hz	Secondary Voltage - KV
140	4.44	190	3.80
200	3.70	430	2.74
210	3.60	440	2.67
220	3.53	450	2.60
230	3.46	460	2.54
240	3.40	470	2.48
250	3.34	480	2.42
260	3.30	490	2.37
270	3.26	540	2.22
280	3.23	550	2.18
290	3.22	560	2.13
300	3.21	570	2.09
310	3.20	580	2.05
320	3.20	590	2.04
330	3.21	600	2.01
340	3.23	610	1.98
350	3.26	620	1.94
360	3.30	630	1.90
370	3.18	640	1.86
380	3.13	650	1.83
390	3.04	660	1.80
400	2.97	700	1.66
410	2.89	750	1.28
420	2.81		

TABLE 16

NON-DESTRUCTIVE TESTS CORRESPONDING TO FIGURE 5
RUN 13

Reactor : 3

Electrode : Silver Nitrate

Primary Voltage :50V

Gas : Air

Frequency - Hz	Secondary voltage - KV	Power - watts	Frequency - Hz	Secondary voltage - KV	Power - watts
60	6.46	89	140	4.77	59
70	6.24	65	150	4.61	58
80	6.02	64	160	4.46	58
90	5.78	62	170	4.32	58
100	5.56	61	180	4.20	58
110	5.34	60	190	4.10	58

TABLE 16 (Continued)

Frequency - Hz	Secondary voltage - KV	Power - watts	Frequency - Hz	Secondary voltage - KV	Power - watts
120	5.14	60	200	3.99	60
130	4.95	59	210	3.90	60
220	3.81	60	999	0.32	56
230	3.74	60	980	0.38	66
240	3.68	60	970	0.41	66
250	3.62	60	960	0.43	66
260	3.57	59	950	0.46	66
270	3.54	60	940	0.49	66
280	3.51	60	930	0.52	66
290	3.49	60	920	0.54	66
300	3.48	60	910	0.73	66
310	3.47	60	900	0.60	66
320	3.48	60	890	0.63	66
330	3.49	60	880	0.67	65
340	3.47	100	870	0.70	65
350	3.35	100	860	0.74	65
360	3.24	100	850	0.78	65
370	4.47	98	840	0.82	65
380	3.04	97	830	0.87	65
390	3.05	97	820	0.92	65
400	2.96	97	810	0.97	65
410	2.87	97	800	1.03	65
420	2.79	96	790	1.09	69
430	2.72	96	780	1.16	70
440	2.64	95	770	1.23	70
450	2.68	95	760	1.31	70
460	2.63	95	750	1.41	72
470	2.57	95	740	1.51	73
480	2.51	95	730	1.64	75
490	2.45	95	720	1.60	76
500	2.40	94	710	1.62	76
510	2.35	92	700	1.65	76
520	2.30	91	690	1.67	77
530	2.24	91	680	1.68	77
540	2.20	91	670	1.71	77
550	2.16	90	660	1.74	77
560	2.11	90	650	1.76	77
570	2.06	90	640	1.79	77
580	2.02	89	630	1.82	80
590	1.98	87	620	1.84	81
600	1.94	87	610	1.86	81
610	1.91	86	600	1.89	81
620	1.88	85	590	1.93	82
630	1.86	85	580	1.95	82
640	1.84	85	570	1.98	82
650	1.80	84	560	2.02	83
660	1.77	83	550	2.03	83
970	0.39	60	540	2.07	84

TABLE 16 (Continued)

Frequency - Hz	Secondary voltage - KV	Power - watts	Frequency - Hz	Secondary voltage - KV	Power - watts
510	2.20	85	280	4.24	98
500	2.25	85	270	4.36	96
490	2.30	91	260	4.45	90
480	2.35	91	250	4.40	82
470	2.41	92	240	3.70	62
460	2.47	92	230	3.76	60
450	2.53	92	220	3.84	58
440	2.60	92	210	3.92	58
430	2.63	92	200	4.02	58
420	2.68	92	190	4.13	57
410	2.76	92	180	4.24	57
400	2.80	92	170	4.36	57
390	2.88	95	160	4.50	57
380	2.95	96	150	4.65	57
370	2.05	96	140	4.82	57
360	3.15	96	130	5.00	57
350	3.26	97	120	5.20	57
340	3.40	98	110	5.40	57
330	3.49	99	100	5.61	57
320	3.64	99	90	5.85	58
310	3.80	99	80	6.08	60
300	3.95	100	70	6.32	63
290	4.11	100	60	6.54	67

TABLE 17

NON-DESTRUCTIVE TESTS CORRESPONDING TO FIGURE 6 RUN 14

Reactor : 3 — Electrode: SilverNitrate

Primary Voltage : 30V — Pressure: 30 Psig

Gas : Air

Run #	Pressure - Psig	Frequency - Hz	Secondary Voltage- KV
60	4.30	140	4.40
70	4.40	150	4.50
80	4.40	160	4.60
90	4.40	170	4.60
100	4.40	180	4.60

TABLE 17 (CONTINUED)

Run #	Pressure - Psig	Frequency - Hz	Secondary Voltage- KV
110	4.40	190	4.70
120	4.40	200	4.80
130	4.40	210	4.80
220	4.80	680	9.40
230	4.90	690	8.80
240	5.00	700	8.20
250	5.00	710	7.60
260	5.10	720	7.20
270	5.20	730	6.80
280	5.20	740	6.40
290	5.40	750	6.00
300	5.60	760	5.60
310	5.70	770	5.40
320	5.80	780	5.20
330	6.00	790	4.80
340	6.20	800	4.70
350	6.40	810	4.50
360	6.60	820	4.30
370	6.80	830	4.10
380	7.20	840	4.00
390	7.20	850	3.80
400	7.80	860	3.60
410	8.20	870	3.60
420	8.60	880	3.40
430	9.20	890	3.40
440	9.20	900	3.20
450	10.40	910	3.0
460	11.3	920	3.00
470	12.30	930	2.80
480	12.40	940	2.70
490	15.00	950	2.60
500	17.20	960	2.50
510	20.20	970	2.40
520	23.70	980	2.30
530	21.40	990	2.20
540	20.60	999	2.00
670	10.40		

TABLE 18

NON-DESTRUCTIVE TESTS CORRESPONDING TO FIGURE 6
RUN 15

Reactor : 3
Electrode: SilverNitrate

Primary Voltage : 30V
Pressure: 40 Psig

Gas : Air

Frequency - Hz	Secondary Voltage-KV	Frequency - Hz	Secondary Voltage-KV
60	4.20	440	9.60
70	4.30	450	10.20
80	4.30	460	11.00
90	4.40	470	11.9
100	4.40	480	13.00
110	4.40	490	14.30
120	4.40	500	16.20
130	4.40	510	18.4
140	4.40	520	21.50
150	4.50	530	15.20
160	4.50	540	16.00
170	4.60	550	16.40
180	4.60	560	15.20
190	4.60	570	14.20
200	4.70	580	23.80
210	4.80	590	23.40
220	4.80	600	21.4
230	4.90	610	19.2
240	5.0	620	17.4
250	5.00	630	15.70
260	5.10	640	14.30
270	5.20	650	13.00
280	5.30	660	12.00
290	5.40	670	10.90
300	5.50	680	10.00
310	5.60	690	9.20
320	5.80	700	8.60
330	6.00	710	8.00
340	6.20	720	7.50
350	6.20	730	7.00
360	6.40	740	6.60
370	6.60	750	6.30
380	7.00	760	6.00
390	7.40	770	5.60
400	7.80	780	5.40
410	8.10	790	5.20
420	8.60	800	5.00
430	9.00	810	4.70

TABLE 19

NON-DESTRUCTIVE TESTS CORRESPONDING TO FIGURE 6
RUN 16

Reactor : 3 Electrode: SilverNitrate

Primary Voltage : 30V Pressure: 40 Psig

Gas : Air

Frequency - Hz	Secondary Voltage- KV	Frequency - Hz	Secondary Voltage- KV
60	4.40	440	9.50
70	4.40	450	9.20
80	4.40	460	10.90
90	4.40	470	11.80
100	4.40	480	12.90
110	4.40	490	14.20
120	4.40	500	16.20
130	4.40	510	18.40
140	4.40	520	21.20
150	4.50	530	24.80
160	4.50	540	28.20
170	4.60	550	30.40
180	4.60	560	30.40
190	4.60	570	29.00
200	4.70	580	27.30
210	4.80	590	24.80
220	4.80	600	22.20
230	4.80	610	20.00
240	4.90	620	18.00
250	5.00	630	16.00
260	5.10	640	14.40
270	5.20	650	13.20
280	5.20	660	12.00
290	5.40	670	11.00
300	5.50	680	10.00
310	5.60	690	9.20
320	5.80	700	8.60
330	6.00	710	8.00
340	6.20	720	7.40
350	6.40	730	7.00
360	6.60	740	6.50
370	6.80	750	6.20
380	7.00	760	5.80
390	7.40	770	5.50
400	7.70	780	5.20
410	8.00	790	5.00
420	8.50	800	4.80
430	9.00	810	4.60

APPENDIX-C

PRELIMINARY DESTRUCTIVE TEST DATA

TABLE 20

PRILIMINARY DESTRUCTIVE DATA CORRESPONDING TO FIGURE 7
RUN 1

Primary Voltage : 40 V

Flow Rate : 147.2 cc/min

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
500	11.2	710	27.2
510	11.6	720	27.2
520	12.2	730	27.2
530	12.6	740	27.2
540	13.2	750	27.4
550	13.6	760	27.4
560	14.4	770	27.4
570	15.0	780	27.6
580	15.8	790	27.4
590	16.6	800	27.3
600	17.6	810	27.2
610	18.6	820	26.8
620	19.8	830	26.4
630	21.0	840	26.0
640	22.4	850	25.6
650	24.0	860	25.0
660	25.2	870	24.6
670	26.0	880	24.2
680	26.4	890	22.4
690	26.8	900	22.8
700	27.0		

TABLE 21

PRILIMINARY DESTRUCTIVE DATA CORRESPONDING TO FIGURE 7
RUN 2

Primary Voltage : 50 V

Flow Rate : 147.2 cc/min

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
510	14.2	570	18.4
520	15.0	580	19.4
530	15.6	590	20.4
540	16.2	600	21.6

TABLE 21 (CONTINUED)

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
630	25.8	820	32.0
640	27.4	830	32.0
650	28.2	840	31.8
660	28.8	850	31.2
670	28.2	860	30.8
680	28.2	870	30.2
690	28.0	880	29.6
700	28.2	890	29.0
710	29.0	900	28.4
720	28.6	910	27.8
730	28.8	920	27.2
740	25.8	930	26.6
750	25.4	940	25.8
760	25.8	950	25.2
77	30.2	960	24.4
780	30.4	970	23.4
79	31.0	980	22.4
800	31.2	990	21.6
810	31.8	999.9	20.8

TABLE 22

PRILIMINARY DESTRUCTIVE DATA CORRESPONDING TO FIGURE 7
RUN 3

Primary Voltage 50 V

Flow Rate : 147.2 cc/min

Frequency Hz	Secondary Voltage KV	Frequency Hz	Secondary Voltage KV
450	16.4	600	31.4
460	17.0	610	31.8
470	17.4	620	32.4
480	18.0	630	32.8
490	18.6	640	33.0
500	19.4	650	33.2
510	20.2	660	33.0
520	21.0	670	33.2
530	22.0	680	33.6
540	23.0	690	34.0
550	24.0	700	34.0
560	25.4	710	34.4
570	26.6	720	34.6
580	28.2	730	34.8
590	29.8	740	35.0

TABLE 23

PRILIMINARY DESTRUCTIVE DATA CORRESPONDING TO FIGURE 7 RUN 4

Primary Voltage : 70 V

Flow Rate : 147.2 cc/min

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
400	16.4	530	26.6
410	17.0	540	28.0
420	17.4	550	29.6
430	17.8	560	31.0
440	18.4	570	32.0
450	19.0	580	32.4
460	19.8	590	32.6
470	20.4	600	33.2
480	20.6	610	33.2
490	22.2	620	33.4
500	23.2	630	33.8
510	24.2	640	33.8
520	25.2		

TABLE 24

PLRILIMINARY DESTRUCTIVE DATA CORRESPONDING TO FIGURE 8 RUN 5

Primary Voltage : 40V

Flow Rate : 252.5 CC/MIN

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
500	10.0	680	23.2
510	10.2	690	24.2
520	10.4	700	24.6
530	10.8	710	24.8
540	11.2	720	25.0
550	11.6	730	25.0
560	12.0	740	25.0
570	12.6	750	25.0
580	13.2	760	25.0
590	13.8	770	25.0
600	14.6	780	24.8
610	15.4	790	24.2

TABLE 24 (CONTINUED)

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
620	16.2	800	23.6
630	17.2	810	23.2
640	18.4	820	22.6
650	19.6	830	22.0
660	20.8	840	21.6
670	21.0		

TABLE 25

PRILIMINARY DESTRUCTIVE DATA CORRESPONDING TO FIGURE 8
RUN 6

Primary Voltage : 50 V

Flow Rate : 252 cc/min

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
450	8.6	650	22.2
460	8.8	660	22.4
470	9.0	670	22.4
480	9.4	680	22.4
490	9.6	690	22.4
500	10.0	700	22.4
510	10.2	710	22.2
520	10.8	720	22.2
530	11.2	730	22.2
540	11.8	740	22.4
550	12.2	750	22.4
560	12.8	760	22.6
570	13.4	770	22.6
580	14.4	780	22.6
590	15.2	790	22.8
600	17.4	800	22.8
610	18.2	810	22.6
620	19.6	830	21.8
630	20.8	840	21.0
640	21.8	850	20.4

TABLE 26

PRILIMINARY DESTRUCTIVE DATA CORRESPONDING TO FIGURE 8
RUN 7

Primary Voltage : 60 V

Flow Rate : 252.5cc/min

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
550	15.8	750	20.2
560	16.4	760	20.4
570	17.2	770	20.4
580	18.2	780	20.2
590	19.2	790	20.2
600	20.4	800	20.2
610	21.2	810	20.2
620	21.6	820	20.2
630	22.0	830	20.0
640	21.2	840	19.8
650	21.2	850	19.6
660	20.8	860	19.4
670	20.6	870	18.8
680	20.6	880	18.4
690	20.6	890	17.8
700	20.6	900	17.2
710	20.6	910	16.6
720	20.4	920	15.8
730	20.4	930	15.2
740	20.4	940	14.6

TABLE 27

PRILIMINARY DESTRUCTIVE DATA CORRESPONDING TO FIGURE 8
RUN 8

Primary Voltage : 70 V

Flow Rate : 252.5cc/min

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
510	16.6	590	21.8
520	17.4	600	21.4
530	18.0	610	21.4
540	18.8	620	20.8
550	19.6	630	20.2
560	19.8	640	20.6

TABLE 27 (CONTINUED)

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
670	20.2	820	20.6
680	20.1	830	20.6
690	20.2	840	20.4
700	20.0	850	20.0
710	20.0	860	19.6
720	20.0	870	19.2
730	20.0	880	18.6
740	20.0	890	18.0
750	20.0	900	17.4
760	20.2	910	16.8
770	20.4	920	16.2
780	20.4	930	15.4
790	20.8	940	14.8
800	20.8	950	14.2
810	20.6		

TABLE 28

PRILIMINARY DESTRUCTIVE DATA CORRESPONDING TO FIGURE 9
RUN 9

Primary Voltage : 40V

Flow Rate :367.2 cc/min

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
550	10.6	730	22.4
560	11.0	740	22.2
570	11.6	750	22.2
580	12.0	760	22.0
590	12.6	770	22.0
600	13.4	780	21.8
610	14.0	790	21.2
620	15.0	800	20.8
630	15.8	810	20.0
640	17.0	820	19.4
650	18.2	830	18.8
660	19.4	840	18.2
670	20.8	850	17.4
680	21.4	860	16.8
690	22.2	870	16.0
700	22.4	880	15.2
710	22.4	890	14.6
720	22.4	900	13.8

TABLE 29

PRILIMINARY DESTRUCTIVE DATA CORRESPONDING TO FIGURE 9
RUN 10

Primary Voltage : 50 V

Flow Rate : 367.2 cc/min

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
550	11.0	730	21.4
560	11.4	740	21.6
570	12.0	750	21.6
580	12.6	760	21.6
590	13.4	770	21.8
600	13.2	780	21.8
610	15.0	790	21.8
620	16.0	800	21.8
630	19.2	810	21.4
640	20.4	820	21.0
650	21.2	830	20.2
660	21.6	840	19.6
670	21.6	850	19.0
680	21.6	860	18.2
690	21.6	870	17.4
700	21.6	880	16.8
710	21.4	890	16.0
720	21.4	900	15.4

TABLE 30

PRILIMINARY DESTRUCTIVE DATA CORRESPONDING TO FIGURE 9
RUN 11

Primary Voltage : 60 V

Flow Rate : 367.2 cc/min

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
510	13.6	740	22.2
520	14.2	750	22.4
530	14.4	760	22.4
540	15.0	770	22.6
550	15.6	780	22.4
560	16.2	790	22.4
570	17.4	800	22.2

TABLE 30 (CONTINUED)

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
600	19.8	830	21.6
610	20.8	840	21.2
620	21.6	850	20.6
630	22.0	860	19.8
640	22.2	870	19.2
650	22.2	880	18.4
660	22.2	890	17.4
670	22.2	900	16.6
680	22.2	910	15.8
690	22.2	920	15.0
700	22.2	930	14.2
710	22.2	940	13.2
720	22.2	950	12.6
730	22.2		

TABLE 31

PRILIMINARY DESTRUCTIVE DATA CORRESPONDING TO FIGURE 9
RUN # 12

Primary Voltage : 70 V

Flow Rate : 367.2 cc/min

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
450	14.8	680	22.0
460	15.4	690	22.2
470	15.8	700	22.2
480	15.2	710	22.0
490	16.8	720	22.2
500	17.0	730	22.0
510	18.8	740	22.0
520	18.8	750	21.8
530	19.6	760	21.8
540	20.6	770	21.8
550	21.2	780	21.8
560	22.6	790	21.6
570	22.4	800	21.4
580	22.4	810	21.4
590	22.6	820	21.2
600	22.6	830	20.8
610	22.2	840	20.4
620	22.4	860	19.4
630	22.2	870	18.8
640	22.0	880	18.0
650	21.8	890	17.2

TABLE 32
PRILIMINARY DESTRUCTIVE DATA CORRESPONDING TO FIGURE 10
RUN 13

Pnmary Voltage : 40 V

Flow Rate : 481.15 CC/MIN

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
550	10.6	730	22.0
560	11.2	740	22.0
570	11.6	750	21.8
580	12.2	760	21.6
590	13.0	770	21.2
600	13.6	780	20.8
610	14.6	790	20.4
620	15.4	800	19.6
630	16.4	810	19.0
640	17.2	820	18.4
650	18.2	830	17.6
660	19.4	840	17.0
670	20.4	850	16.4
680	21.2	860	15.6
690	21.8	870	15.0
700	22.0	880	14.2
710	22.2	890	13.6
720	22.2	900	12.8

TABLE 33

PRILIMINARY DESTRUCTIVE DATA CORRESPONDING TO FIGURE 10
RUN 14

Primary Voltage : 50 V

Flow Rate : 481.15 cc/min

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
510	11.0	610	18.2
520	11.4	620	19.0
530	12.0	630	20.8
540	12.8	640	21.8
550	13.4	650	22.2
560	14.0	660	22.2
570	14.6	670	22.2
580	15.4	680	22.2

TABLE 33 (CONTINUED)

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
710	22.6	810	21.6
720	22.6	820	21.0
730	22.6	830	20.2
740	22.6	840	19.6
750	22.6	850	18.8
760	22.6	860	18.0
770	22.4	870	17.2
780	22.4	880	16.4
790	22.2	890	15.6
800	22.0	900	14.8

TABLE 34

PRILIMINARY DESTRUCTIVE DATA CORRESPONDING TO FIGURE 10
RUN 15

Primary Voltage : 60 V

Flow Rate : 481.15 CC/MIN

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
500	13.8	700	22.0
510	14.4	710	22.0
520	15.0	720	22.0
530	15.4	730	21.8
540	16.2	740	21.6
550	17.0	750	21.6
560	18.2	760	21.6
570	19.0	770	21.4
580	20.0	780	21.4
590	20.8	790	21.2
600	20.8	800	21.2
610	22.0	810	21.0
620	22.0	820	20.6
630	22.2	830	20.4
640	22.4	840	19.8
650	22.2	850	19.2
660	22.2	860	18.4
670	22.2	870	17.8
680	22.0	880	17.0
690	22.0	890	16.2

TABLE 35

PRILIMINARY DESTRUCTIVE DATA CORRESPONDING TO FIGURE 10
RUN 16

Primary Voltage : 70 V

Flow Rate : 481.15cc/min

Frequency - Hz	Secondary Voltage KV	Frequency - Hz	Secondary Voltage KV
450	13.2	680	21.2
460	13.8	690	21.2
470	14.2	700	21.2
480	14.6	710	21.2
490	15.0	720	21.2
500	15.6	730	21.2
510	16.2	740	21.2
520	16.8	750	21.0
530	17.4	760	21.0
540	18.2	770	20.8
550	18.8	780	20.6
560	20.0	790	20.6
570	21.4	800	20.4
580	22.6	810	20.2
590	23.0	820	20.0
600	22.0	830	19.8
610	22.0	840	19.6
620	22.0	850	19.2
630	21.2	860	18.8
640	21.6	870	18.2
650	21.6	880	17.8
660	21.8	890	17.2
670	21.4	900	16.6

APPENDIX-D

DESTRUCTIVE TEST DATA

TABLE 36

DESTRUCTIVE TEST DATA CORRESPONDING TO FIGURE 11

Primary Voltage : 40V

Flow Rate : 147.2 cc/min

Duration of Each Run : 10 minutes

Run Number	Frequency Hz	Secondary Voltage KV	Power Watts	Temperature C	Amount of Sulphur in Grams	Percentage Conversion
1	690	22.4	92	21	0.0297	1.522
3	720	22.4	90	23	0.0871	4.46
4	730	22.8	90	23	0.0187	0.96
5	740	21.8	94	20	0.0233	1.19
6	750	22.2	90	25	0.0228	1.17
7	770	22.0	89	27	0.0179	0.93
8	780	20.6	84	27	0.0230	1.2
9	800	19.2	81	28	0.0139	0.73

TABLE 37

DESTRUCTIVE TEST DATA CORRESPONDING TO FIGURE 12

Primary Voltage : 50V

Flow Rate : 147.2 cc/min

Duration of Each Run : 10 minutes

Run Number	Frequency Hz	Secondary Voltage KV	Power Watts	Temperature C	Amount of Sulphur in Grams	Percentage Conversion
10	660	22.2	90	29	0.0732	3.85
11	670	21.8	94	25	0.1160	6.02
12	690	22.0	94	22	0.0710	3.65
14	710	21.6	95	21	0.1285	6.60
15	720	21.4	90	30	0.0646	3.41
17	730	23.6	88	27	0.0244	1.27
18	740	23.2	86	28	0.0313	1.64
19	750	24.0	90	28	0.0690	3.62
20	770	24.2	89	29	0.0319	1.68
21	790	21.6	89	28	0.0447	2.34
22	820	18.8	85	28	0.0396	2.07

TABLE 38

DESTRUCTIVE TEST DATA CORRESPONDING TO FIGURE 13

Primary Voltage : 60V

Flow Rate : 147.2 cc/min

Duration of Each Run : 10 minutes

Run Number	Frequency Hz	Secondary Voltage KV	Power Watts	Temperature C	Amount of Sulphur in Grams	Percentage Conversion
23	610	21.4	90	28	0.1299	6.81
24	630	21.4	91	28	0.1600	8.4
25	640	21.2	95	28	0.1559	8.2
26	650	24.8	95	26	0.1683	8.77
27	670	25.0	92	26	0.1431	7.45
28	680	21.4	93	22	0.1535	7.9
30	690	21.0	92	25	0.1518	7.88
31	710	22.2	93	24	0.1381	7.15
32	730	19.4	96	25	0.1649	8.56
33	750	22.6	94	25	0.1109	5.76

TABLE 39

DESTRUCRTIVE TEST DATA CORRESPONDING TO FIGURE 14

Primary Voltage : 70V

Flow Rate : 147.2 cc/min

Duration of Each Run : 10 minutes

Run Number	Frequency Hz	Secondary Voltage KV	Power Watts	Temperature C	Amount of Sulphur in Grams	Percentage Conversion
34	550	20.4	88	26	0.1396	7.27
35	560	21.0	94	28	0.2437	12.8
36	570	20.2	98	30	0.2147	11.34
37	580	20.8	94	31	0.1622	8.60
38	590	20.6	94	31	0.2097	11.11
39	600	21.0	98	32	0.1733	9.21
40	620	20.4	95	32	0.2007	10.67
42	630	23.0	100	21	0.1791	9.17
43	650	22.6	97	23	0.3114	16.06
44	670	22.6	97	24	0.2854	14.77
45	690	21.8	100	28	0.1673	8.78

TABLE 40

DESTRUCTIVE TEST DATA CORRESPONDING TO FIGURE 15

Primary Voltage : 90 V

Flow Rate : 147.2 cc/min

Duration of Each Run : 10 minutes

Run Number	Frequency Hz	Secondary Voltage KV	Power Watts	Temperature C	Amount of Sulphur in Grams	Percentage Conversion
46	500	22.0	120	28	0.2230	11.66
47	510	21.6	116	32	0.2050	11.00
48	520	21.6	117	31	0.2127	11.30
49	530	22.4	120	22	0.1943	10.00
50	550	22.0	120	22	0.1928	10.07
51	560	22.4	117	28	0.2050	10.77
52	570	22.0	115	30	0.1780	9.41
53	590	21.8	115	30	0.1857	10.18
54	490	21.6	110	33	0.1827	9.75
55	610	21.2	110	38	0.1796	9.74

TABLE 41

DESTRUCTIVE TEST DATA CORRESPONDING TO FIGURE 16

Primary Voltage : 100 V

Flow Rate : 147.2 cc/min

Duration of Each Run : 10 minutes

Run Number	Frequency Hz	Secondary Voltage KV	Power Watts	Temperature C	Amount of Sulphur in Grams	Percentage Conversion
56	460	19.2	135	27	0.4337	22.7
59	470	19.0	135	28	0.2925	15.4
60	480	18.4	130	30	0.2778	14.7
62	500	17.8	130	31	0.2667	14.05

TABLE 42

DESTRUCTIVE TEST DATA CORRESPONDING TO FIGURE 17

Fixed Condition : Flow Rate = 147.2 cc/min

Primary Voltage, Volts	Average Percentage Conversion
40	1.1
50	3.15
60	8.06
70	9.28
90	10.74
100	16.0

TABLE 43

DESTRUCTIVE TEST DATA CORRESPONDING TO FIGURE 18

Primary Voltage : 70 V

Duration of Each Run : 10 minutes

Run Number	Frquency Hz	Secondary Voltage KV	Flow Rate cc/min	Power Watts	Temperature C	Amount of Sulphur in Grams	Percent Conversion
100	560	10.0	8.70	100	26	0.1043	91.90
78	560	11.4	18.18	105	24	0.1036	43.42
67	560	14.2	56.00	97	24	0.1550	21.00
35	560	21.0	147.20	94	28	0.2437	12.80
107	590	10.4	8.70	98	25	0.0893	78.47
81	590	11.0	18.18	102	26	0.1209	51.10
68	590	13.8	56.00	101	25	0.1670	22.80
38	590	20.6	147.2	94	31	0.2097	11.11
99	650	9.4	8.70	95	26	0.0980	86.40
83	650	11.2	18.18	98	29	0.1418	60.43
69	650	13.8	56.00	100	27	0.1470	20.20
42	650	22.6	147.2	97	23	0.3114	16.06

TABLE 44

DESTRUCTIVE TEST DATA CORRESPONDING TO FIGURE 19

Primary voltage : 70 V

Flow Rate : 56 cc/min

Frequency : 590 Hz

Run Number	Secondary Voltage KV	Power Watts	Duration of Each Run in minutes	Temperature C	Amount of Sulphur in Grams	Percent Conversion
84	13.4	105	5	22	0.0808	21.80
85	13.0	100	10	32	0.1412	19.73
86	14.2	101	15	27	0.1753	16.00
87	14.2	100	20	26	0.2658	18.20
88	14.4	102	30	25	0.4008	18.24

APPENDIX-E

DISCUSSION ON FREE ENERGY MINIMIZATION TECHNIQUE

The Free Energy Minimization Technique

Equilibrium can be defined as that state of a system in which the total free energy of that system is a minimum. In essence, all that is required is to obtain an expression for the free energy of the system in terms of the potentials of the species involved, and to minimize this quantity subject to certain external constraints.

For chemical systems, the technique was first suggested by White et al. [31] and involves the chemical potentials of the species present. The technique as followed by the NASA scientists is enumerated here. The equations given below are directly taken from reference [7]. The free energy of a mixture of n chemical species containing x_i moles of the i^{th} species (in the ideal gaseous state) can be expressed as

$$g = \sum_{j=1}^{n} \mu_j n_j$$

where the chemical potential per kilogram mole of species j is defined to be

$$\mu_j = (\partial g / \partial n_j)_{T,P,n_j \neq 1}$$

and n_j is the number of kilogram- mols of species j per kilogram of mixture.

To determine the equilibrium composition, it is necessary to find a set of non-negative values of n_j which minimizes equation 5.1, while also satisfying the mass balance constraint expressed as

$$\sum_{j=1}^{n} a_{ij} n_j - b_{io} = 0 \qquad i=1, \ldots\ldots\ldots, l \quad -(5.2)$$

where the stoichiometric coefficients a_{ij} are the number of kilogram atoms of element i per kilogram-mole of species j and b_{io} is the assigned number of kilogram-atoms of element i per kilogram of total reactants.

Now defining a term G to be,

$$G = g + \sum_{i=1}^{\ell} \lambda_i (b_i - b_{io}) \qquad \text{-(5.3)}$$

where λ_i are lagrangian multipliers. The condition for equilibrium becomes

$$\partial g = \sum_{j=1}^{n} (\mu_j + \sum_{i=1}^{\ell} \lambda_i a_{ij}) \partial n_j + \sum_{i=1}^{\ell} (b_i - b_{io}) \partial \lambda_i = 0 \qquad \text{-(5.4)}$$

Treating the variations $\partial\ n_j$ and $\partial\lambda\ _i$ as independent gives

$$\mu_j + \sum_{j=1}^{\ell} \lambda_i a_{ij} = 0 \qquad j=1,\ldots\ldots\ldots\ldots,n \qquad \text{-(5.5)}$$

and the mass balance equation 5.2 .

Based on the ideal gas assumption, the chemical potential may be written as

$$\mu_j = \mu_{jo} + RT\ln(n_j / n) + RT\ln P_{atm} \quad j=1,\ldots\ldots\ldots\ldots,m$$

where μ_{jo} for gases (j=1 to m) is the chemical potential in the standard state. Equations 5.2 and 5.5 permit the determination of equilibrium compositions for thermodynamic states specified by an assigned temperature T_0 and P_0.

The mathematics of the minimization technique can be summarized as follows :

1. An expression in the form of equation (5.1) is written for the free energy of the systems starting with a positive set of values n (n_1,.........n_n) for the species involved, which satisfies the mass balance constraint in equation 5.2. This set constitutes the initial guesses for the product distribution.
2. A Taylor series expansion of this expression is generated in terms of the differences between the final solution seat of values and the starting values assigned.
3. The expanded function, which is an approximation to the free energy subject to the mass balance constraint using Lagrange multipliers. This is done by setting the partial differential of the free energy expression with respect to each species equal to zero.

4. A set of simultaneous equations result from these equations. Solution of these yields a new composition which approximates more closely to the true value than did the original guessed set , at the starting of the computation.
5. This procedure is repeated until the new composition is the same as the previous one within desired limits. This set of product values corresponds to the composition having the minimum free energy for that system.

One of the advantages of this technique, is that little prior knowledge of the system to be investigated is required.. The method does not require any knowledge of reactions or specific equilibria involved. The generality and flexibility of this technique make it ideal for tackling a variety of problems involving equilibria.

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