# REMOVING $SO_2$ AND $NO_X$ FROM FLUE GAS OF ABADAN REFINERY BY NONTHERMAL PLASMA

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***ABSTRACT*****:** *Air pollution is one of the major problems facing the world resulted from concentration of airborne pollutants. The major sources of air pollution are combustion processes of fossil fuels. Flue gases generated by combustion contain air pollutants such as $SO_2$, $NO_x$ which are of great concern and they must be treatment before they can be released to the atmosphere. In this study the flue gas from distillation unit 75 of Abadan refinery was treated by corona discharge to remove $SO_2$ and $NO_x$. A glass reactor with an id of 20 mm consisting of one inlet pipe with an id of 5 mm and one exit pipe with an id of 5 mm including two electrodes with an id of 3 mm was used for this purpose. The electrodes were connected to an AC power supply with a high-voltage transformer to initiate the corona discharge. The inlet voltage (220 V) can be increased to a maximum of 7 kV by using the transformer. A facility of taking samples from inlet and outlet gases was included in the reactor. The effects of electrode distance (3, 5, and 5 mm) and discharge power (3, 4, 5, 6, and 7 kV) on NO, $NO_2$, and $SO_2$ removal were investigated. The results indicated that as the discharge power of the plasma increased the removal efficiency of three pollutants increased. The effect of electrode distance on removal efficiency of the pollutants indicated that between plasma powers of 3 to 5 kV the removal efficiency increased to a maximum and afterward the removal efficiency decreased as the plasma power increased further to 7 kV.*

Keywords: Flue gas, $SO_2$ removal, $NO_x$ removal, corona discharge, petroleum refinery

## INTRODUCTION

Air pollution is one of the major problems facing the world resulted from concentration of airborne pollutants. The major sources of air pollution are combustion processes of fossil fuels used in power plants, motor vehicles, and other form of transportation, including ships, airplanes and trains [1]. In addition to combustion some industrial processes such as refineries, chemical and petrochemical worsen the dilemma of air pollution.

Major combustion-generated air pollutants are $SO_x$, $NO_x$, particulate matter, carbon monoxide and unburned hydrocarbons. These pollutants are judged the primary pollutants of the atmosphere that bring about environmental problems similar to photochemical smog, acid rain, ozone layer depletion, and global warming [2]. Air pollutants can affect vegetation, soil composition, bodies of water and aquatic life, atmospheric visibility, surfaces of buildings, works of art, and personal property and produce unpleasant odors and unwanted noises [3].

Each of air pollution sources (power plants, car engines, etc.) in combustion processes has its own preferred methods of $SO_x$ and $NO_x$ emission control. However, some technologies can find application in $SO_x$ and $NO_x$ emission abatement for many types of sources of these pollutants [1]. Some methods and technologies for controlling the $SO_x$ and $NO_x$ emissions from flue gas include using appropriate fuels, control air and fuel mixing to achieve staged combustion, flue gas recirculation, absorption, adsorption, reduction, ozone destruction, and plasma destruction [1].

Plasma technologies have been the area of extensive inspection for more than five decades [4, 5]. Plasma is a collection of partly ionized gases containing molecules, atoms, ions, electrons, and free radicals in a neutral background gas [6]. In non-thermal plasma, the bulk gas, which is the main constituent, has a temperature very close to the ambient temperature, whereas the electrons are hotter and can thus lead to a reaction through excitation, ionization and dissociation processes [7]. In the plasma the collision between the highly velocity charged particles and other chemical species such as $SO_x$ and $NO_x$ takes on. This collision can contribute to the destruction of the pollutants species [8].

Non-thermal plasma can be generated by different kinds of gas discharge, including glow discharge, microwave discharge, dielectric barrier discharge (DBD), and corona discharge [9]. This process is capable of treating both small and large gas volumes, making them suitable for use with a wide variety of industrial and commercial processes [10, 11].

Corona discharges are transient discharges generated by strongly inhomogeneous electric fields associated with thin wires, needles or sharp edges of an electrode. Various geometries are used for generating corona discharges, the most common being pin-to-plate, wire-cylinder, and wire-plane. The discharge can be supplied by a constant voltage (DC corona), an alternating voltage (AC corona) or a pulsed voltage [12].

The first effort to use nonthermal plasma to clean flue gas was initiated by Ebara Corporation in Japan in 1970 and 1971. It was shown that electron beam could be used to remove $SO_2$ and $NO_x$ resulting from flue gases. The success of these initial batch tests indicated a future potential use for this method of treating flue gases. Later development of the process continued in many countries such as USA [11], Germany [13], Poland [14], Japan [15, 16], and China [16, 17, 18, 19], and Bulgaria [1].

Abadan refinery is located in Abadan city near the Iran-Iraq border. It is the oldest refinery in the Middle East and one of the biggest refineries in Iran which was completed in 1912. Since it is located within the city there is a need to remove $SO_2$ and $NO_x$ resulting from flue gases from combustion processes. The objective of this paper is to perform a preliminary study of using nonthermal plasma to treat Abadan refinery flue gas.

## MATERIALS AND METHODS

Fig. 1 shows the experimental setup used in this experiment. One glass reactor (with an id of 20 mm) consisting of one inlet pipe (with an id of 5 mm) for inlet gas, one exit pipe (with an id of 5 mm) for exit gas, two electrodes (with an id of 3 mm) was made by glass casting workshop at Science College of Shiraz University. The distance between the electrodes could be varied. The electrodes were connected to an AC power supply with a high-voltage transformer (Iran RADAR Electric 30 mA) to initiate the corona discharge. The inlet voltage (220 V) can be increased to a maximum of 7 kV by using the transformer. A facility of taking samples from inlet and outlet gases was included in the reactor. Flue gas exiting distillation unit 75 stack of Abadan refinery was drawn by a pump and was sent to the reactor inlet. The inlet flow rate of flow gas to the reactor was kept constant at 20 ml/min during the experiment. The reactor inlet and outlet gases were analyzed by a gas analyzer (Testo 350 M/XL). The effects of electrode distance (3, 5, and 5 mm) and discharge power (3, 4, 5, 6, and 7 kV) on $NO_x$ and $SO_2$ removal were investigated.

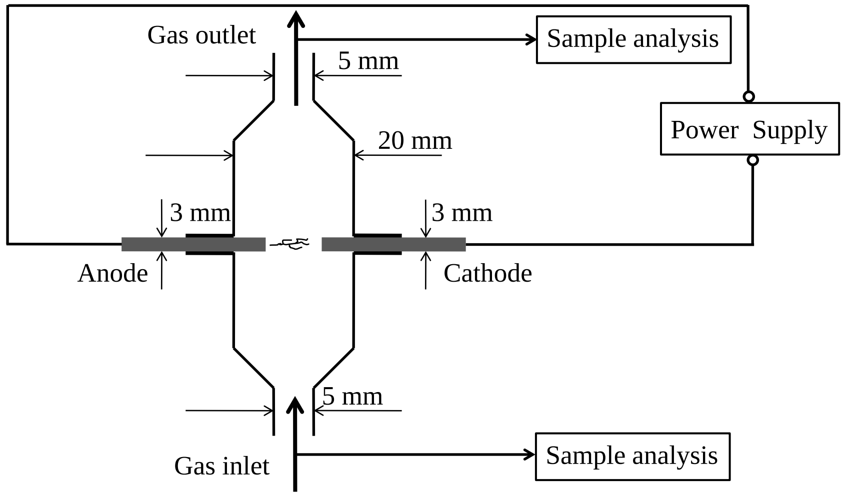


Fig. 1. Experimental setup

## RESULTS AND DISCUSSION

The analysis of stack flue gas (reactor feed) is shown in Tables 1. The effect of discharge power on NO, $NO_2$, and $SO_2$ reduction is demonstrated in Fig. 2. As the discharge power of the plasma increases the removal efficiency of the technique for the three pollutant increases which is similar to what is reported in the literature [18, 19].

Table 1. Flue gas composition (reactor feed)

| Component | Amount | Component | Amount | Component | Amount |
|---|---|---|---|---|---|
| $O_2$ (%) | 7.64 | $CO_2$ (%) | 6.87 | $NO_2$ (ppm) | 2.0 |
| $N_2$ (%) | 85.49 | NO (ppm) | 38 | $SO_2$ (ppm) | 19 |

In this method, electrons are produced, accelerated, and multiplied in plasma. Many kinds of radicals are subsequently produced by collision of the accelerated electrons with combustion gas molecules such as water vapor, oxygen, and nitrogen. Then $NO_x$ and $SO_2$ are oxidized or reduced by the reaction with the radicals [20]. The removal efficiency is increased with plasma power due to higher powers provided to the plasma that produce a larger concentration of radicals to destroy the pollutants. In the lower range of plasma power (between 3 to 4 kV) the effect of plasma power on pollutants removal is the same for all pollutants; however, at higher range of plasma power (between 4 to 7 kV) the effect on $NO_x$ removal is much more pronounce.

The effect of electrode distance on removal efficiency of the pollutants is illustrated in Fig. 3. Increasing the electrodes distance from 3 to 5 mm increases the removal efficiency to a maximum and afterward the removal efficiency decreases as the electrodes distance increases further to 7 mm. A possible explanation is given in the next paragraphs.

Two factors affect the plasma power between the electrodes; gap spacing between the electrodes and the possibility of corona to bridge between the electrodes. If the applied voltage is too high, the corona may bridge in the gas space and produce a spark that reduces plasma power [8]. As the gas spacing decreases the plasma power increases [10].

Increasing the electrodes distance from 3 to 5 mm increases the pollutants removal efficiency as a result of increase in plasma power which increases the radical concentration; Between 3 to 5 mm the increase in plasma power due to lower possibility lowering corona bridge formation is probably much more than the plasma power decreasing due to higher gas spacing. However, increasing the electrodes distance further (from 5 to 7 mm) reduces the plasma power due to the more effect of plasma power weakening owing to higher gas spacing.

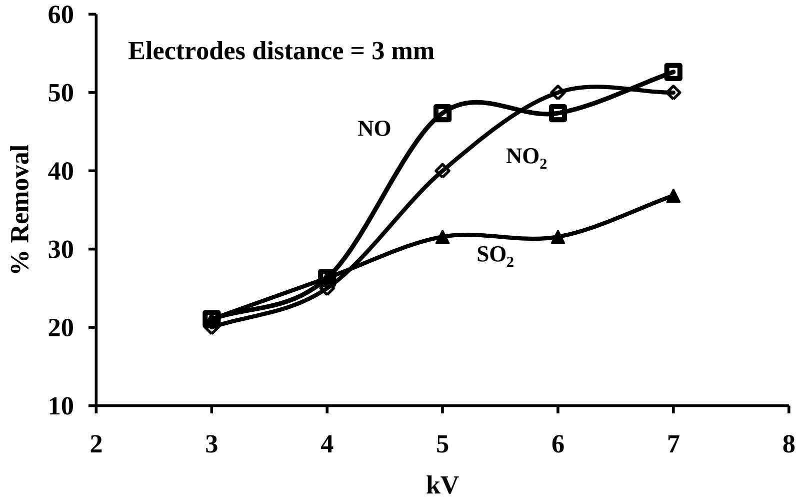


Fig. 2. Effect of plasma voltage on removal efficiency at an electrode distance of 3 mm

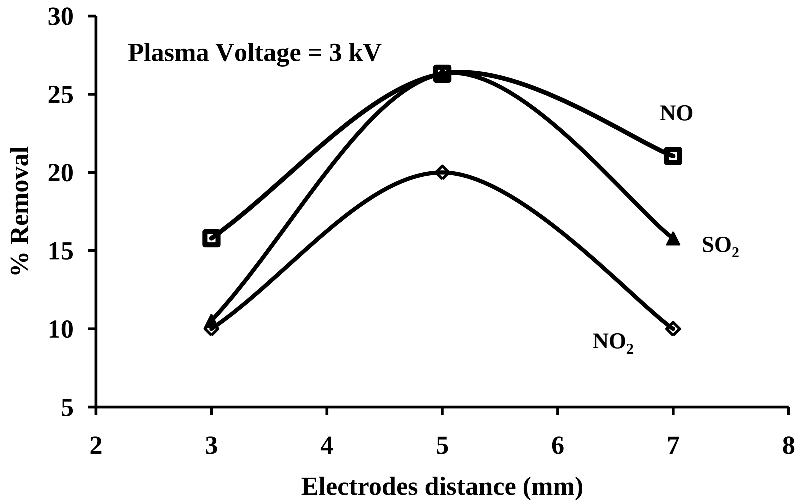


Fig. 3. Effect of electrode distance on removal efficiency at plasma voltage of 3 kV

## CONCLUSION

The capability of non equilibrium plasma power in removing three pollutants (NO, $NO_2$, and $SO_2$) was investigated. In this research the effects of electrode distance and plasma discharge power on NO, $NO_2$, and $SO_2$ removal were studied. The removal efficiency was increased with plasma power due. In the lower range of plasma power (between 3 to 4 kV) the effect of plasma power on pollutant removal was the same for all pollutants; however, at higher range of plasma power (between 4 to 7 kV) the effect on $NO_x$ removal was much more pronounce. The effect of electrode distance on removal efficiency of the pollutants indicated that at gap spacing of 3 to 5 mm the removal efficiency increased to a maximum and afterward the removal efficiency decreased as the electrode distance increased further to 7 mm.

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