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Development of Plasma-Chemical Technology for Purification of Acid Gases from Hydrogen Sulfide

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ABSTRACT: The main aim of the paper is an investigation of associated petroleum gas and acid gas conversion by plasma waste-free technology. Plasmachemical technology helps to achieve the hydrogen supplied final fracture down to elementary sulphure and hydrogen by streamer electric discharge. Availability and efficiency of offered technology are rate, herewith high selectivity to hydrogen sulphide. It has developed actual physical model for hydrogen sulphide conversion in to molecular hydrogen elemental sulphure. In experimental work a tendency to use nanosecond width impulse is observing.

KEY WORDS: Plasmachemical Technology, Associated Petroleum Gases, Hydrogen Sulfide Removing, Elemental Sulfur Recovery, Electric Discharge of Streamer Corona, Nanosecond Width Impulse, Actual Physical Model

I.INTRODUCTION

Purification of gas streams from sulfur-containing components is currently carried out mainly by traditional absorption and adsorption methods, as well as their various modifications. In industry, especially oil and gas complex at various stages of extraction and processing, sulfur-containing, so-called acidic (H_2S, CO_2, mercaptans R-SH, COS and CS_2) components in gases are found in the aggregate, and their purification is carried out in a complex manner at the same installations.

There are theoretically many methods for gas desulfurization. All of them have certain advantages and a number of disadvantages [1-2].

Among the non-traditional methods of gas purification from hydrogen sulfide, an important place is occupied by plasma technologies, the scope of which is expanding in various industries. Scientific research is being carried out on the use of plasma in the oil and gas industry. Various types of electrical discharges are investigated, with a large range of parameter values. The most promising is the use of a streamer corona discharge, the possibilities of which for gas purification from environmentally harmful impurities are being actively studied both in our country, and in many countries of Europe, China, Japan, North America [3].

II. SIGNIFICANCE OF THE SYSTEM

The prospects for the development of this method are determined by the expectation of its high efficiency and environmental friendliness.

As part of this work, laboratory studies were carried out on the process of purification of associated gases from hydrogen sulfide and their processing into elemental sulfur in electric corona discharge, the form of which is a streamer. Streamers are narrow luminous branched channels formed in the pre-breakdown stages of spark and corona discharges.

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The use of a streamer corona for gas purification, in particular from hydrogen sulfide, is based on the creation of active radicals and molecules, followed by removal which presents a simpler problem. The potential of the streamer corona is considered both in the field of processing of primary oil and gas raw materials, and in the field of waste disposal and synthesis of new types of hydrocarbon fuels and raw materials for other petrochemical industries.

Plasma-chemical technology for the decomposition of hydrogen sulfide opens up the possibility of creating a waste-free technology for the simultaneous production of gaseous sulfur and hydrogen.

III. LITERATURE SURVEY

One of the main advantages of this technology is its high selectivity of impact on the molecules of environmentally harmful impurities. Wherein Both the simplicity of purification technologies, the combination of the reaction chamber with existing technological schemes, and the relatively low energy costs for the purification process are attractive. Compared to other electrophysical methods, cleaning using a streamer corona does not involve solving complex engineering problems of ensuring a high resource of an energy source - an electron accelerator or a radiation source in an aggressive environment of the gas being cleaned. The presence of only corona electrodes in the gas is an undoubted advantage of this purification method compared to other electrical methods. The simplicity of the design of the reaction chamber determines the easy scalability of the technology by changing the number and diameter of the reaction chambers. The physics of the purification process is such that it is possible to decompose practically any gaseous impurities in the air in the concentration range from tens to tens of thousands ppm [4].

In experimental works, there is a tendency to use a pulsed corona discharge with nanosecond pulses. The conducted studies show that the use of a pulsed corona discharge for the decomposition of environmentally harmful gaseous compounds, such as hydrogen sulfide, is promising. The method makes it possible to almost completely decompose hydrogen sulfide into elemental sulfur and hydrogen.

The process is simple in hardware design and management.

The operability of a nanosecond streamer discharge was proven in the plasma-chemical technology developed by us in an experimental laboratory form. The effectiveness was evaluated and the energy intensity and energy consumption of the proposed technology for the purification of associated petroleum gas from hydrogen sulfide with the production of elemental sulfur at the production facilities of OAO Tatneft were determined [5].

The schematic diagram of the laboratory setup is shown in Figure 1.

A laboratory plant for the purification of associated petroleum gases from hydrogen sulfide with the production of elemental sulfur in the electric discharge of a streamer corona consists of the following main nodes [6-7]:

- unit for preparing a model gas mixture and supplying real gases;
- reactor assembly with streamer corona;
- nanosecond pulse generator unit;
- power unit, pulse transformer and ballast unit with voltage regulator;
- node for obtaining the final product - sulfur;
- sampling unit.

IV. METHODOLOGY

The laboratory unit is a prefabricated structure assembled on a metal frame with dimensions of 1185x900x210 mm, made of steel profiles with a section of 20x20 mm and steel panels with a size of 500x610 mm. The frame over the entire area

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is covered with a screen made of organic glass to protect against electric shock. Also, the installation has a protection system for turning on the high-voltage generator with an uncovered screen.

The main components of the installation are placed on steel panels, which can be conditionally divided into electric and gas parts. The main unit of the laboratory setup is the reactor. The streamer corona discharge reactor is designed to study on a laboratory scale the process of purification of associated gases from hydrogen sulfide into elemental sulfur in an electric discharge.

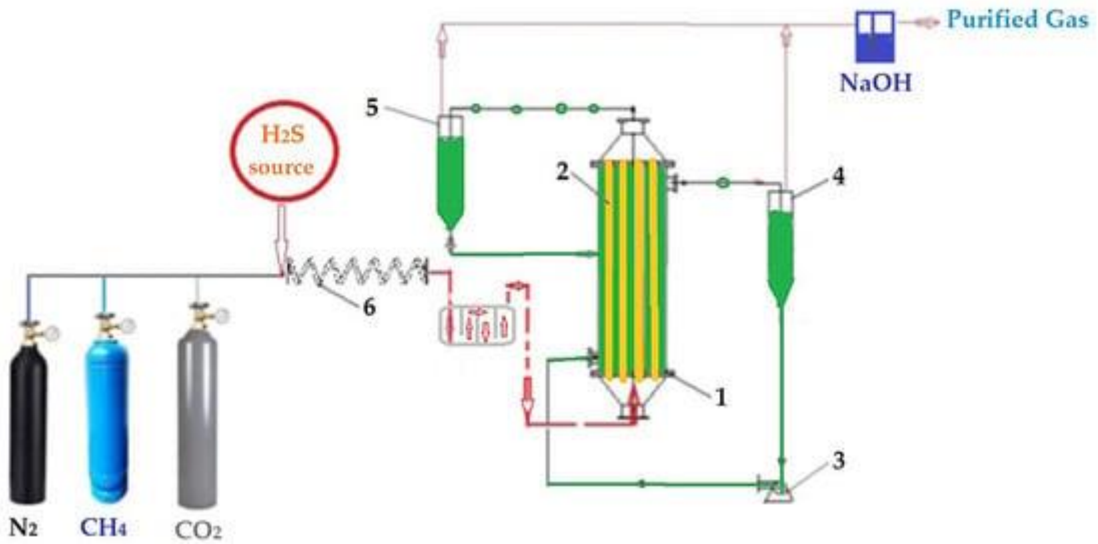

Figure 1. Schematic presentation of the laboratory installation for hydrogen sulfide sequestration from a gaseous mixture: *1-membrane contactor; 2-composite hollow fiber membranes; 3-pump for metal ions acidic solutions; 4 and 5-gas-liquid separator; 6-homogenization*

V. EXPERIMENTAL RESULTS

The reactor is a prefabricated structure, has the form of a cylindrical pipe with flanges. The tubular part consists of outer and inner cylinders (the main chamber of the reactor) made of heat-resistant glass. The cylinders are arranged coaxially.

The outer glass cylinder has a length of 300 mm and a wall thickness of 3 mm, closed from the sides with metal flanges. The diameter of the working area is 70 mm. On the walls

The outer cylinder has two holes for supplying power to the inner electrode. In the center of the flange there is a hole with a diameter of 14 mm through which the tubes pass to the inner cylinder. Tubes are intended for supply and removal of gases and formed products. Parallel to the main gas flow, there is a nitrogen gas flow entering the space between the cylinders. The function of gaseous nitrogen supplied to the cavity between the cylinders is to protect against fire in case of leakage of the main chamber of the reactor, and is also a dielectric. The gas pressure is kept within 1.3-1.4 atmospheres, which increases the breakdown voltage behind the main chamber of the reactor (Paschen's law).

The inner cylinder with a diameter of 42 mm has a length of 270 mm and is closed from the ends with round plugs made of organic glass. A central electrode made of a metal rod is attached to the plugs and metal tubes are connected. The volume of the inner cylinder is isolated from the outer space.

The overall height of the reactor assembly is 335 mm. The volume of the working zone of the reactor is 350 cm^3.

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The central electrode consists of a metal rod. On this rod are mounted round metal plates, pointed along the perimeter. Number of plates varies between 10-20 pieces. At both ends of the rod, clamps are installed, which are a round flat washer. In the washer, located on the side of the gas inlet, holes are made that serve to swirl the incoming flow.

Large-scale turbulence is given to the main gas to be cleaned, supplied to the central electrode, to optimize the destruction processes. After Process decomposition gas with decay products goes to the sulfur trap. The pressure difference at the inlet and outlet of the main chamber of the reactor is measured using a differential pressure gauge.

The laboratory unit has three main material streams: hydrogen sulfide, propane-butane gas mixture, nitrogen gas 99.5%. A hydrogen sulfide flow is supplied if a model gas mixture is being investigated.

Hydrogen sulfide is produced in a hydrogen sulfide generator, then fed into a mixer, where a propane-butane mixture flow is also supplied, which is supplied from a cylinder through an adsorption column.

An adsorption column with an inner diameter of 30 mm and a height of 170 mm is filled with a layer of silica gel with a fraction of 1.5-2 mm and a height of 130 mm and serves to remove propane-butane mixture of water vapor. Grids are installed in the lower and upper parts of the adsorption column to prevent entrainment of silica gel particles.

With the help of shut-off and control valves, the content of the components of the model hydrogen sulfide-propane-butane mixture varies. Gas for the determination of hydrogen sulfide content taken at the sampling point.

The prepared model gas mixture enters the electric heater. In the electric heater, the model gas mixture, reaching the required temperature (maximum possible value 120°C) is sent to the lower part of the inner cylinder of the reactor, in which the pressure is slightly above atmospheric. Under the action of a nanosecond corona discharge in the inner cylinder of the reactor, hydrogen sulfide contained in the test gas decomposes with the formation of hydrogen and elemental sulfur in the form of a highly dispersed mist. Thus, the products of the plasma-chemical reaction - sulfur and hydrogen, hydrocarbon gases and residues - exit from the reactor undecomposed hydrogen sulfide.

The mixture is then fed into a sulfur trap where gaseous sulfur is captured. The sulfur trap is a cylindrical container made of heat-resistant glass with a wall thickness of 2.0 mm, a height of 170 mm and an internal diameter of 85 mm. The container is filled with cleaned and dried sand with a fraction of 0.5-1.0 mm. The height of the sand layer is 140mm, at the top and bottom of the trap to prevent the entrainment of grains of sand broken glass with a particle size of 7.5–15.0 mm was laid with a layer height of 15 mm. It is necessary to note the peculiarity of the sulfur trap, which consists in the fact that the gaseous components from the reactor come from its upper to the lower part through a stainless steel tube mounted inside the cylindrical vessel, then rise up through a layer of sand and, already cleaned of fine sulfur, are removed through the upper branch pipe.

Elemental sulfur already in a viscous form is removed from the lower part of the apparatus.

After the sulfur trap, the gas passes through the sampling point, the safety tank. The container is a canister made of polymeric material with a capacity of 5 liters, filled in 3/7 parts of 10% of the mass. aqueous solution of sodium hydroxide. The gas bubbles through the alkali solution, and the hydrogen sulfide remaining undecomposed in the streamer discharge, when passed through the absorbent, is absorbed to form sodium salts. After the container, the remaining gas mixture is discharged into the exhaust ventilation. The flow of gaseous nitrogen is supplied from the cylinder, divided into two streams: one stream - the main one - is sent to the space between the inner and outer cylinders at an overpressure of 0.3-0.4 atm., The second one is 2-3% by volume. - is fed into the pulse generator to create a neutral insulating medium and then leaves as losses through leaks between parts of the installation. The first flow of gaseous nitrogen is introduced into

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the reactor through the lower part, and is discharged through a pipe from above. Similarly, an experiment is being carried out to study the process of destruction of hydrogen sulfide using a nanosecond streamer discharge in associated petroleum gases.

The electrical part of the installation works in the order shown below. When the protective screen is covered, the contact closes, which allows you to turn on the high-voltage pulse generator 5. Then, a voltage of 6 kV is supplied from the power source to the high-voltage pulse generator, where the voltage rises to 300 kV. To limit the charging current of the capacitor, voltage is applied through the ballast resistor block. The operating mode of the pulse generator is controlled from an external control panel. The high voltage current is fed through the high voltage cable to the input contacts of the streamer discharge reactor.

The high voltage applied to the reactor electrodes is formed using the following devices: a nanosecond pulse generator, a power supply unit, a pulse transformer, block of ballast resistors and voltage regulator. All devices have a protective earth.

The main parameters and dimensions of the laboratory setup are presented in the form of Table 1.

The proposed mechanism of hydrogen sulfide decomposition under the influence of a streamer discharge is as follows. The gas stream containing hydrogen sulfide passes through the reaction chamber, to which high voltage pulses are applied so short duration, so that the breakdown of the chamber does not occur. In this case, an intense pulsed corona discharge occurs in the chamber, which is the simultaneous development of a large number of thin luminous discharge channels - streamers. During the growth of streamers in the interelectrode gap, due to the high electric field strength, a large number of electrons are produced on the streamer heads, which have relatively high energy. In the zone of a pulsed nanosecond corona discharge, a strongly nonequilibrium plasma is formed, in which the ion temperature practically does not differ from the temperature of the surrounding neutral gas, and the overwhelming majority of the energy received from the source goes to increase the temperature of the electrons or, what is the same, their energy. Electrons possessing the necessary energy, in turn, are the main participants in plasma-chemical reactions leading to gas purification. Interaction these electrons with hydrogen sulfide molecules leads to the formation of reactive species such as HS^- and H^+, which is a prerequisite for purification.

Table 1
Main parameters and dimensions of the laboratory setup

Main parameters and dimensions		**Parameter limit range**
Productivity on the purified gas maximum, m^3/hour		0,1
The content of hydrogen sulfide in the purified gas, % wt.		0,1 – 10
Degree of purification of associated petroleum gas, %, not less than		99,6
Acid gas conversion degree, %, not less than		97,0
Gas pressure at the inlet to the installation, kg/cm^2 (abs.)		1,2
Power, kWt		0,1 – 0,4
Estimated efficiency of the installation in terms of electricity, %		87
Dimensions, mm	length	1500
	width	400
	height	1000
Total weight, kg		90

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VI. CONCLUSION AND FUTURE WORK

The scheme of the hydrogen sulfide dissociation process can be represented as follows:

$$H_2S \rightarrow H^+ + HS^- - 4\ eV$$
$$H^+ + H_2S \rightarrow H_2 + HS^- + 0,55\ eV$$
$$HS^- + HS^- \rightarrow H_2S + S + 0,39\ eV$$
$$S + H_2S \rightarrow H_2 + S_2 + 1,83\ eV$$

Secondary reactions (2–3) are fast exothermic processes with low activation energies; therefore, the rate of the process is determined by the limiting stage (1), the reverse endothermic reaction.

An operating facility has been created that can be used to test the technology for cleaning real gases using a corona plasma discharge in laboratory conditions.

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