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Industrial wastewater is often contaminated with hydrogen sulfide and sulfides. This poses significant risks to both the environment and human health and life as H2S is extremely toxic. Therefore, water purification from it is vital, and the choice of an effective desorber device is an urgent issue.

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This paper investigates the process of H2S desorption from wastewater in mass exchange devices with a continuous bubbling bed (DCBB), a column with falling plates (CFP), and a horizontal device with bucket-like dispersers (HDBD). To analyze the kinetic and technological characteristics of the process, the following indicators were selected: the product of the mass exchange coefficient on the contact surface of phases $(K \cdot F)$, reduced to 1 m^3 of the volume of the apparatus, and *the degree of hydrogen sulfide desorption.*

The most complete desorption of hydrogen sulfide occurs at pH≤*5. For practical needs, it is suggested to acidify the water to pH=5.5...6.0. It was established that the partial pressure of H2S increases linearly with increasing temperature, and an increase in salinity from 2...4 to 130...160 kg/m3 leads to its increase by 1.45...1.5 times.*

The best desorption indicators can be achieved in HDBD at pH=4.97. The efficiency of cleaning in CFP and DCBB is significantly affected by the specific air flow rate. The highest values (K \cdot *F) per 1 m³ that were achieved in desorbers are,* $mol/(s \cdot Pa \cdot m^3)$: HDBD – 1.94 \bullet 10⁻⁵, in CFP – 5.55 \cdot 10⁻⁶ *DCBB – 6.9•10-6. The ratio of the product (K•F) in HDBD to CFP is 3.5, and in HDBD to DCBB 2.8. It was possible to achieve the maximum degree of desorption of 37.8 % in DCBB; in CFP, this indicator is 74.1 %, and in HDBD – 77.7 %. Experimental studies have generally confirmed the effectiveness of using HDBD, and the results obtained under production conditions on real drainage and reservoir waters could find be practically implemented in hydrogen sulfide utilization technologies*

Keywords: hydrogen sulfide, water purification, mass exchange devices, desorption rate, bubbling, solution dispersion

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CHOOSING AN EFFICIENT MASS EXCHANGE APPARATUS FOR DESORPTION OF HYDROGEN SULPHIDE FROM RESERVOIR AND DRAINAGE WATERS

Andriy Helesh

Corresponding author Doctor of Technical Sciences, Professor* E-mail: andrii.b.helesh@lpnu.ua **Yaroslav Kalymon**

Doctor of Technical Sciences, Professor* *Department of Chemistry and Technology of Inorganic Substances Lviv Polytechnic National University S. Bandery str., 12, Lviv, Ukraine, 79013

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1. Introduction

Reservoir and drainage waters and liquid effluents from the chemical, food, mining, and other sectors of the national economy are often contaminated with hydrogen sulfide and sulfides. These compounds are always found in drainage and reservoir waters in contact with sulfur ores, and also occur in reservoir waters of those areas where there are no sulfur ores and in sulfate-containing waters of anaerobic treatment. During the extraction of minerals, reservoir waters contaminated with carbon disulfide and sulfides come to the surface. Especially a lot of water containing hydrogen sulfide is formed during the extraction of sulfur by the method of underground heating. The content of hydrogen sulfide in the wastewater under this method is $70...400$ g/m³. Industrial wastewater from ferrous metallurgy enterprises, coke plants, gas generating stations, oil production enterprises, artificial fiber productions, etc. is even more concentrated in terms of hydrogen sulfide, the content of the latter in it reaches $500 \frac{g}{m^3}$ [1, 2].

The presence of hydrogen sulfide in drainage and reservoir waters creates significant risks for both the environment and human health. H_2S is toxic to aquatic organisms. Its presence in water bodies can lead to a critical decrease in the content of dissolved oxygen, which leads to hypoxia of living organisms. For humans, exposure to H_2S , even at low concentrations (5 ppm), is dangerous and can cause respiratory problems, eye irritation, and headaches, while higher concentrations of $500-700$ ppm can be fatal, and at 1000–2000 ppm, death occurs instantly [3]. In addition, H2S dissolved in water can cause corrosion of metal pipes and concrete structures, etc. Hydrogen sulfide has a particularly negative effect on people who live near mining enterprises, in particular coal ones. Hydrogen sulfide reservoir waters, having entered drinking water aquifers, pose a health hazard to the population, including an increased risk of respiratory diseases [4].

Many methods have been proposed for cleaning liquids from hydrogen sulfide, among them methods in which the first stage is desorption of hydrogen sulfide occupy a prominent place [5]. Next, the desorbed hydrogen sulfide is oxidized to sulfur or to other sulfur-containing compounds. Taking into account the current trends of sustainable development, highly concentrated solutions of hydrogen sulfide should be considered as a complex raw material for the production of hydrogen and sulfur. In particular, work [6] carried out a critical analysis of methods for obtaining hydrogen from H2S (thermal dissociation, catalytic cracking,

multi-stage thermochemical cycles, photocatalytic splitting, etc.). Paper [7] also focuses on plasma-chemical and electrochemical methods of decomposition of hydrogen sulfide with the production of hydrogen.

Desorption and oxidation of hydrogen sulfide are carried out using various types of equipment [5], in many cases without proper justification. However, the intensity and its technical and economic indicators depend on the correct choice of the device that corresponds to the physical-chemical basis of this process. Therefore, the choice of an appropriate device for the disposal of hydrogen sulfide is an urgent issue, and considering the high toxicity of hydrogen sulfide, purification of water from it is vital.

2. Literature review and problem statement

Mass exchange devices with a fixed surface (film, spraying) and a contact surface that is formed during the movement of flows (plate; nozzle) are widely used in industry. This is due to the simplicity of the design, reliability of operation, low hydraulic resistance, etc. But the issue of intensification of their work remains unresolved, which is related to the peculiarities of mass exchange, which is determined by molecular diffusion, in the first case, and vortex diffusion, in the second type of devices. The reason for this is the fundamental impossibility of increasing the relative velocity of the phases without a noticeable increase in hydraulic resistance and droplet entrainment. That is, it can be assumed that these devices have exhausted their potential for intensification, which makes the corresponding research not expedient [8].

An effective way to intensify mass exchange and hydrogen sulfide oxidation is to supply the system with external energy directly to the contact zone. It is easiest to implement the supply of energy directly to the reaction system in electrochemical methods. These methods and their hardware implementation have a number of advantages: they are simple, compact, do not require reagents, are easy to manage, and can use renewable energy sources (solar, wind, etc.) for operation [7, 9]. At the same time, the issue of disposal of electrocoagulation and electro flotation sludges remained unresolved. One can get rid of these disadvantages by using electrooxidation methods. Thus, in work [10], the electrochemical oxidation of hydrogen sulfide on platinum electrodes was studied. Given the high cost of platinum, other materials were investigated as "cheap" electrodes. One of the directions in the intensification of the electrochemical oxidation of dissolved hydrogen sulfide is the use of catalytically active electrodes. In work [11], electrochemical cleaning was carried out on a stainless anode coated with nickel. In [12], the possibility of using sulfide solutions for anodic depolarization and reducing electricity consumption for the production of electrochemical hydrogen was investigated.

In all cases, it was possible to oxidize about $90\ \%$ of $\rm H_2S$ to sulfate. For the implementation of the above-described electrochemical processes, significant electricity consumption is required for the oxidation of sulfide ion (S^2) to sulfate ion, which is the main drawback of the method. Oxidation to sulfur is economically and energetically more profitable. To overcome this problem, work [13] proposed an interesting approach – an electrochemical method for cleaning with the help of a fuel cell that oxidizes the sulfide ion, producing electricity, and the product of oxidation is sulfur, which is widely used in many industries. Despite the advantages of this method, it has not yet been implemented industrially, which is related to the passivation of electrodes with sulfur and the need for its periodic removal from the electrodes. In addition, there are problems with the corrosion resistance of electrodes in the aggressive environment of the electrolyzer [5].

One option to overcome difficulties with effective supply of external energy to the system is to use centrifugal devices. The imposition of centrifugal forces contributes to the increase of the interfacial surface, the expansion of the stable range of operation under the phase inversion mode, and the reduction of droplet entrainment. This is the approach applied in the HiGee (High Gravity Technology) technology, which is used for many mass exchange processes in gas-liquid systems. It is implemented in rotating devices (rotating packed beds, RPB). In the role of the active volume, contactors of various designs are used, which are placed inside the rotor, which rotates at a high frequency. Depending on the structure of the contactor, phase interaction can be carried out in film or film-droplet modes. These devices have high productivity, the intensity of mass exchange can be 1–3 orders of magnitude higher than in conventional nozzle devices [14].

However, rotary devices have many disadvantages, in particular, they have a significant centrifugal load, which is why they impose special requirements for the rotating elements of the rotor. Accordingly, the diameter of the rotary devices is structurally limited, the distance between the rotor and the body or elements of the contactor nozzle does not exceed several centimeters. This leads to significant hydraulic resistance (2500–4000 Pa) and short contact time in the gas-liquid system. That is, the increase in the mass exchange coefficient is less than the increase in energy consumption. The reason for this is that with a high intensity of phase interaction, energy is evenly distributed in the volume of the device, while the resistance to mass exchange is concentrated near the interface of the phases, where it is advisable to direct the energy [8].

The solution to this problem is related to the redistribution of the supplied energy and localization of its action in the area of phase contact. To do this, it is necessary to ensure the operation of the device under non-stationary, discrete-pulse mode of supplying energy to the interface of the phases from the side of the liquid phase. This is the approach used when designing a horizontal apparatus with bucket-shaped dispersers (HDBD) [15]. In HDBD, the energy of shaft rotation is directly transmitted to the surface of the liquid through the dispersers. The absorption solution is repeatedly distributed in the volume of the gas flow, which allows the pulsating movement of the medium as a result of the periodic alternation of the stages of liquid capture and its dispersion. The device implements the cross movement of streams. The hydraulic resistance of HDBD remains at the level of the resistance of the spraying hollow devices, while the specific energy costs for dispersing the liquid are much lower compared to nozzles and do not exceed 50 W/m3.

The expediency of its use in dust collection systems [15] and $SO₂$ absorption [8, 16] has been theoretically substantiated. Experimental studies were also carried out under laboratory and production conditions, which confirmed the high efficiency of HDBD for these processes [17, 18]. Paper [19] reports the results of theoretical studies of $\rm H_2S, SO_2,$ and $\rm O_2$ absorption processes in mass exchange devices with a continuous bubbling bed (DCBB) and HDBD. It is calculated

that, with the same driving force of the process, the rate of absorption in HDBD is 2–3 orders of magnitude higher compared to DCBB, and the throughput is 54–74 times higher.

However, many assumptions were accepted in the calculation process, so practically obtained values may differ significantly from theoretical ones. In addition, the selection of industrial devices for gas desorption is guided by the properties of the absorbent (water) – gas system, mathematical models of the devices, and production experience of their operation. But the use of such methods for selecting devices is justified only under temperature regimes close to normal, provided that the absorbent and absorbent are practically pure substances, and the mathematical model adequately describes the process. When using highly concentrated H_2S water, with high salinity, at temperatures different from normal, objective difficulties arise because of the lack of relevant experimental data and empirical dependences. This is due to a number of reasons, in particular variable quantitative and qualitative composition of hydrogen sulfide-containing waters. Also, conducting research under industrial conditions, using enlarged laboratory or experimental industrial facilities, is a complex, long and expensive process. In addition, there are no single criteria for comparing the efficiency of mass exchange devices based on different physical processes.

All this gives reason to assert that it is expedient to conduct experimental studies under production conditions on the use of HDBD for desorption of H_2S from reservoir and drainage waters.

Industrial processes of desorption of hydrogen sulfide from water are carried out in devices with a continuous bubbling layer, columns with failed plates. Devices with nozzles (Raschig rings, etc.) and nozzles are usually not used because of their possible clogging with mechanical impurities. Therefore, these two types of devices were chosen for experimental studies and for comparison of their operational indicators with those of HDBD.

3. The aim and objectives of the study

The aim of our study is to establish the kinetic and technological parameters of hydrogen sulfide desorption from reservoir and drainage waters in mass exchange devices of various types. This will make it possible to compare the efficiency of mass exchange equipment and use experimentally obtained results to improve existing and devise new technologies for the disposal of highly concentrated hydrogen sulfide-containing waters.

To achieve the goal, the following tasks must be solved:

– to establish the influence of pH, salinity, and temperature on the value of partial pressure of H_2S above the surface of formation and drainage waters;

– to investigate the efficiency of desorption of hydrogen sulfide in HDBD, CFP, and DCBB;

– to compare the efficiency of desorption of hydrogen sulfide in HDBD, CFP, and DCBB, justify the choice of desorber.

4. The study materials and methods

4. 1. The object and hypothesis of the study

The object of our study is the process of desorption of hydrogen sulfide from reservoir and drainage waters. The sub-

ject of research is the effect of various types of mass exchange devices on the efficiency of hydrogen sulfide desorption.

The hypothesis of the study assumes that the use of a modern mass exchange apparatus HDBD can intensify the process of desorption of hydrogen sulfide from water.

During the research, it was assumed that the value of product of the coefficient and the mass exchange surface related to the volume of the investigated device (*K·F*) is a determining indicator in the process of choosing a mass exchange device for desorption of hydrogen sulfide from reservoir and drainage waters. In the course of research, the following simplifications were adopted: the degree of dissociation of hydrogen sulfide (*Хdiss*, %) depends on the temperature and pH of the water and does not depend on the salinity, the desorption temperature practically does not affect the value (*K·F*).

4. 2. Methodology for calculating the intensity of desorption in mass exchange devices

The process of hydrogen sulfide desorption from reservoir and drainage waters can be described by the mass exchange equation:

$$
w = K \cdot F \cdot \Delta P,\tag{1}
$$

where w is the desorption rate, mol/s;

 K – mass exchange coefficient, mol/(N·s);

 F – mass exchange surface, m²;

 ΔP is the driving force, N/m².

To compare desorption indicators in devices with a continuous bubbling layer, mechanical water spraying, and in columns with falling plates, it is necessary to mathematically describe the complex mass exchange processes of moving gas bubbles and water droplets. Mathematical models of mass exchange from gas and liquid phases, as well as calculations of the mass exchange surface, are based on many assumptions [19]. These assumptions make the calculations of the mass exchange coefficient and the contact surface between the phases very imprecise. Therefore, it is proposed not to calculate the mass exchange coefficient and the phase contact surface based on mathematical models for different devices but to operate with product of the mass exchange coefficient (K) by the phase contact surface area (F) . The value of this product (*K·F*) in different types of devices is related to 1 m3 of desorption volume. The speed of desorption (*w*) and the driving force of the process (Δ*Р*) were determined experimentally on the appropriate types of devices: HDBD, CFP, and DCBB. Then the value (*K·F*) is accurately and easily calculated: it can serve as an important characteristic during the selection of a mass exchange apparatus for desorption of hydrogen sulfide from reservoir and drainage waters.

4. 3. Methodology for determining the partial pressure of hydrogen sulfide above hydrogen sulfide-containing waters

It is possible to estimate the partial pressure of H_2S over sulfur-containing waters based on literature data, based on pH, temperature, and concentration of hydrogen sulfide in water. The partial pressure of hydrogen sulfide above water depends on the state in which it is in water (physically dissolved or chemically bound). Actually, the amount of physically dissolved hydrogen sulfide and the temperature determine the partial pressure of hydrogen sulfide above the solution.

To evaluate the relationship between different forms of hydrogen sulfide depending on pH and temperature, the dissociation constants of hydrogen sulfide of the first and second degree of dissociation in aqueous solutions at different temperatures were used.

At a certain pH value, all possible forms of hydrogen sulfide can be in equilibrium.

According to the material balance, one can write down:

$$
C_{\rm H_2S} = C_{\rm H_2S}^{\text{undiss}} + C_{\rm HS} + C_{\rm S^{2-}},\tag{2}
$$

or:

$$
a_{\text{H}_2\text{S}} = a_{\text{H}_2\text{S}}^{\text{undiss}} + a_{\text{HS}^-} + a_{\text{S}^2},\tag{3}
$$

where $C_{\text{H}_2\text{S}}\left(a_{\text{H}_2\text{S}}\right)$ is the total concentration (activity) of hydrogen sulfide in water, mol/dm3;

 $C_{\text{H}_2\text{S}}^{undiss}$ $\left(a_{\text{H}_2\text{S}}^{undiss}\right)$ – concentration (activity) of physically dissolved hydrogen sulfide, mol/dm3;

 C_{HS} *a*_{HS} \int – concentration (activity) of hydrogen sulfide ions, mol/dm3;

 $C_{S^2} \left(a_{S^2} \right)$ – concentration (activity) of sulfide ions in water.

Taking into account the dissociation constant of hydrogen sulfide in water and the value of the activity coefficient of undissociated hydrogen sulfide, which is equal to 1 (a small value of the molar concentration of hydrogen sulfide in water). The value of the molar concentration of hydrogen sulfide in the form of equation (3) can be written:

$$
a_{\text{H}_2\text{S}} = \frac{a_{\text{H}^+}^2 + K_1 \cdot a_{\text{H}^+} + K_2 \cdot a_{\text{H}^+}}{a_{\text{H}^+}^2},\tag{4}
$$

where:

$$
K_1 = \frac{a_{H^+} a_{H^S}}{a_{H^S}} , \quad K_2 = \frac{a_{H^+} a_{S^2}}{a_{H^S}}.
$$

The ratio $\frac{a_{\text{H}_2\text{S}}^{\text{undiss}}}{a_{\text{H}_2\text{S}}}$ represents the degree of undissociated $_{\rm H_2S}$

hydrogen sulfide molecules in water. The right-hand part of equation (4), in addition to the activity of hydrogen ions, implicitly includes the activity coefficients of ions that are formed during the dissociation of dissolved hydrogen sulfide. As is known, the activity coefficient of a certain ion is affected differently by the ionic strength of the solution. Taking into account the high mineralization (ionic strength) and the varied salt composition of the water solution to be purified, it is difficult to determine a priori the exact value of the activity coefficients of HS^- and S^2 ions. Therefore, we calculated the value of the degree of undissociated hydrogen sulfide molecules at different pH and temperature of water, assuming that the ionic strength of water is zero. The temperature dependences $K_1=f(T)$ and $K_2=f(T)$ are borrowed from the literature [20].

Reservoir hydrogen sulfide-containing waters differ not only in hydrogen sulfide content, temperature, pH, but also in the quantitative and qualitative composition of soluble salts. Thus, according to the results of preliminary exploration studies, the Yavoriv reservoir waters (YRW) of underground sulfur melting have relatively low mineralization $(2...4 \text{ kg/m}^3)$, caused mainly by sulfates and hydrogen carbonates of sodium and calcium, high temperature (50... 60 °C), their hydrogen sulfide content is $0.14...0.25 \text{ kg/m}^3$, pH=6.5...7. Gaurdak drainage wa-

ters (GDW) have high mineralization (130...160 kg/m³), mainly caused by sodium chloride, temperature 30...35 °C, hydrogen sulfide content in them is $0.28...0.30$ kg/m³, pH=5.5...7. Experimental studies were conducted to determine the influence of mineralization and temperature on the value of the partial pressure of hydrogen sulfide over the studied waters. The pH of the studied samples was adjusted to pH=5.

The study of the partial pressure of hydrogen sulfide over water was carried out on the installation shown in Fig. 1.

Fig. 1. Diagram of a laboratory setup for studying the partial pressure of hydrogen sulfide over water

The laboratory setup (Fig. 1) consists of two flasks 3 and 4, a magnetic stirrer 2, Drexel beakers (not shown in the diagram) filled with a solution of cadmium acetate, burette 5, and pH meter 1, the electrodes of which are mounted in flask 3. Methodology of the experiment was as follows: "hydrogen sulfide" water is poured into flask 3, which is placed on magnetic stirrer 2. Flask 3 was hermetically closed and connected with hoses to flask 4. The excess alkalinity of the solution was neutralized with HCl solution from burette 5, and the acidity of the liquid phase was determined based on the readings of pH meter 1. Flask 4 was alternately placed lower and higher than flask 3, as a result, the solution was poured from one container to another, washing the gas phase that was in the free volume of flasks 3 and 4. These manipulations were carried out until a stable pH value was established. This indicated the achievement of an equilibrium distribution of H_2S between the gas and liquid phases. After stabilization of the pH, the washing cycle was repeated 5–6 times, and only then were samples of the gas and liquid phases taken for analysis.

Analysis of gas and liquid phases for hydrogen sulfide content was carried out by the iodometric method [21].

4. 4. Research methodology for hydrogen sulfide desorption in a horizontal apparatus with bucket-shaped dispersers

The desorption of hydrogen sulfide from water was studied at the experimental installation for the treatment of reservoir waters of underground sulfur heating at the Yavoriv Sulfur Plant, the main device of which was a horizontal device with bucket-shaped sprinklers with a diameter of 0.5 m and a length of 0.7 m (Fig. 2)

Fig. 2. Image of the pilot experimental installation for studying the process of hydrogen sulfide desorption in HDBD

The research was conducted as follows. Hydrogen sulfide water from the water discharge wells of the underground sulfur heating with a flow rate of 1.5...4.5 m3/h entered the desorber. At the entrance to the desorber, reservoir water with pH=6.52 and a temperature of 50.5 °C was acidified with water containing sulfur dioxide. Aqueous solution of $SO₂$ was obtained as a result of combustion of desorbed H2S, with its subsequent capture in the HDBD absorber, the design of which is similar to the structure of the H_2S desorber (Fig. 2). This organization of the process makes it possible not only to save reagents but also to obtain a valuable product – finely dispersed colloidal sulfur, which is formed in sedimentation tanks during the slow interaction of $H₂S$ and SO₂. Water consumption for acidification was 3.0 m³/h, and its temperature was 22 °C. Air was supplied counter currently to the flow of water in the desorber with a flow rate of 10 m^3/h .

The process of desorption of hydrogen sulfide was studied under different air, reservoir and acidifying water consumption and other factors. Water consumption for reservoir water acidification was set so that the pH of the mixed waters before the desorber did not exceed 6.0...6.2, the pH of the water was monitored using a laboratory pH meter (Ezodo PL-700PC), the pH value at the entrance to the desorber was adjusted by changing reservoir water consumption.

The analysis of gas and liquid phases for the content of hydrogen sulfide was carried out by the iodometric method according to the procedure described in the literature [21].

4. 5. Research methodology for hydrogen sulfide desorption in a column with falling plates

The specificity of the design of columns with falling plates and obtaining data for modeling the mass exchange process require the manufacture of an apparatus of rather large sizes and significant consumption of both liquid and gas flows. It was impossible to carry out such studies under laboratory conditions. Therefore, the process of desorption of hydrogen sulfide from water in a column with falling plates was studied at the experimental unit, which was installed at the Gaurdak Sulfur Plant (Fig. 3).

Drainage water through a rotameter (not shown in the diagram) was fed into desorber 8. To increase the speed of H2S blowing, part of the water from absorber 3 was fed into the desorber. Next, the water entered container 6, where the process of H_2S and SO_2 interaction with the production of elemental sulfur took place. Water from the upper part of container 6 was fed into plate absorber 3 for SO_2 absorption by means of circulation pump 7. After desorber 8, the mixture of air and hydrogen sulfide was sent to furnace 1 for burning through droplet catcher 2. To maintain the temperature of 600 °C, diesel fuel and air were supplied to the furnace. After the furnace, the gas was directed to absorber 3, and after the absorber, the purified gas, passing through droplet catcher 4, was discharged into the atmosphere with the help of fan 5.

Fig. 3. Diagram of an experimental installation for the treatment of drainage water from hydrogen sulfide at the Gaurdak Sulfur Plant

> The desorber consisted of 8 plates with an inner diameter of 0.5 m and a height of 0.4 m each, between which were placed 7 plates with a thickness of 4·10–3 m with holes with a diameter of 1.10^{-2} m and a live section of 25 %. Viewing windows were installed on the second floor from the bottom. Samplers for gas and liquid phases are installed on each stage. The plates are enameled, and the plates are made of Х18Н10Т steel.

> During our studies, the pH of drainage waters was between 5.32 and 5.42, so acidification was not carried out. The studies were conducted at a drainage water temperature of 31 °C. The process of desorption of hydrogen sulfide from drainage water under the bubbling, emulsified, and wave modes of operation of the plates was studied. Experiments characterizing the operation of a column with falling plates in the bubbling and emulsifying modes are given in Table 4.

> Analysis of gas and liquid phases for hydrogen sulfide content was carried out by the iodometric method [21].

4. 6. Research methodology for hydrogen sulfide desorption in an apparatus with a continuous bubbling layer

The process of desorption of hydrogen sulfide in an apparatus with a continuous bubbling layer was studied under laboratory conditions on simulated reservoir waters of underground sulfur heating at the Yavoriv sulfur plant, acidified to pH=5. The main apparatus of the laboratory installation (Fig. 4) was column 2 with a diameter of 0.032 m and a height of 0.5 m.

At the bottom of the column, there was a soldered glass plate with holes that dispersed air to the size of bubbles of

2...3 mm. The installation included pressure vessel 1, compressor for air supply 3, rheometer for measuring air flow 4.

Experiments were performed at an air flow rate $(1.10^{-5} \text{ m}^3/\text{s})$, which corresponded to a gas content of ≈ 0.05 , that is, the flow rate at which the bubbling mode of mass exchange still occurs. Water consumption was changed from $3.\overline{3}\cdot 10^{-6}$ m³/s to $1.25 \cdot 10^{-5}$ m³/s. Other parameters were constant and were as follows: water height in the

desorber – 0.4 m; desorption temperature – 21 °C; the content of hydrogen sulfide in simulated drainage water – 190 g/m³.

Analysis of gas and liquid phases for hydrogen sulfide content was carried out by the iodometric method [21].

Fig. 4. Diagram of a laboratory installation for the study of desorption of hydrogen sulfide from reservoir waters of underground sulfur heating at the Yavoriv Sulfur Plant

4. 7. Methodology for comparing the efficiency of mass exchange devices

The comparison of effectiveness of using HDBD, CFP, and DCBB in the process of desorption of H2S from drainage and reservoir waters was carried out by the value $(K \cdot F)$ related to 1 m³ of the device volume. The depth of desorption of hydrogen sulfide from water was also compared by the degree of desorption.

5. Results of hydrogen sulfide desorption research

5. 1. Results of investigating the values of partial pressures of hydrogen sulfide over waters containing hydrogen sulfide

The rate of desorption of hydrogen sulfide from hydrogen sulfide-containing waters depends on the form of sulfide compounds, their concentration, temperature, and the content of dissolved salts. The total influence of the factors listed above is expressed by the partial pressure of hydrogen sulfide above water, which directly determines the driving force of de-

sorption. Therefore, the value of the partial pressure of hydrogen sulfide over hydrogen sulfide-containing waters, depending on their characteristics, is absolutely necessary for calculations and development of the technological mode of desorption.

The results of theoretical calculations are summarized in Table 1, and experimental studies are presented graphically (Fig. 5).

Table 1

Dependence of the degree of undissociated hydrogen sulfide (*Хundiss*, %) on the temperature ($t_{des.}$, °C) and pH of water

No. of entry	$^{\circ}C$ t_{des}	Dissociation constants H_2S		X_{undiss} , %								
		$K_1 \cdot 10^7$	K_2 -10 ¹²	$\rm pH_{des.}$								
				4	5	6			9	10		
		0.33	0.10					99.97 99.67 96.81 75.19 23.10	2.94	0.30		
$\overline{2}$	31	0.95	0.72	$99.90 \mid 99.06 \mid 91.32 \mid 51.28 \mid$				8.52	1.42	0.10		
3	50	1.23	1.22				99.88 98.78 89.05 44.84	7.52	0.80	0.08		

Note: $t_{des.}$ – desorption temperature, °C; pH_{des.} – pH of the water in the desorber.

Fig. 5. Dependence of the partial pressure of hydrogen sulfide above YRW and GDW on temperature and mineralization at pH=5

> For ease of use, the obtained dependences were mathematically treated using the method of least squares. The resulting equations and values of approximation probability values (R^2) are given in Table 2.

> > Table 2

Approximation equation of the experimentally obtained results from studying the effect of temperature on the partial pressure of hydrogen sulfide above water (Fig. 1)

One can see from the experimentally obtained results that the partial pressures of H_2S over all solutions increase linearly with increasing temperature (Fig. 5), and the resulting dependences of the partial pressure of H_2S on temperature are described with high probability by linear equations (Table 2).

5. 2. Results of investigating hydrogen sulfide desorption processes in mass exchange devices

The results of our experiments reflecting the desorption process in HDBD, CFP, and DCBB are given in Tables 3–5 respectively.

In addition to experimentally establishing the most important indicator of the product desorption process (*K·F*), two important technological indicators were also determined for all devices: the degree of desorption and specific air consumption. Also, for each device, Tables 3–5 give parameters that are important for their technological use for: HDBD – conditional irrigation density $(W, m^3/m^2)$; CFP – operating mode (bubbling, emulsified); DCBB – liquid level in the desorber (*h*, m).

5. 3. Results of comparing the efficiency of hydrogen sulfide desorption in the studied mass exchange devices

The highest values $(K \cdot F)$ per 1 m³ obtained in each desorber were used to compare the efficiency of the devices. The results of our research are given in Table 6.

Comparing the results of the research (Table 6), one can state that the values of (*K·F*) for CFP and DCBB are 3.5 and 2.8 times smaller compared to HDBD, respectively. The values of the degree of desorption for HDBD and CFP are comparable, and for DCBB – significantly lower. Comparing the *Xdes*. values obtained in DCBB with the values from HDBD and CFP is not completely correct since the desorption process, due to the limitations associated with research on large-scale factory installations, took place at different temperatures. One can see from the results described above (Fig. 5, Table 2) that the partial pressure of H_2S above the surface of the studied waters increases in direct proportion with increasing temperature. Thus, for Yavoriv reservoir waters, when the temperature increases from 21 °C to 31 °C, the partial pressure increases by 1.39...1.42 times.

Table 3

Results of investigating the hydrogen sulfide desorption process in a horizontal apparatus with bucket-shaped dispersers

No. of entry	Desorption parameters								Desorption indicators				
	$Q_{wat.}$	$\mathrm{pH}_{des.}$	$t_{des.}$	$[H_2S]_{ent.}$	$[H_2S]_{out.}$	q_{air}	W	ΔP	$v \cdot 10^3$	$K\text{-}F\text{-}10^6$	$(K \cdot F) \cdot 10^5$	$X_{des.}$	
	1.5	4.86	31	52.0	12.6	2.23	22.9	567	1.45	2.55	1.86	75.8	
\mathcal{D}	2.0	4.97	33	62.4	13.9	2.00	25.5	744	1.98	2.66	1.94	77.7	
	3.0	5.48	35	78.0	26.4	1.67	30.6	1042	2.53	2.43	1.77	66.2	
4	4.0	6.0	38	89.1	34.0	1.43	35.7	1338	3.15	2.36	1.72	61.8	
	4.5	6.1	39	93.6	41.0	1.33	38.2	1621	3.22	1.99	1.45	56.2	

Note: H2S content in reservoir water is 156 g/m3; reservoir water pH – 6.52; reservoir water temperature – 50.5 °C; acidifying water temperature – 22 °C; water consumption for acidification – 3.0 m³/h; air consumption in the desorber – 10 m³/h; Q_{wat.} – consumption of drainage *(reservoir) water, m*³/h; $[H_2S]_{ent.} - H_2S$ content in the water at the entrance to the desorber, g/m^3 ; $[H_2S]_{out.} - H_2S$ content in the water at *the outlet of the desorber, g/m3; qair – specific air consumption, m3 of air/m3 of water; W – conditional irrigation density, m3/m2·h;* ∆*P is the driving force of desorption, Pa;* υ*des. – desorption rate, (mol/s); K·F is the product of the mass exchange coefficient (K) by the mass exchange surface (F), mol/(Pa·s); (K·F) is the product of the mass exchange coefficient (K) on the mass exchange surface (F), reduced to 1 m3 of the desorber, mol/(s·Pa·m³);* $X_{des.}$ *– degree of desorption, %.*

Table 4

Table 5

Results of investigating the process of desorption of hydrogen sulfide in a column with falling plates

Note: B – bubbling; E – emulsified.

Results of investigating the hydrogen sulfide desorption process in an apparatus with a continuous bubbling layer

Note: h is the liquid level in the desorber, m.

It can be predicted that the degree of desorption will also increase within the same limits, however, its value will be smaller compared to HDBD and CFP. Therefore, to compare the efficiency of desorption of hydrogen sulfide in HDBD, CFP, and DCBB, it is more appropriate to use the value of (*K·F*). When calculating (*K·F*), the value of partial pressures of H_2S is taken into account, so the temperature has practically no effect on its value.

Analyzing the findings obtained in HDBD, CFP, and DCBB, it was established that the determining factor of the desorption process is pH. Thus, for HDBD, with a stable consumption of water for acidification and with an increase in the consumption of drainage water, the pH of the mixture naturally increases from 4.86 to 6.10. The best desorption rates $((K \cdot F) = 1.94 \cdot 10^{-5} \text{ mol/(s} \cdot \text{Pa} \cdot \text{m}^3), X_{des} = 77.7 \%$ were achieved at pH=4.97, which is explained by the fact that

Table 6

Results of comparing the efficiency of hydrogen sulfide desorption in HDBD, CFP, and DCBB

No. of entry	Device type	$Q_{wat.}$	$\rm pH_{des.}$	$t_{des.}$	$[H_2S]_{ent.}$	q_{air}	ΔP	$(K \cdot F) \cdot 10^6$	$X_{des.}$
	HDBD	2.0	4.97	33	62.4	2.00	744	19.4	77.7
2	CFP	15.0	5.41	31	304	6.0	8237	5.55	67.4
		10.0	5.36	31	296	12.0	7978	4.12	74.7
3	DCBB	8.33	5.0	21	190	1.2	4302	6.90	20.5
		2.50	5.0	21	190	4.0	4280	3.81	37.5

6. Discussion of results related to the study of desorption of hydrogen sulfide from reservoir and drainage waters

One can see from our calculations (Table 1) that a sharp transition from the non-dissociated form of hydrogen sulfide to the dissociated form is observed within rather narrow pH ranges and depends on the temperature. Thus, for hydrogen sulfide-containing waters at a temperature of 31 °C, when the pH changes from 6 to 8, the proportion of physically dissolved hydrogen sulfide decreases from 91.3 % to 8.5 %.

The results of calculations show that at $pH\leq 599\%$ of hydrogen sulfide in water is in the form of undissociated molecules; at pH≤7, approximately half of the hydrogen sulfide molecules are in undissociated form, the rest in dissociated form. That is, the most favorable conditions for hydrogen sulfide degassing will be pH≤5. At the same time, there is not always a technological need for deep desorption of hydrogen sulfide. For example, under the "wet Claus" method, it is necessary to blow off only one third of the hydrogen sulfide to oxidize it to SO2. That is, the depth of the desorption process can be controlled by changing the pH. When the pH value of water changes from 5 to 8, the partial pressure of hydrogen sulfide over hydrogen sulfide-containing water decreases by 10...12 times. Therefore, it is impractical to desorb hydrogen sulfide from waters at pH values ≥7 without first acidifying them to pH values equal to 5.5...6.0.

The partial pressure of H_2S above reservoir and drainage waters at $pH=5$ (Fig. 1) increases linearly with increasing temperature. This experimentally confirmed that at pH≤5, regardless of the quantitative and qualitative composition of the studied waters, almost all of the sorbed H_2S is in a physically bound state.

As a result of our experimental studies, it was established that the partial pressure of hydrogen sulfide above hydrogen sulfide-containing waters under the same conditions (temperature, pH) above the drainage waters at the Gaurdak sulfur plant is 1.45...1.5 times higher than the pressure above the Yavoriv reservoir water. (Fig. 5), which is explained by a higher salinity and, accordingly, a decrease in the solubility of hydrogen sulfide. There are no results of similar studies in the literature.

within pH=4...5, the content of physically bound H2S remains practically unchanged at 99.97...99.67 % (Table 1). At the same time, the driving force of the process increased by a third (567...744 Pa).

In the case of using CFP, the specific air flow, and the mode of conducting the process also play a significant role. Under the emulsified mode at a specific air flow rate of $6 m_{air}^3 / m_{water}^3$, the maximum value of $(K \cdot F) = 5.55 \cdot 10^{-6}$ mol/(s·Pa·m³) was reached, and the greatest depth of cleaning (**Х**=74.7 %) can be achieved at a specific air flow 12 m_{air}^3/m_{water}^3 . Such results are explained by the fact that the value of the contact surface

for a certain type of falling plates is determined by the ratio of air flow to liquid flow and has an optimal value. That is, the mass exchange takes place most intensively at a specific air flow rate of 6 m_{air}^3/m_{water}^3 , and the increase in the degree of desorption at a specific air flow rate of $12 \text{ m}^3_{\text{air}}/\text{m}^3_{\text{water}}$, is explained by a better "blowing" of hydrogen sulfide as a result of an increase in the air volume.

In DCBB, as in the previous case, the efficiency of the process largely depends on the specific air consumption, so, when this indicator increases from 0.8 to 4 m_{air}^3/m_{water}^3 , the degree of desorption naturally increases from 13.7 to 37.5 %. At the same time, the maximum value of $(K \cdot F) = 6.90 \cdot 10^{-6}$ mol/(s·Pa·m³) was reached at a specific air flow rate of 1.2 $\mathrm{m}^3_{\text{air}}/\mathrm{m}^3_{\text{water}}$.

The highest values of (KF) per 1 m³, which were achieved in desorbers, are, mol/(s·Pa·m³): HDBD – 19.4·10⁻⁶, in CFP – 5.55 \cdot 10⁻⁶, and in DCBB – 6.9 \cdot 10⁻⁶ mol/(s \cdot Pa) (Table 6). That is, to achieve the desired degree of desorption, the desorption volume of CFP should be 3.5 times greater than the desorption volume of HDBD, and 2.8 times that of DCBB. At the same time, it should be noted that the theoretically calculated efficiency of mass exchange in HDBD was 2–3 orders of magnitude higher compared to DCBB. This discrepancy between practically obtained results and theoretically calculated results can be explained by the effect of pH and H2S concentration. Thus, in the course of the research, the pH value of the water was: HDBD – 4.86...6.1; KTP – 5.32...5.42, and for DCBB – 5. Accordingly, the H₂S content in the initial water was, $g/m³$: HDBD – 156; KTP – 290...304 and DCBB –190. As a result, the driving force of the process in CFP was 5.3...13.8, and in DCBB – 2.6...8.0 times greater in comparison with HDBD. Despite the comparable values of the degree of desorption for HDBD and CFP, it should be noted that the specific air consumption for hydrogen sulfide desorption was 1.3...2.2 and 4.7...12.0, respectively. That is, under all other conditions being the same, the content of H_2S in the gas phase at the exit from CFP will be 3.6...5.5 times lower compared to HDBD.

In other words, on the basis of our results, one can state that HDBD is significantly more efficient than the devices that have become widely used in industrial processes (CFP and DCBB).

The results reported for HDBD are comparable to the characteristics of HiGee devices [14]. At the same time, "HiGee" type devices are characterized by a significant centrifugal load on the rotor shaft, which is due to the high frequency of rotation of the shaft $(5.0...13.3 \text{ s}^{-1})$ and high hydraulic resistance (2500–4000 Pa). Unlike "Hi-Gee", the HDBD frequency of rotation of the shaft is not high, $2.6...3.8$ s⁻¹, and the hydraulic resistance does not exceed 120...300 Pa, which allows the desorption process to be carried out efficiently at significantly lower energy costs. This became possible due to the fact that, unlike HiGee, the external energy is transferred directly to the liquid, giving its drops significant kinetic energy and initial speed, which significantly reduces the resistance from the gas film. In addition, a pulsating movement of the medium is created due to the alternation of phases of liquid capture and spraying.

Our research carried out under industrial conditions at large-scale factory and laboratory facilities showed high efficiency of HDBD for H2S desorption and is a reliable basis for industrial introduction of HDBD in processes of purification of H_2S -containing waters.

It should be noted that the study was conducted for two types of hydrogen sulfide waters. This leads to certain limitations in the use of our results since the sample is insufficient for the possibility of deriving empirical dependences. The main shortcomings of the research are that the experiments with each mass exchange device were carried out on waters of different composition, under different technological regimes, on installations of different desorption volumes. Accordingly, it is not entirely correct to compare the obtained specific indicators since our results are difficult to scale. As mentioned above, these disadvantages and limitations are due to the complexity and high cost of research on an industrial scale. At the same time, these limitations open up new prospects for further research, in particular, it is advisable to investigate the influence of salt content on the desorption process in mass exchange devices of the same working volume.

7. Conclusions

1. Our theoretical and experimental studies on the influence of pH, temperature, and salinity on the partial pressure of H2S have made it possible to choose the values of the process management parameters necessary for desorption. It is technologically advisable to pre-acidify the water to pH=5.5...6.0, and for deep desorption to pH≤5. With an increase in salinity from $2...4 \text{ kg/m}^3$ (YRW) to $130...160 \text{ kg/m}^3$ (GDW), the partial pressure of H_2S above the water surface increases by 1.45...1.5 times.

2. As a result of experimental research, it was established that the pH of water has a decisive influence on the desorption process, in particular, the best indicators of the process in HDBD $((K \cdot F)=1.94 \cdot 10^{-5} \text{ mol/(s} \cdot \text{Pa} \cdot \text{m}^3), X_{des} = 77.7 \%)$ were achieved at pH=4.97. At the same time, the efficiency of cleaning in CFP and DCBB is significantly affected by the hydrodynamics of the process, in particular, the specific air flow rate.

3. The highest values $(K \cdot F)$ per 1 m³, which were managed to be achieved in desorbers, are mol/(s·Pa·m3): HDBD – $1.94 \cdot 10^{-5}$, in CFP – 5.55 $\cdot 10^{-6}$, and in DCBB – 6.9 $\cdot 10^{-6}$. The ratio of the product (*K·F*) in HDBD to CFP is 3.5, and such a ratio in HDBD to DCBB is 2.8. At the same time, it was possible to achieve the maximum degree of desorption of 37.8 % in DCBB, while in CFP this indicator is 74.1 %, and in HDBD – 77.7 %. As the results of the research showed, HDBD is a highly efficient mass exchange device for the process of desorption of hydrogen sulfide from drainage and reservoir waters. Its kinetic and technological indicators exceed those of CFP and DCBB. In addition, HDBD is characterized by very low hydraulic resistance and has a significant potential for intensification of the desorption process.

Conflicts of interest

The authors declare that they have no conflicts of interest in relation to the current study, including financial, personal, authorship, or any other, that could affect the study, as well as the results reported in this paper.

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Data availability

All data are available, either in numerical or graphical form, in the main text of the manuscript.

Use of artificial intelligence

The authors confirm that they did not use artificial intelligence technologies when creating the current work.

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