Hydrogen argonide {H_2Ar}: artifact or an objective reality?

A. N. Startsev

Retiree, 630090 Novosibirsk, Russia

E-mail: anatolii.startsev@gmail.com

The decomposition of hydrogen sulfide on solid catalysts at room temperature revealed a unique phenomenon: when using argon or nitrogen as a carrier gas, molecular hydrogen is not detected in the reaction products, despite the high conversion of H_2S and the presence of gaseous sulfur as the main product of the reaction. A hypothesis is proposed about the possibility of interaction of atomic hydrogen formed (*in statu nascendi*) at the moment of its origin in a catalytic reaction with molecules of carrier gas – argon or nitrogen adsorbed on the catalyst. The hypothesis is based on the previously developed conception on the mechanism of chemosynthesis of organic matter from (H_2S + CO_2) with the participation of sulfur bacteria, where H_2S is a supplier of hydrogen and energy for the activation of chemically inert molecules of carbon dioxide. To verify and substantiate this hypothesis, purposeful experimental studies by modern physical and chemical methods with the use of theoretical calculations of non-equilibrium thermodynamics of the open system and the mechanism of catalytic stages of interaction of atomic hydrogen with adsorbed argon and nitrogen molecules are necessary. Currently, the author does not have such an opportunity, so the author will be very grateful to any expressions of interest from the scientific community and is ready to share all available information for the implementation of the goals.

1. Introduction.

The reaction of low-temperature catalytic decomposition of hydrogen sulfide

$$2\,H_2S \xrightarrow{\text{metal, 25oC}} 2\,H_2\uparrow + {}^3S_2^{(gas)} \qquad (1)$$

occurs on metal catalysts at room temperature to form hydrogen and diatomic gaseous sulfur in the ground triplet state [1,2] . To study the reaction (1), two flow setups were built at the Boreskov Institute of Catalysis (Fig. 1), in which the gas mixture at the outlet of the reactor was analyzed by two methods:

1. on a gas chromatograph LHM-8 or Zwet-500 by periodic sampling with a 6-way volumetric feeder;

2. in a continuous mode on the hydrogen gas analyzer INV-8 “Zircon” with the thermo-chemical sensor.

Initially, the chromatogram showed the appearance of the only poorly resolved peak with a shoulder in the region of smaller retention times (Fig. 2). Later a chromatographic column was developed, which was able to separate the two peaks

[1] - Startsev AN, Kruglyakova OV, Ruzankin SPh, Bulgakov NN, Chesalov YuA, Kravtsov EA, Jeivot VI, Larina TV, Paukshtis EA. Peculiarity of low temperature catalytic decompositionof hydrogen sulfide. Russ J Phys Chem. 2014;88:943–956 (in Russian).. DOI: 10.7868/S004445371406034X

[2] - Startsev AN. Low-Temperature Catalytic Decomposition of Hydrogen Sulfide into Hydrogen and Diatomic Gaseous Sulfur. Kinetics Catalysis. 2016; 57: 511–522. DOI: 10.1134/S002315841604011X

with a good resolution (Fig. 3). The first peak was attributed to hydrogen, since its retention time coincided well with the retention time of molecular hydrogen used in calibration, while the second peak was attributed to gaseous diatomic sulfur.

The curves of the signal change on the thermo-chemical sensor of the “Zircon” analyzer are shown in Fig. 4. Thermo-chemical sensors are based on the measurement of the thermal effect of the reaction of catalytic oxidation of hydrogen containing gas, they are used to determine the concentrations of combustible gases. Its essence consists in measuring the thermal effect (additional temperature increase) from the oxidation reaction of combustible gases and vapors on the catalytic active element of the sensor and further transformation of the received signal.

In both cases, the formation of hydrogen as a product of the reaction of hydrogen sulfide decomposition by reaction (1), seemed to be the most likely alternative, beyond doubt. This is confirmed by the data of mass spectrometric analysis (Fig. 5), which, along with the hydrogen, was the diatomic gaseous sulfur, for the first time it was discovered in our research[3].

2. The flow set-up in "SKTB Katalizator»

As a matter of fact the similar flow setup was built in “SKTB Katalizator” (Fig. 6). The main difference was that for the quantitative analysis of hydrogen the gas analyzer "Test-1", produced by JSC "Boner", Novosibirsk, was used, which is a sensor with an electrochemical cell that allows selective analysis of only hydrogen in any gas mixture. The software of the device allows you to determine the amount of detected hydrogen, which is fundamentally important for the preparation of the material balance of the process under study.

The analyzed gas reacts chemically with the electrolyte filling the cell. As a result, charged ions appear in the solution, an electric current proportional to the concentration of the analyzed component in the sample begins to flow between the electrodes. The electrical sensor processes the resultant electrical signal. For each specific gas, a sensor is developed that allows you to selectively control the concentration of only this gas in any gas mixture. Therefore, in our case, we measure only the concentration of hydrogen.

3. Calibration of the hydrogen analyzer "Test-1".

Initially, using a calibrated gas mixture (nitrogen + hydrogen) with known hydrogen content, the required hydrogen concentration is set on the gas analyzer scale. Gas flows are set by gas flow controller (RGS), measured by a foam flow gauge and recorded on the PC screen.

To calibrate the analyzer, the flow of argon is passed through the detector until the zero mark on the scale of the analyzer, and then the flow of hydrogen is introduced into the flow of argon in a known proportion (Fig. 7). The gas flow rate at the outlet of the analyzer corresponds to the sum of the supplied flows (H_2 + Ar) at the inlet to the system. Hydrogen in the gas mixture is recorded by the analyzer's electrochemical sensor and displayed on the PC as its volume concentration in the gas stream. The concentration of hydrogen in the gas stream corresponds to its theoretical value. After a certain period of time, the hydrogen supply is stopped and the analyzer is blown with argon to zero reading of the device. The total amount of hydrogen supplied is

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[3] - Startsev A.N.. *Diatomic sulfur: a mysterious molecule.* J. Sulfur Chemistry. 2019, v. 40, No 4, p. 435 – 450. https://doi.org/10.1080/17415993.2019.1588273

determined by the integration of the area under the hydrogen evolution curve in accordance with the analyzer software. As follows from the calibration experiments (Fig. 7), the error in determining the volume of registered hydrogen does not exceed 3 % of its volume actually submitted using independent means of regulating gas flows.

A similar calibration technique was used to determine the volume of hydrogen fed through a real catalytic system, including a reactor with a catalyst under a layer of monoethanolamine (MEA) aqueous solution, an absorber with zinc acetate, and a protective layer of solid absorber (Fig. 8). As it was shown, when passing the gas mixture (argon + hydrogen) through the catalytic system, no hydrogen losses are observed, and the measurement error of its volume determined by the integration under the curve of its detection does not exceed 3 % of the theoretically submitted (Fig. 8).

Conclusion. Molecular hydrogen is not absorbed (does not dissolve) in the rubber tubes connecting the absorbers, and the technological solutions and the sinks absorb hydrogen reversibly.

4. Gas-phase decomposition of hydrogen sulfide

In studies performed at the Boreskov Institute of Catalysis, the reaction was carried out by passing a mixture of hydrogen sulfide with argon through the catalyst bed, while the conversion of hydrogen sulfide in the flowing gas phase mode did not exceed 20 %. A synchronous appearance of two reaction products was observed on the chromatograph (Fig. 3).

The scheme of the experimental setup in the “SCTB Katalizator” is shown in Fig. 9. After the catalytic reactor, an absorber with Al_2O_3 is installed to absorb sulfur gas, the unreacted hydrogen sulfide is extracted from the flow in the absorber with zinc acetate, and the gas flow, after passing through the protective layer of the absorber, is detected on the hydrogen analyzer "Test-1". However, it turned out that, despite the high conversion of hydrogen sulfide (20 -30 %), as well as the color of aluminum oxide in yellow (both of these criteria prove the reaction (1) [1,2]) , the hydrogen concentration at the outlet of the installation, detected by the gas analyzer "Test-1", did not exceed 0.1 %. This pattern was repeated with surprising constancy both in the case of using argon as a carrier gas and when replacing it with nitrogen.

Therefore, a different method of implementation was used. After purging the system with an inert gas, the carrier gas supply was stopped, and the reactor was filled with pure hydrogen sulfide until it appeared in the absorber with zinc acetate. As the reactor is filled with hydrogen sulfide, hydrogen appears at the outlet of the installation, the concentration of which (1 - 1.5 % vol.) significantly exceeded its concentration in the flow mode (see above). Then hydrogen sulfide was stopped, argon or nitrogen was fed again, and the whole system was purged to remove unreacted hydrogen sulfide (Fig. 9). Despite the high conversion of hydrogen sulfide, the amount of hydrogen released was only 17 % of the expected amount, based on the conversion of hydrogen sulfide. This experiment excludes the assumption that the active hydrogen is dissolved in the rubber tubes connecting the absorbers.

Conclusion. The absence of hydrogen at the outlet of the setup may mean that the atomic hydrogen formed (in statu nascendi) at the time of its formation in the catalytic reaction either dissolves in the rubber tubes connecting the absorbers or interacts with the carrier gas. However, the experiment in Fig. 9 indicates in favor of the latter assumption.

5. Three-phase mode of hydrogen sulfide decomposition

Without any doubt, the immersing solid catalyst in a liquid layer capable of dissolving hydrogen sulfide and sulfur[4] is a fundamental solution to the problem of implementing the process of low-temperature decomposition of hydrogen sulfide on an industrial scale. The obvious advantages of this method have been repeatedly discussed in the scientific literature [2,5,6], however, as it turned out, for the realization of all the potential possibilities of this method in full, additional fundamental research is necessary.

The main unexpected and difficult to explain problem that arose during the decomposition of hydrogen sulfide on solid catalysts under a layer of an aqueous solution of monoethanolamine (MEA) was the lack of hydrogen at the outlet of the catalytic unit. To understand this phenomenon, special experiments were performed (Fig. 10,11), when nitrogen was used as a carrier gas.

Stainless steel chips were placed in a glass adsorber equipped with a magnetic stirrer (Fig. 10), filled with an aqueous solution of MEA and purged with an inert gas to remove the dissolved air 5. Then a stream of hydrogen sulfide was introduced into the carrier gas stream. The criterion for the activity of catalysts was the absence of hydrogen sulfide in the absorber with zinc acetate and staining the solution in yellow. The flow of hydrogen sulfide was passed up to his appearance in the absorber with zinc acetate.

After stopping the supply of hydrogen sulfide and purging the system with an inert gas, only traces of non-reacted hydrogen sulfide were found in absorbers with zinc acetate, which allowed us to conclude that the conversion of H_2S is close to 100 %. It is very important that in the process of supplying hydrogen sulfide, the rate of gas flow at the outlet of the installation corresponded to the rate of supply of the carrier gas to the installation, while an additional increase in the total flow rate due to hydrogen sulfide was not observed. According to the data of the analyzer "Test-1", the amount of hydrogen released at the outlet of the plant was less than 1 % of the amount of hydrogen sulfide supplied (Fig. 10). Therefore, it was hypothesized that the "active" hydrogen released during the decomposition of hydrogen sulfide reacts with the carrier gas nitrogen.

To test this hypothesis, a litmus test was placed in the nitrogen gas stream at the reactor outlet (Fig. 11). Although the MEA has an alkaline reaction, the colouring of the litmus test in the gas stream has not changed, apparently due to the very low partial pressure of the MEA vapors. At the same time, the introduction of a litmus test into the gas stream during the decomposition of hydrogen sulfide clearly indicates the existence of an alkaline component $\{N_xH_y\}$ in the gas stream, for example, N_2H_2, N_2H_4, NH_3.

Most of the experiments in "SCTB Katalizator" were performed using argon as a carrier gas, as it was necessary to reproduce the results obtained earlier at the Institute of Catalysis. In a typical experiment (Fig. 12) on a chip stainless steel under a layer of

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[4] - Старцев А.Н., Пашигрева А.В., Ворошина О.В., Захаров И.И., Пармон В.Н., // Патент РФ № 2,261,838, опубл. 10.10.2005

[5] - Startsev AN, Kruglyakova OV, Chesalov YuA, Paukshtis EA, Avdeev VI, Ruzankin SPh, Zhdanov AA, Molina IYu, Plyasova LM. Low temperature catalytic decomposition of hydrogen sulfide on metal catalysts under layer of solvent. J Sulf Chem. 2016; 37: 229-240. DOI: 10.1080/17415993.2015.1126593

[6] - Startsev AN. Hydrogen sulfide as a source of hydrogen production. Russ Chem Bull. 2017;66:1378–1397. DOI: 10.1007/s11172-017-1900-y. https://link.springer.com/journal/11172

aqueous MEA, a stream of hydrogen sulfide was introduced in the flow of argon. The criterion of catalyst activity is the absence of hydrogen sulfide in the absorber with zinc acetate and staining the solution in the reactor in yellow. As noted above, the rate of total flow at the outlet of the setup corresponded to the rate of argon supply at the inlet of the plant, while the expected increase in the flow rate due to the additional flow from hydrogen sulfide was not observed (Fig. 12). After stopping the supply of hydrogen sulfide and purging the installation with argon, only traces of hydrogen sulfide were found in the absorber with zinc acetate, which indicates 100 % conversion of hydrogen sulfide. At the same time, the analyzer "Test-1" registered the formation of only 11.5 ml of hydrogen. Therefore, it is concluded that the activated hydrogen formed at the moment of hydrogen sulfide decomposition reacts with argon on the catalyst surface, resulting in the final product of the reaction – hydrogen argonide {H_2Ar}. Based on the material flows balance, half of the argon fed reacted with activated hydrogen.

To test this incredible assumption, the experimental setup was upgraded (Fig. 13). The area of active hydrogen of two absorbers was connected by glass Schliff sections, and rubber tubes were replaced by a steel capillary. In addition, in the reaction a fundamentally new type of catalysts with the conventional name "Start-1" is used which in all its technological parameters is significantly superior to stainless steel chips. During the catalytic experiment, it was possible to clearly observe the release of gas bubbles (Fig. 13). Hydrogen sulfide conversion at this catalyst was still close to 100 %, and, despite the measures taken, the hydrogen concentration at the outlet of the installation did not exceed 0.1 % (Fig. 13). Unfortunately, under the influence of external forces, work in "SCTB Catalyst" for low-temperature catalytic decomposition of hydrogen sulfide is stopped.

Conclusion. The obtained results convince us of the need to accept the incredible hypothesis about the possibility of interaction of active hydrogen, formed at the time of decomposition of hydrogen sulfide on the surface of solid catalysts, with chemically very inert gases – nitrogen and argon.

6. The hypothesis of the interaction of active hydrogen with the carrier gas

If the majority of the unexpected experimental facts discovered by us in this study could be explained from the standpoint of the known laws of catalytic processes, then the absence of hydrogen as a mandatory target product of hydrogen sulfide decomposition cannot be logically described within the existing laws of chemistry. It would seem that this phenomenon could be attributed to the dissolution of the activated atomic hydrogen, which is formed at the moment of decomposition of hydrogen sulfide, in rubber tubes connecting glass absorbers (see, for example, Fig. 6,9). However, this assumption seems unlikely, since the lifetime of atomic hydrogen in condensed media (as opposed to the gas phase) should be very small, which should be accompanied by its recombination into molecular form. As shown in the calibration experiments (Fig. 7.8), losses of molecular hydrogen during its transmission through the catalytic system were not observed. Taking into account the experimental evidence obtained previously at the Institute of Catalysis, an assumption was made about the possibility of interaction of activated hydrogen which is formed at the moment of its origin (in statu nascendi) with the carrier gas – nitrogen or argon, and this process can be realized only on the catalyst surface.

This assumption is based on the concept of the role of hydrogen in the processes of ***chemosynthesis*** of organic matter from hydrogen sulfide and CO_2, with the participation of sulfur bacteria, discussed in detail in 3. The process of chemosynthesis (in contrast to the then known photosynthesis involving sunlight) of organic matter from inorganic precursors – hydrogen sulfide H_2S and carbon dioxide CO_2, was discovered by S.N. Vinogradsky in 1887. He suggested used the term "sulfur bacteria" for the microorganisms that assimilate hydrogen sulfide as an energy source and hydrogen to the activation and implementation of quite inert in chemical terms of molecules of carbon dioxide. The proposed by us mechanism of chemosynthesis of organic matter from CO_2 and H_2S includes two stages. In the first stage the dissociation of hydrogen sulfide by colorless sulfur bacteria is carried out at a temperature and pressure of the environment and results in the formation of active hydrogen and diatomic sulfur in the ground triplet state:

$$2\,H_2S \xrightarrow{\text{sulfur bacteria}} 2\,H_2^* + \{\,^3S_2\,\}^{\text{(globules)}} \qquad (2)$$

as a result of nucleation of diatomic sulfur in the form of condensed hydrophilic phase S_2. The originated active hydrogen is involved in chemosynthesis reactions:

$$H_2^* + CO_2 \xrightarrow{\text{enzymes of sulfur bacteria}} \{\,HCOOH\,\}\ \text{(carbohydrates)} \qquad (3)$$

Thus, toxic and "useless" hydrogen sulfide, which has not found any practical application in human activty, is, apparently, the very substance that underlies the nature-created process of chemosynthesis of organic matter from CO_2, laid the Foundation of biological life on Earth. In contrast to the chemical analogue – water, which is the fundamental basis of the existence of biological organisms, H_2S is a supplier of hydrogen and energy for the processes of life support of these organisms. This extremely important role of hydrogen sulfide is apparently due to the unique property of this molecule – the standard enthalpy of formation ($\Delta_f H^o_{298} = -4.82$ kcal/mol) is the smallest among all known potential sources of hydrogen, which means minimal energy consumption when splitting the molecule. The driving force of hydrogen sulfide decomposition processes in nature is the formation of final products in the ground electronic state (i.e., having minimal free energy) – singlet hydrogen, solid sulfur and diatomic sulfur molecule in the ground triplet state.

There is no doubt that at present, in connection with the problem of global warming, the reactions involving carbon dioxide CO_2 as the initial molecule for the synthesis of organic compounds are of greatest interest. CO_2 chemistry has been the focus of attention of many researchers over the past half-century, as reflected in numerous publications (see e.g.,[7]). Direct gas-phase hydrogenation of CO_2 by molecular hydrogen is a thermodynamically unfavorable process:

$$CO_2 + H_2 \longleftrightarrow HCOOH \qquad (4)$$

since ΔG^o_{298} = +33 kJ/mol (+7.8 kcal/mol) [8]. However, in the aqueous medium, the thermodynamics of this reaction is more favorable to its implementation

$$H_2(aq) + CO_2(aq) \longleftrightarrow HCOOH(aq) \qquad (5)$$

because ΔG^o_{298} = - 4 kJ/mol (- 0.95 kcal/mol). In this regard, at the Institute of Catalysis, we conducted exploratory studies of the possibility of using hydrogen obtained *in situ* in the H_2S decomposition reaction to hydrogenate CO_2 at room temperature. It turned out

[7] - A.M. Appel, J.E. Bercaw, A.B. Bocarsly et al. *Frontiers, Opportunities, and Challenges in Biochemical and Chemical Catalysis of CO_2 Fixation.* // Chem. Rev., 2013, Vol. 113, pp. 6621-6658. dx.doi.org/10.1021/cr300463y.
[8] - [W. Leitner, E. Dinjus, F. Gassner in: *“CO_2 Chemistry—Aqueous- Phase Organometallic Catalysis”* (Eds.: B. Cornils, W. A. Hermann), Wiley-VCH,Weinheim, 1998, p. 488].

that when using stainless steel chips as the simplest and most affordable metal catalyst, carbon dioxide completely inhibits the decomposition reaction of hydrogen sulfide. However, we have found fundamentally new metal catalysts that were insensitive to the atmosphere of carbon dioxide while maintaining their efficiency in the decomposition of hydrogen sulfide. This gives some optimism to the creation of low-temperature catalytic systems for the processing of "acid" hydrogen sulfide containing gases to produce hydrogen, which can be used *in situ* for the assimilation of carbon dioxide and the synthesis of organic molecules.

Thus, for catalytic processes occurring at ambient temperature and atmospheric pressure in the absence of external energy sources, the reactions occur due to the kinetic and potential energy of substrate molecules[9]. In this case, a solid catalyst or enzyme captures the energy of the substrate (exothermic processes of chemisorption and dissociation) and introduces it into a catalytic or biological system, where the energy is converted into other types of internal energy of the system – the formation of new chemical bonds and the removal of reaction products from the system. In the coupled processes of chemosynthesis of carbohydrates of (CO_2 + H_2S) free energy systems generated through the exothermic reactions of the dissociation of hydrogen sulfide and is used to activate CO_2 molecules.

The processes of absorption or formation of hydrogen in biological systems occur with the participation of a specific class of enzymes whose substrate (or product) is molecular hydrogen. These enzymes have a trivial name "hydrogenase".

As is known, the atoms in the nitrogen molecule are bound by a strong triple bond, which is why it does not enter into oxidation-reduction reactions under normal conditions without the use of catalysts – enzymes of diazotrophic bacteria, the most important of which are nitrogenase, hydrogenase, ferredoxin, etc.. Nitrogen fixation is a strictly anaerobic process. Many bacteria, including sulfate-reducing diazotrophs, have now been isolated. The main part of molecular nitrogen (about $1,4 \cdot 10^8$ t/year) is fixed by biotic way.

Currently, more than half of the hydrogen produced in the industry is used for the synthesis of ammonia. Ammonia synthesis is carried out at a temperature of 400-600 °C and pressures of 10-1000 atmospheres. According to the set of properties (catalytic activity, resistance to poisoning, cheapness), the catalyst based on metal iron with impurities of aluminum and potassium oxides was most used.

At the same time, molecular nitrogen can be activated by transition metal compounds and then converted at normal temperature and pressure to NH_3, hydrazine or aromatic amines. The most active compounds in these reactions are Ti, V, Cr, Mo, Fe. The process is carried out in aproton media (esters or hydrocarbons), while some systems are so active that they absorb nitrogen at high rate even at temperatures of —80 °C.

Chemical inertness of mono-atomic argon molecules are explained primarily by the saturation limit of the electron shells. However, talk about argon chemistry today is not pointless, because there are many examples of chemical compounds with inert gases. However, most of these compounds are obtained under very exotic conditions necessary for the activation of an inert gas, and most of these compounds are stable

[9] - А.Н. Старцев. *Определяющая роль катализаторов в процессе низкотемпературного разложения сероводорода: неравновесная термодинамика открытой системы.* Обзор отклонен для публикации в редакциях журналов «Кинетика и катализ» и «Журнал физической химии» и будет опубликован на сайте http://startsev-an.ru

only at low temperatures. For argon we know of only one chemical compound HArF – hydride-fluoride argon, a molecule which is electrically neutral. In addition, the molecular complex of argon {Ar – H_2} [10], an extremely unstable compound existing only at low temperatures, was isolated and characterized.

Taking into account that chemically quite inert CO_2 molecule interacts with the activated *in statu nascendi* hydrogen formed during the decomposition of the hydrogen sulfide by sulfur bacteria at the temperature and pressure of the environment[3] come to the conclusion that incredible hypothesis about the interaction of the activated hydrogen with nitrogen and argon has a right to exist. Since the reaction takes place in very mild conditions with a high yield of the target product, then the creation of appropriate financial conditions and in the presence of accompanying circumstances, this hypothesis can acquire direct experimental evidence.

Conclusion. A hypothesis about the possibility of fixation of molecular nitrogen and chemical activation of argon by hydrogen, which is originated *in statu nascendi* during decomposition of hydrogen sulfide at room temperature on solid catalysts, is proposed. High yields of target products allow us to optimistically consider obtaining positive results of experimental studies to prove the proposed hypothesis. The proposed chemical compound of argon with hydrogen {H_2Ar} is hydrogen argonide by analogy with hydrogen sulfide.

7. Conclusions

The long-suffering scientific study of the process of low-temperature catalytic decomposition of hydrogen sulfide received another portion of incredible experimental results and once again lost financial support for the study of these fundamental laws. Once again, the human factor, called "obscurantism"since the middle ages, is at the forefront of countering scientific progress. However, in this case, the situation is aggravated by the lack of real prospects for the revival of scientific research in the area of reach of my daily movement opportunities, taking into account that the leadership of science in our region still have unsinkable apologists of scientific archaism.

In this regard, I appeal to the progressive layers of the scientific community, to the extent of their ability to pay special attention to obtaining evidence (or, conversely, counterarguments) of the proposed hypothesis. Apparently, "ovchinka worth the candle", because at stake is the possibility of fixing molecular nitrogen at ambient temperature and pressure, and the existence of a new chemical – ***hydrogen argonide {H_2Ar}*** will solve the fundamental problem of activation of inert gases. In addition, the utilization of carbon dioxide in mild conditions will offer an alternative to combat global warming. Exploratory studies of the latter problem give some optimism to the creation of carbon dioxide assimilation processes, since we have found a catalytic system capable of activating a CO_2 molecule at room temperature. I am ready to share my knowledge and experience to solve the tasks.

[10] - A.R.W. McKellar. The infrared spectrum of H_2 – Ar. In: Structure and Dynamics of Weakly Bound Molecular complexes. A. Weber (Ed.). 1987. D. Reidel Publ. Co., P. 141-147.

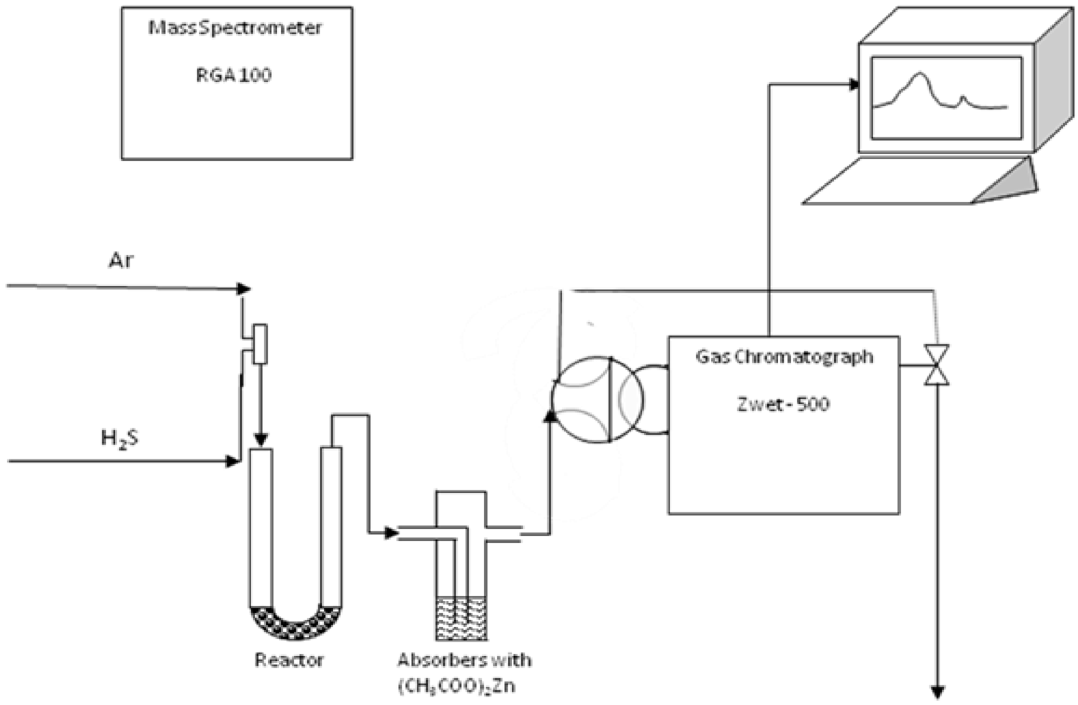


Fig. 1. Scheme of the experimental setup for low-temperature catalytic decomposition of hydrogen sulfide.

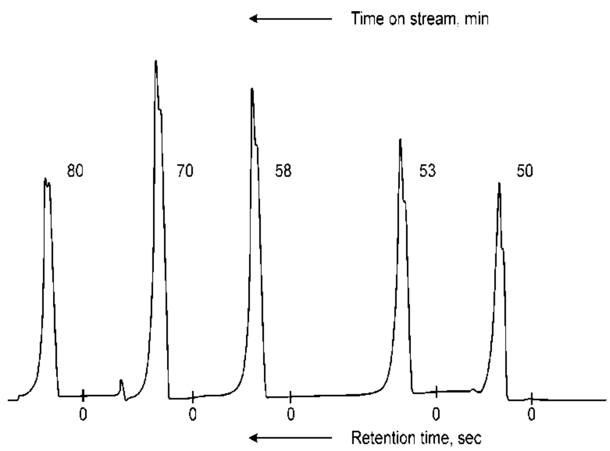


Fig. 2. Chromatogram of the products of the H_2S decomposition on a 0.5 Pt /SiO_2 catalyst. The chromatographic column (3 m x 2 mm) was filled with a Hayser A porous polymer modified with 10% phosphoric acid, column temperature 120 ° C. Retention time S_2 = 98 s, H_2 = 108 s; (for comparison, the retention time of nitrogen / oxygen 140-150 s, hydrogen sulfide 5 min)

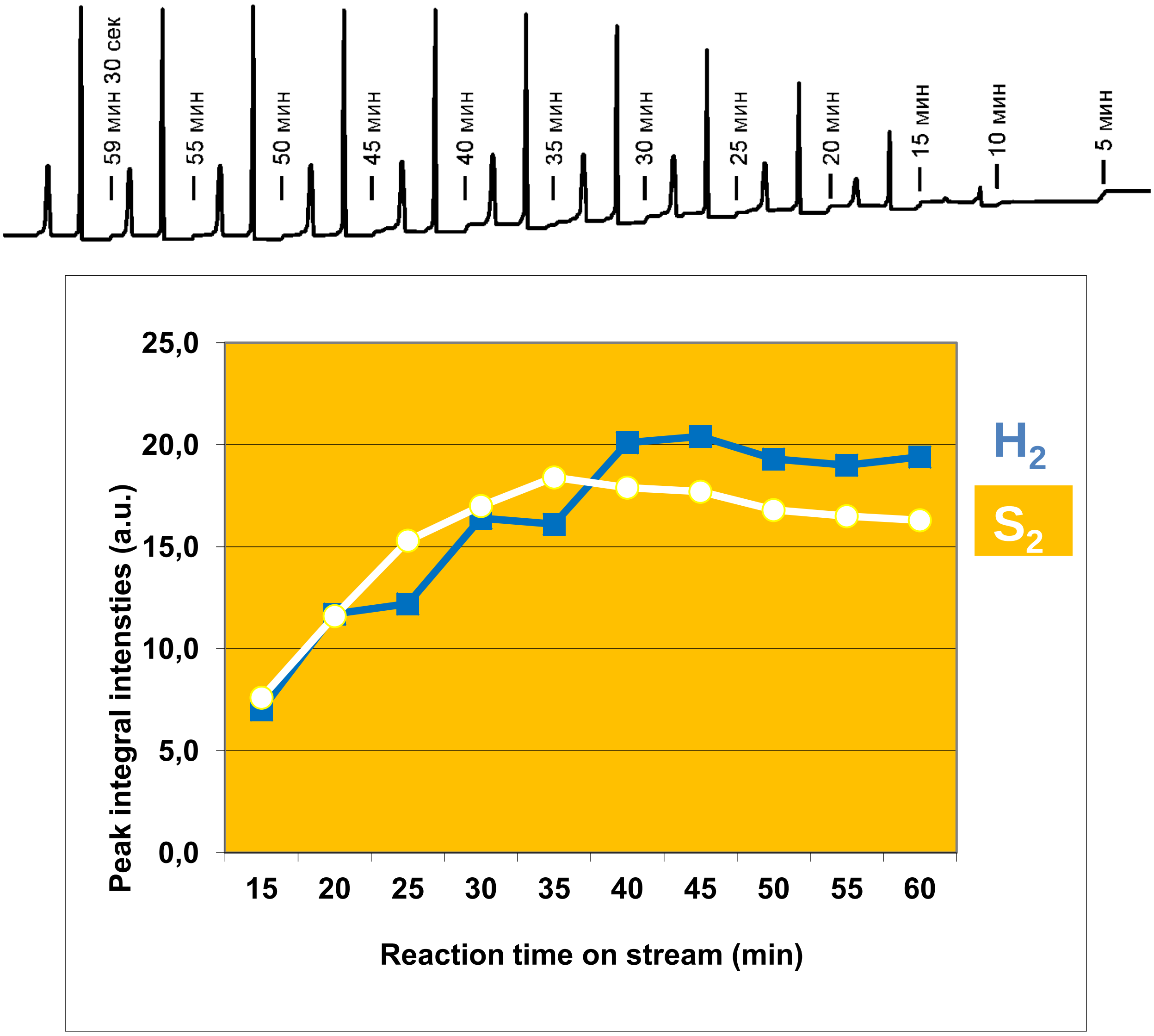


Fig. 3. Separation of the products of H_2S decomposition on a chromatographic column (5 m x 3 mm) filled with AG-3 microporous carbon sorbent, the temperature of the column is room temperature. The retention time of hydrogen is 1 min 51 s, sulfur - 3 min 41 s.

Catalyst - bulk Stainless Steel Chip

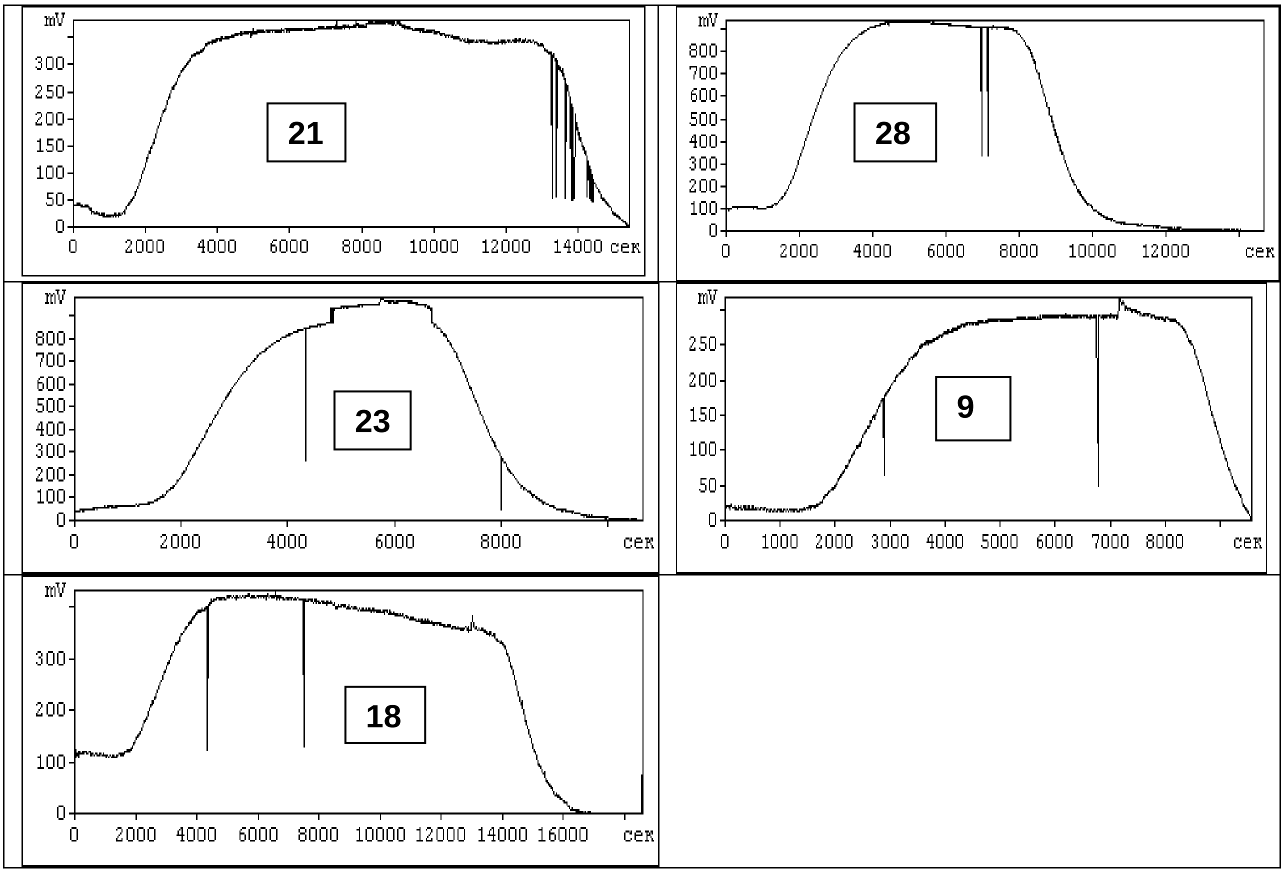


Fig. 4. Hydrogen evolution during the H_2S decomposition of on a 0.5% Pt / SiO_2 catalyst at room temperature. "Zircon" hydrogen analyzer. The frame indicates the turn over number (TON) of the reaction, mol H_2S / mol Pt. The TON for 5 consecutive experiments was 99.

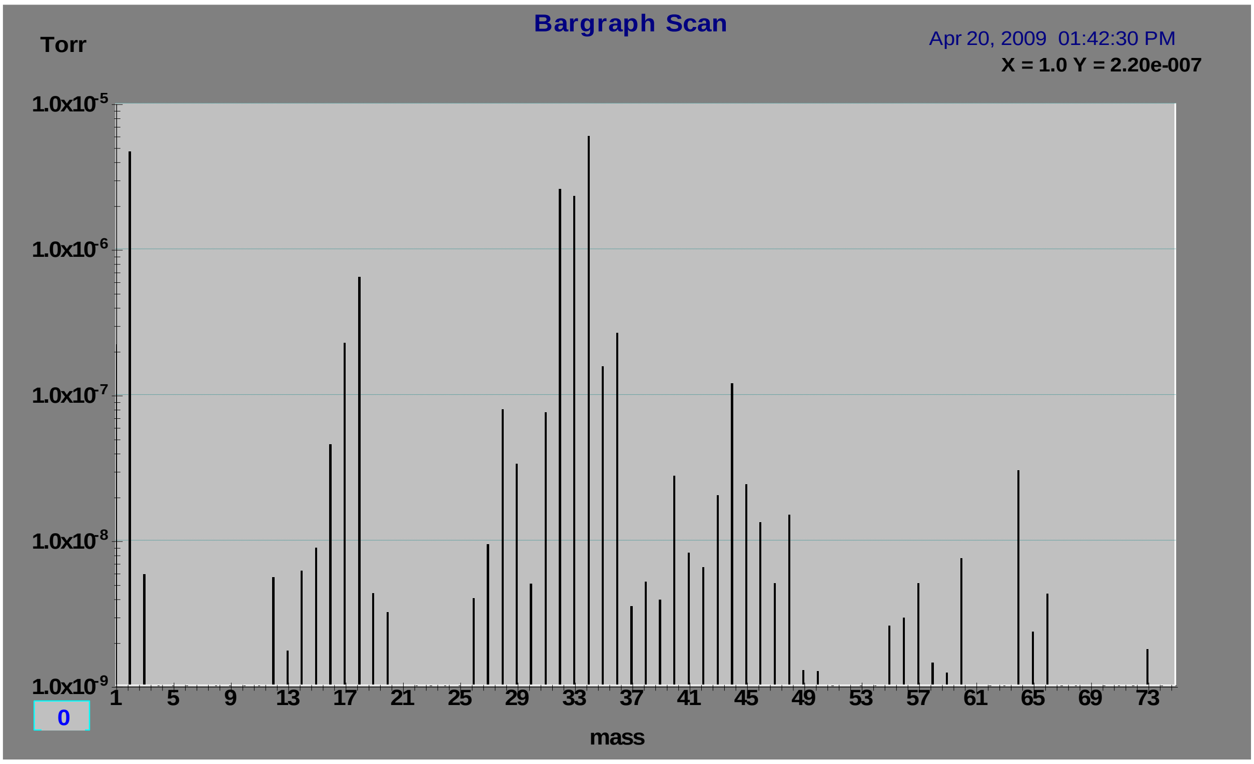


Fig. 5. Mass spectrum of H_2S decomposition products on Pt catalysts at room temperature. Along with hydrogen, gaseous sulfur is formed m / e = 64 amu (atomic mass units) with isotopes 65 and 66 amu, the signal intensity of which is comparable to the signal from argon 40 amu.

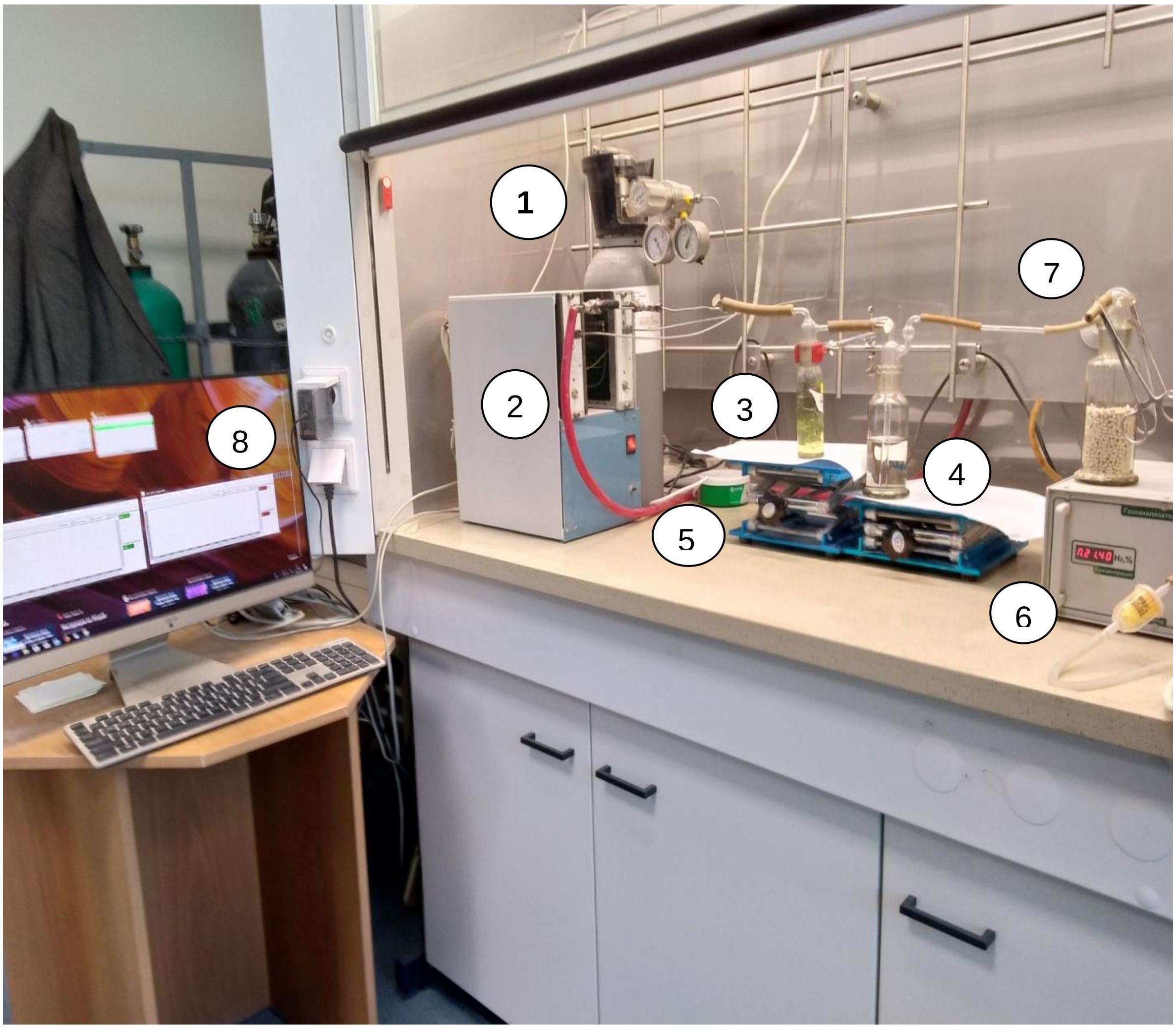
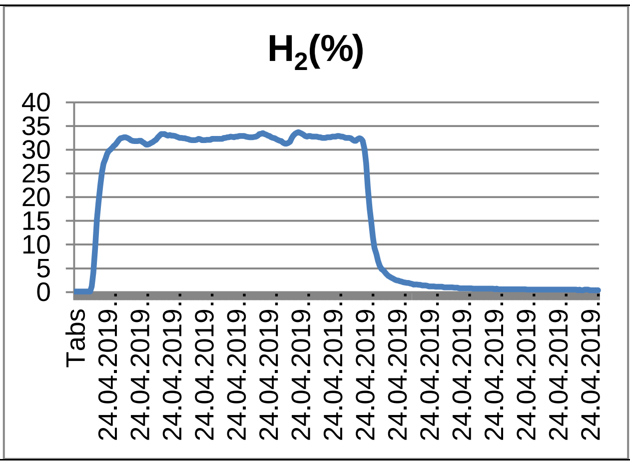
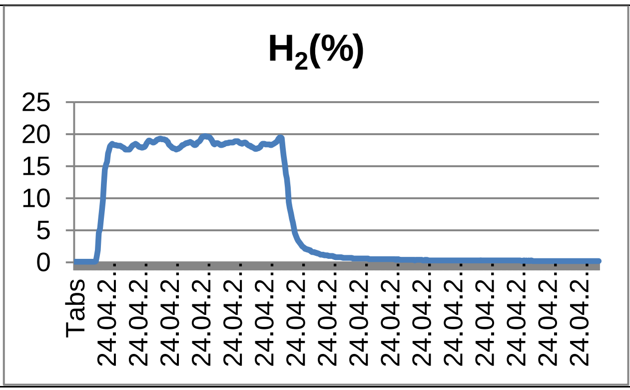


Fig. 6. An experimental setup, created in “SKTB Catalyst”, for low-temperature decomposition of hydrogen sulfide on stainless steel chips under a solvent layer.

1. Cylinder with hydrogen sulfide.
2. The controller with gas flow regulators (GFG) of argon (nitrogen) and H_2S.
3. Catalytic reactor.
4. Absorber with an aqueous solution of zinc acetate.
5. Magnetic stirrer.
6. “Test-1” hydrogen analyzer.
7. A trap to absorb moisture and gaseous sulfur.
8. Computer

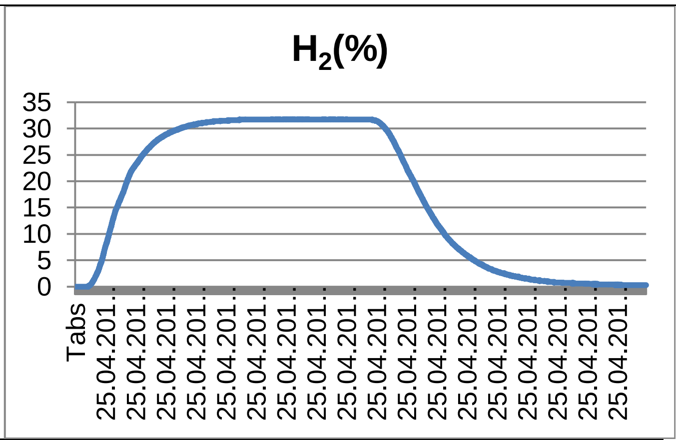


Flows: Argon 20.2 ml/min + H_2 10.1 ml/min,
Σ Flow output 30 ml / min
The time for the supply of hydrogen is 5 minutes
H_2 supplied 50.5 ml
H_2 Received (integral) 50 ml
Theoretical concentration of H2 in the gas phase **33.3%**



Flows: Argon 20.0 ml / min + H_2 5.0 ml / min,
ΣFlow at the exit of 25 ml / min.
The time for the supply of hydrogen is 5 minutes
H_2 supplied 25 ml
Received (integral) 24.3 ml of H_2
Theoretical concentration of H_2 in the gas phase **20%**

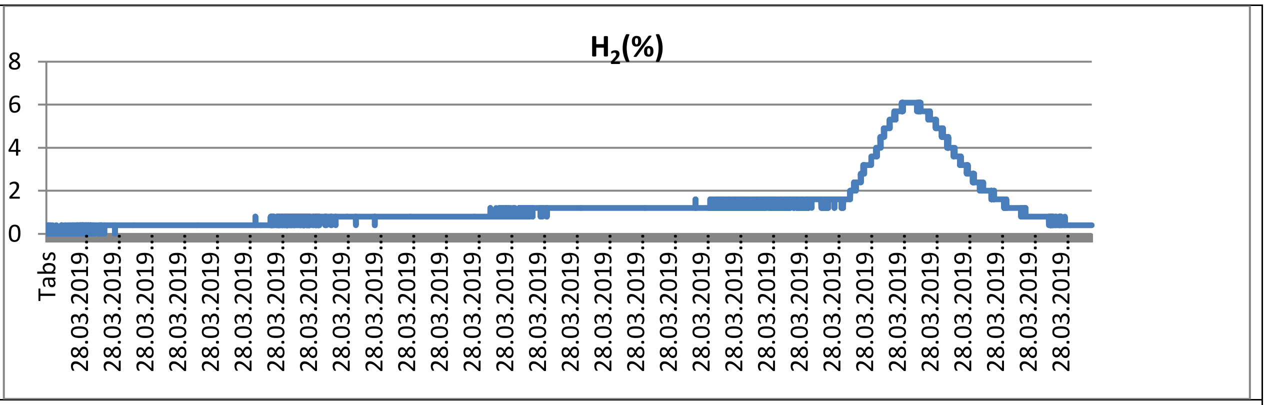
Fig. 7. Calibration of the hydrogen analyzer “Test-1”.



Catalyst stainless steel chip in 10% MEA,
Flows: Argon 20.6 ml/min + H_2 10.3 ml/min,
Σ Flow output 30 ml / min
The time of hydrogen supply is 90 min
H_2 supplied 927 ml
H_2 received (integral) 939 ml
Theoretical concentration of H_2 in the gas phase **33.3%**

The catalyst and MEA are the same, argon 11.8 ml / min, H_2 5.0 ml / min, time 110 min,
550 ml H_2 supplied, obtained (integral) **535 ml H_2**

Fig. 8. Calibration of the hydrogen analyzer "Test-1" in an experimental setup



Reaction conditions:

gas phase, supply of nitrogen 0 ml / min, hydrogen sulfide 2.55 ml / min.

After 3 hours and 50 minutes, the supply of hydrogen sulfide was stopped,

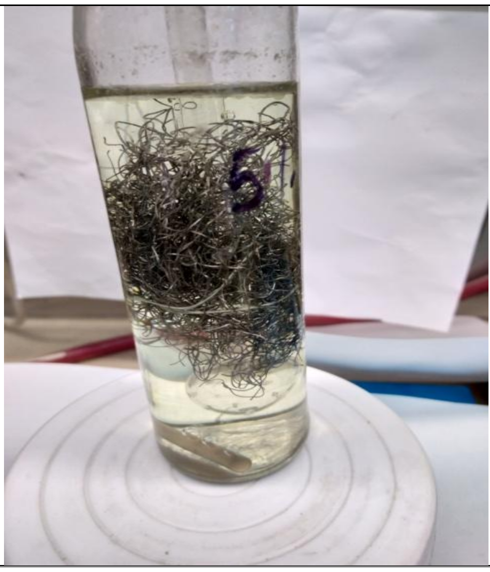
a purge of 50 ml / min of nitrogen.

587 ml of hydrogen sulfide was fed, **16.6 ml** in an absorber.

The conversion of hydrogen sulfide is **97.1%.**

Received **hydrogen v = 100 ml**

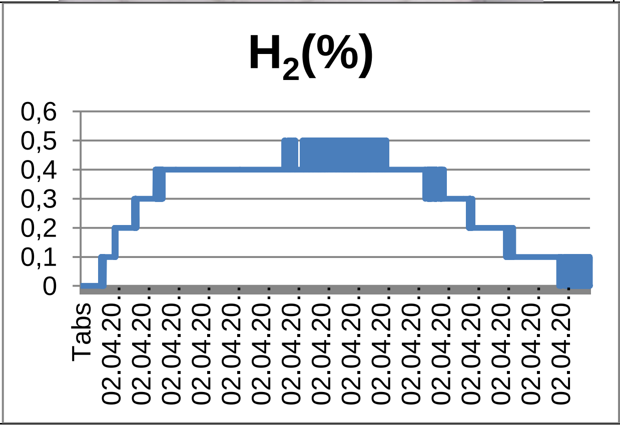
Fig. 9. The gas-phase process of H_2S decomposition. The color of alumina changed to yellow due to sulfur adsorption.



Reaction conditions:
5% MEA
feed (nitrogen 15 ml / min + 10 ml / min of hydrogen sulfide),

2200 ml of hydrogen sulfide was fed in 2 days, **14.5 ml** in an absorber,
H_2S conversion is **99.7%.**

The volume of hydrogen **(31 + 14) = 45 ml**.



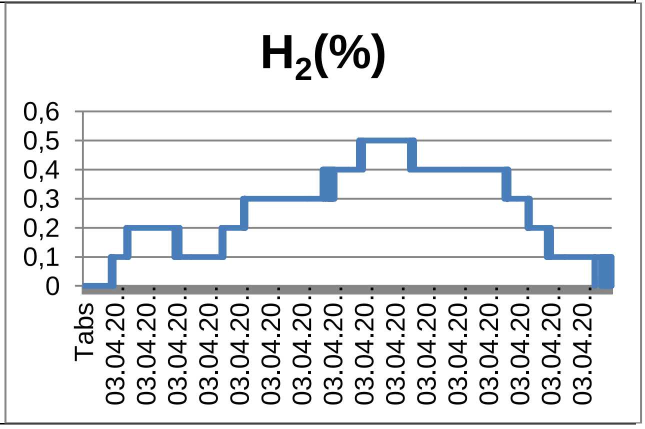


Fig. 10. The H_2S decomposition in a stream of nitrogen at room temperature on stainless steel chips under a layer of an aqueous MEA

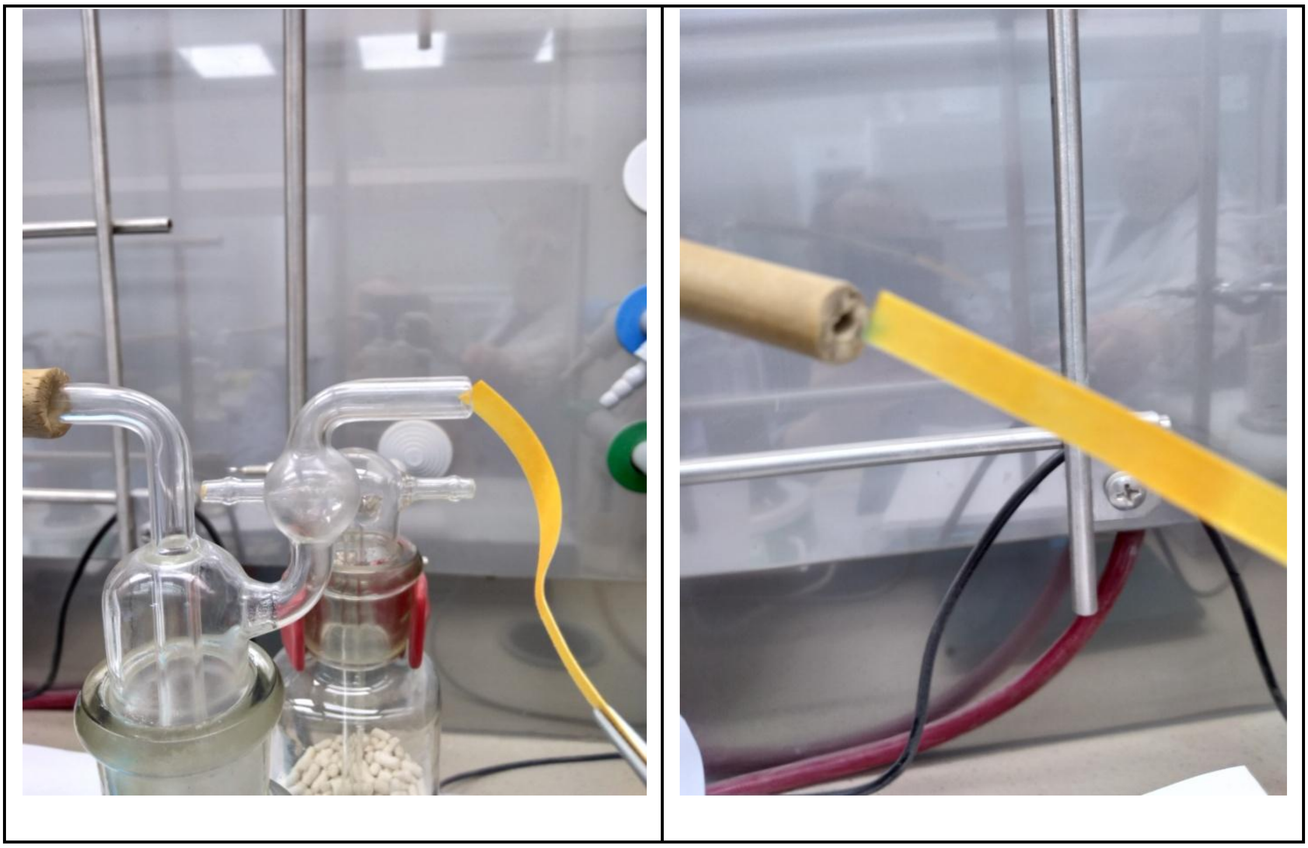


Fig. 11. The appearance of an alkaline gas component at the outlet of the catalytic reactor of hydrogen sulfide decomposition in a stream of nitrogen.

Catalyst: stainless steel chips placed in 10% MEA 150 ml.

Gas flows: **Argon 20.0 ml / min** + **H_2S - 10.6 ml / min**
(total flow at the inlet to the unit **Σ = 30.6 ml / min**)

H_2S feed time 120 min,

Supplied - **1268 ml H_2S**, **2400 ml Ar**

At the outlet of the installation, a flow rate is **20.2 ml / min**

1268 ml of H_2S reacted (conversion of about 100%)

Hydrogen produced **11.5 ml**

1256 ml of Ar reacted
Product Selectivity = 52.3%

Theoretical concentration of H_2 in the gas phase 34.6%

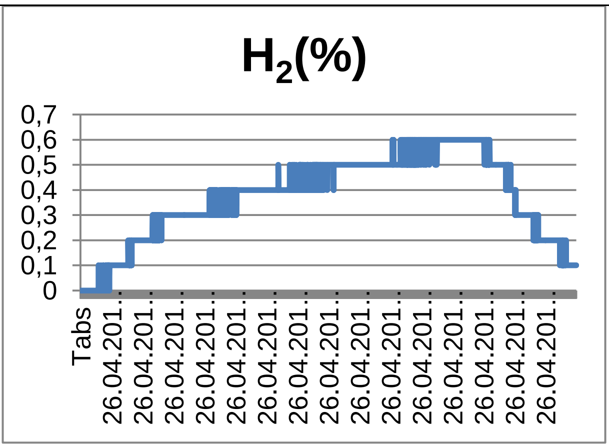


Fig. 12. The decomposition of hydrogen sulfide in a stream of argon

Fig. 13. The decomposition of hydrogen sulfide on the “Start-1” catalyst under a 5% aqueous MEA solution. The carrier gas is argon.

To the right in the frame are clearly visible gas bubbles released during the supply of hydrogen sulfide through the catalyst.