

## Optimization of Adsorption Processes with Cyclic Variable Pressure in Gas Mixture Separation

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### Abstract

The paper considers the problems of optimization of the regime variables (pressure at the adsorption stage and the time of the adsorption cycle), and automation of the adsorption separation process of the gas mixture and production of hydrogen in a 4-adsorption plant for pressure swing adsorption. Since the optimization problem belongs to the class of nonlinear programming problems, penalty methods and methods of sequential quadratic programming have been used for its solution. A two-level SCADA system for adaptive optimization and control has been developed to solve the optimization problem and automatically control the hydrogen production process. The results of simulation studies of the effectiveness of the SCADA control system with a stepwise change in the disturbances (concentration of carbon dioxide, temperature and pressure of the gas mixture) in the feed of the pressure swing adsorption plant are presented.

### Keywords

Pressure swing adsorption; gas mixture separation; hydrogen; carbon dioxide; process dynamics; mathematical model; optimization; disturbances; control.

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### Introduction

The formulation of optimization and automation problems begins with the analysis of its functioning as a control object that involves the study of the static and dynamic properties of the process, identification of input, main disturbing and control actions, output (controlled) variables, and finding acceptable ranges for their change. The analysis of the adsorption

separation of gas mixture and hydrogen production in a 4-adsorption pressure swing adsorption (PSA) plant as a control object conducted in [1, 2] made it possible to determine (Fig. 1):

– input variables  $\mathbf{x}$  (disturbing actions) of PSA plant (composition  $y^{in} = (y_1^{in}, y_2^{in}, y_3^{in})$ , temperature  $T_g^{in}$ , consumption  $G^{in}$  and pressure  $P^{in}$  of gas

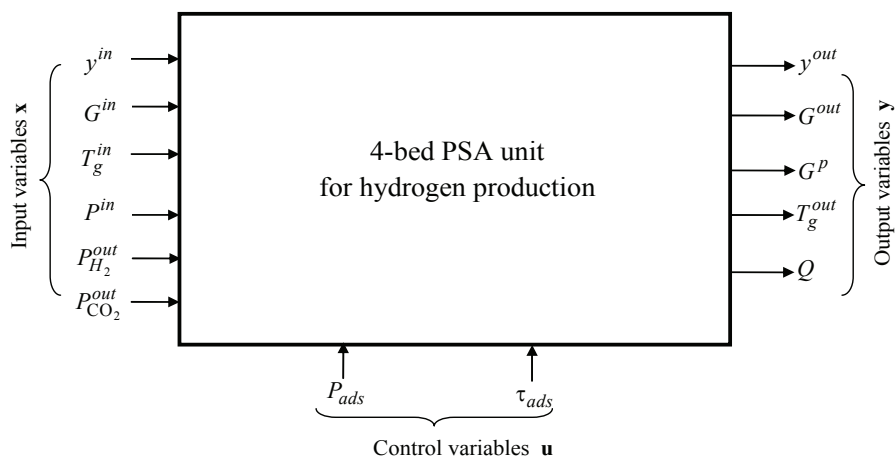


Fig. 1. Structural diagram of the PSA plant as a control object in hydrogen production

mixture in the plant feed, pressure on the production  $P_{H_2}^{out}$  and discharge  $P_{CO_2}^{out}$  outlets of the PSA plant  $P_{H_2}^{out}$ ), i.e.  $x = \{y^{in}, T_g^{in}, G^{in}, P^{in}, P_{H_2}^{out}, P_{CO_2}^{out}\}$ ;

– control variables  $u$  pressure  $P_{ads}$  and cyclic time  $\tau_{ads}$  of adsorption stage), i.e.  $u = \{P_{ads}, \tau_{ads}\}$ ;

– output variables  $y$  of the PSA plant (compositions  $y^{out} = (y_1^{out}, y_2^{out}, y_3^{out})$ , consumptions  $G^{out}$ ,  $G^p$  and temperature  $T_g^{out}$  of the gas mixture at the PSA plant outlet, plant productivity  $Q = y_1^{out} G^p$ , where  $G^p = G^{out} - G^{des}$ ,  $G^{des}$  is volumetric flow rate of hydrogen-rich gas directed to the regeneration of adsorbent, i.e.  $y = \{y^{out}, G^{out}, G^p, T_g^{out}, Q\}$ .

### Mathematical modules for the hydrogen production process in the 4-adsorption PSA plant

A mathematical description of the dynamics of the adsorption process in the PSA plant adsorbers for hydrogen production [1] can be given in the form of a block diagram of the “Adsorber” mathematical module (Fig. 2). The mathematical model differs from the known ones, as it takes into account the impact of mass and heat transfer processes and the velocity of a multicomponent gas mixture flow on the kinetics of mixed-diffusive transport of adsorbent ( $H_2$ ,  $CO_2$ ,  $CO$ ), and allows calculating the profiles of component concentrations and temperatures in the gas and solid phases, pressure and velocity of the gas mixture by the adsorbent height as a function of time. The tuning parameters of the mathematical model are the mass transfer coefficients  $\beta_k^1$  (1 –  $H_2$ , 2 –  $CO_2$ , 3 –  $CO$ ) and formal kinetic parameters  $\beta_k^2$  and  $\theta$  of the equation of mass transfer of adsorbent ( $H_2$ ,  $CO_2$ ,  $CO$ ) from the gas phase to the solid phase of the adsorbent (through the phase interface):

$$\frac{\partial a_k}{\partial t} = \frac{F_k^2 - F_k^1}{2} \tanh(\theta(v_g - v_g^*)) + 1 + F_k^1,$$

$$k = H_2, CO_2, CO,$$

where  $F_k^1$  is the right-hand side of the kinetics equation of non-stationary convective (external) mass transfer,  $F_k^1 = \beta_k^1(c_k - c_k^*)$ ;  $\beta_k^1$  is the mass transfer factor referred to the concentration of adsorbent in the gas phase, 1/s;  $c_k$  is molar concentration of the  $k$ -th component of the gas mixture, mol/m<sup>3</sup>;  $a_k$  is sorption value of the  $k$ -th component in the adsorbent, mol/m<sup>3</sup>,

$c_k^*$  is the adsorption concentration at the phase interface or the equilibrium current adsorption value  $a_k$ , mol/m<sup>3</sup>,  $F_k^2$  is the right-hand side of the kinetics equation inside the diffusion adsorption process,  $F_k^2 = \beta_k^2(a_k^* - a_k)$ ;  $\beta_k^2$  is the kinetic coefficient, 1/s;  $a_k^*$  is the adsorption value, which is the equilibrium current concentration of adsorbent  $c_k$  in flow of the gas mixture on the outer surface of the granules, mol/m<sup>3</sup>;  $v_g$  is velocity of the gas mixture, m/s;  $v_g^*$  is the critical velocity of the gas mixture, which determines the transition from the diffusion region (external mass transfer) to the kinetic region (internal diffusion in adsorbent granules) of adsorption transfer, m/s.

A specific type of equations for determining the tuning parameters of the model is given in [2, 3]. However, the numerical values of the parameters obtained from these equations are recommended to be used as initial approximations for solving the inverse coefficient problem using the equations of the mathematical model [3]:  $v_g^* = 0.022$  m/s;

$$\beta_{H_2}^1 = 0.816 \text{ 1/s}; \quad \beta_{CO_2}^1 = 0.021 \text{ 1/c}; \quad \beta_{CO}^1 = 0.084 \text{ 1/s};$$

$$\beta_{H_2}^2 = 0.73 \text{ 1/c}; \quad \beta_{CO_2}^2 = 0.012 \text{ 1/s}; \quad \beta_{CO}^2 = 0.059 \text{ 1/s},$$

$$\theta = 18.2.$$

The state of the valve is described by the components of the vector  $\sigma$  that are Boolean variables taking the values 0 or 1. The specific form of the function  $\sigma = \sigma(t)$  is determined using the valve switching cycle of the PSA plant [1, 2]. A mathematical description of the operating mode of the valve system of the 4-adsorption PSA plant for hydrogen production can be presented in the form of a block diagram of the mathematical module “Valve system” (Fig. 3).

In Fig. 2, the following notation is adopted:  $\varepsilon, \varepsilon_0$  – are porosity coefficients of the adsorbent with and without the porosity of the granules, respectively, m<sup>3</sup>/m<sup>3</sup>;  $D_g^k$  is the effective coefficient of longitudinal mixing of the  $k$ -th component in the gas mixture, m<sup>2</sup>/s;  $x$  is a spatial coordinate along the axis of the adsorber (height in the adsorbent bed), m;  $t$  is time, s;  $c_v^g, \rho_g$  – are the specific heat and molar density of the gas mixture, respectively, J/(mol·K), mol/m<sup>3</sup>;  $T_g$  is the temperature of the gas mixture, K;  $T_a$  is the adsorbent temperature, K;  $\lambda_g, \lambda_a$  – are the coefficients of thermal conductivity of the gas mixture and adsorbent, respectively, W/(m·K);  $\alpha$  – is the heat transfer

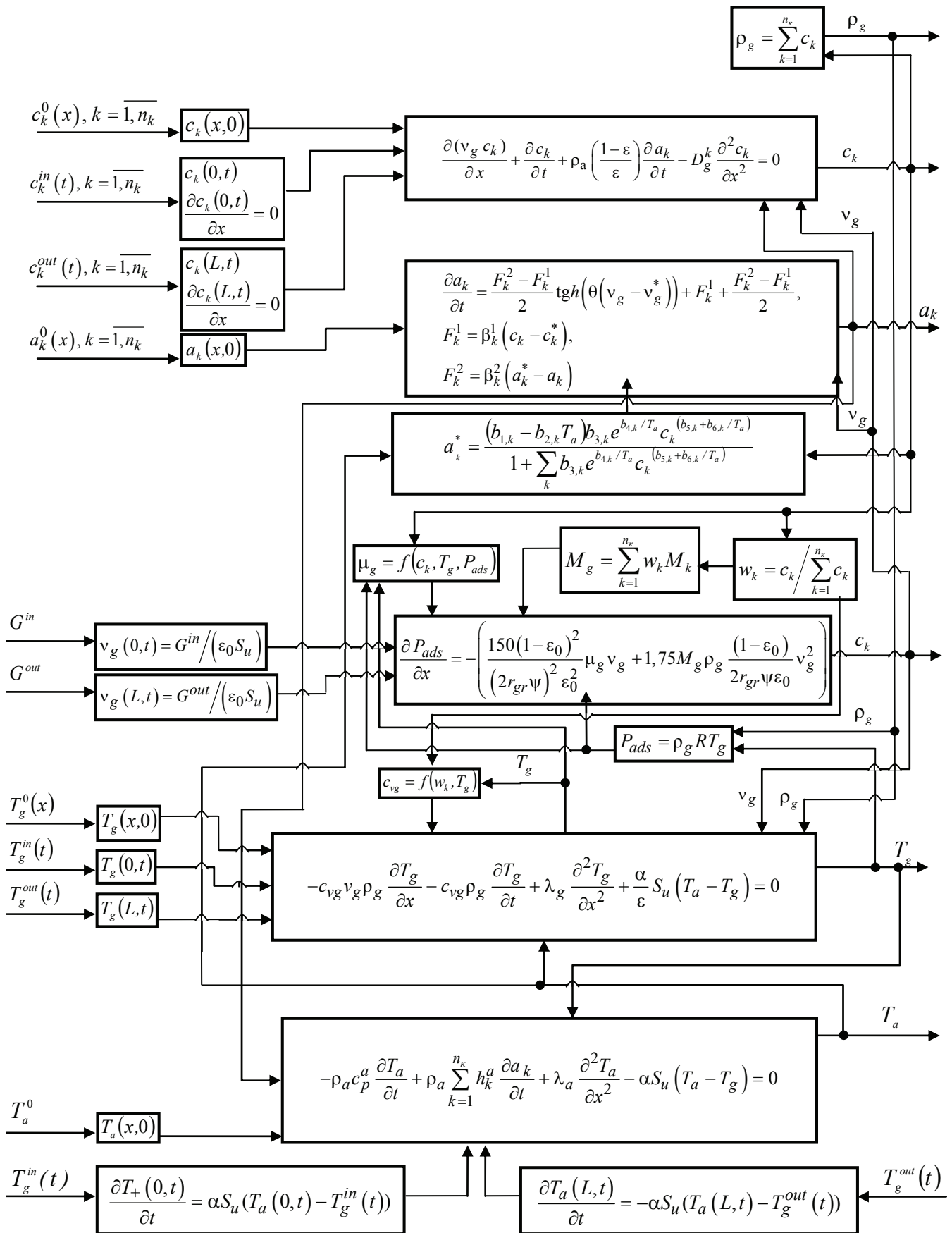
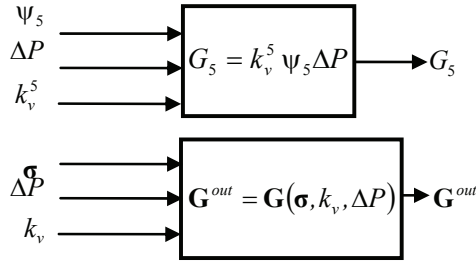


Fig. 2. The block diagram of the mathematical description of the “Adsorber” module



**Fig. 3. Block diagram of the mathematical description of the module “Valve system”:**

$G_5$  – is the flow through the control valve  $K_5$ ,  $\text{m}^3/\text{s}$ ;

$k_v^5$  – is the valve throughput,  $\text{m}^3/(\text{s}\cdot\text{Pa})$ ;  $\psi_5$  – is the valve opening

degree  $K_5$ ;  $\Delta P$  – is the pressure drop across the valve  $K_5$ ,  $\text{Pa}$ ;

$G = G(\sigma, k_v, \Delta P)$  is the system of equations determining the flow of gas passing through the shut-off valves K1.1- K4.1, K1.2-K4.2, K1.3-K4.3, K12, K13, K14, K23, K24, K34 of the PSA plant [1]

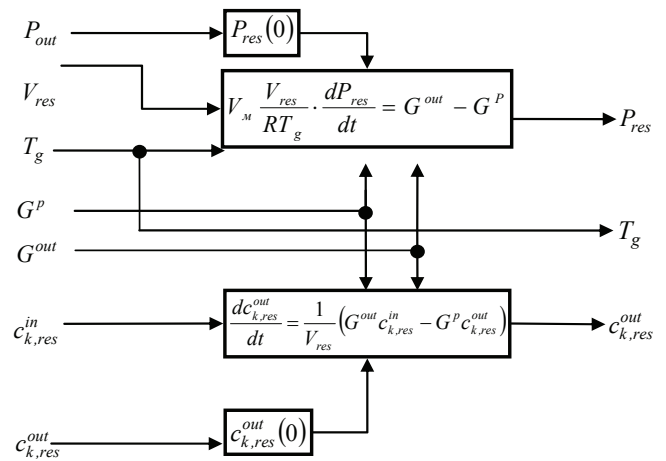
coefficient from the surface of the adsorbent granules to the gas mixture flow,  $\text{W}/(\text{m}^2\cdot\text{K})$ ;  $S_u = (1-\varepsilon)\frac{3}{r_{gr}}$  is

the specific surface coefficient of the adsorbent granules,  $\text{m}^2/\text{m}^3$ ,  $r_{gr}$  is the radius of the adsorbent granule,  $\text{m}$ ;  $c_p^a$  is the specific heat capacity of the adsorbent,  $\text{J}/(\text{kg}\cdot\text{K})$ ;  $\rho_a$  is the density of the adsorbent,  $\text{kg}/\text{m}^3$ ;  $c_p^g$  is the adsorbate specific heat,  $\text{J}/(\text{mol}\cdot\text{K})$ ;  $h_k^a$  is the heat of adsorption of the  $k$ -th component of the gas mixture,  $\text{J}/\text{mol}$ ;  $\psi$  is the sphericity coefficient of the adsorbent granules;  $\mu_g$  is the dynamic viscosity of the gas mixture,  $(\text{N}\cdot\text{s})/\text{m}^2$ ;  $M_g$  is the molar mass of the gas mixture,  $\text{kg}/\text{mol}$ ;  $b$  is the parameter vector of the Langmuir-Freundlich sorption isotherm [3].

Finally, the mathematical description of the receiver includes equations for the dynamics of pressure and concentration of the components of the gas mixture to be separated, and it can be visually represented in the form of a block diagram of the mathematical module “Receiver” (Fig. 4), where  $V_M$  – is the molar volume,  $\text{m}^3/\text{mol}$ ;  $V_{res}$  is the volume of the receiver,  $\text{m}^3$ ;  $P_{res}$  is the pressure of the gas mixture in the receiver,  $\text{Pa}$ ;  $P_{out}$  is the pressure of the production gas mixture,  $\text{Pa}$ ;  $G^P$  is the consumption of production gas mixture,  $\text{m}^3/\text{s}$ ;  $G^{out}$  is the consumption of production gas mixture at the outlet of adsorbers  $A_1$ – $A_4$ ,  $\text{m}^3/\text{s}$ , is determined by a logical expression:

$$G^{out} = G_{1,2}\sigma_{1,2} \vee G_{2,2}\sigma_{2,2} \vee G_{3,2}\sigma_{3,2} \vee G_{4,2}\sigma_{4,2};$$

$c_{k,res}^{in}$ ,  $c_{k,res}^{out}$  is the concentration of gas mixture components at the inlet and outlet of the receiver, respectively.



**Fig. 4. The block diagram of the mathematical description of the module “Receiver”**

Further, the concentrations of hydrogen, dioxide and carbon monoxide will be denoted by  $y = (y_1, y_2, y_3)$ , vol. %.

### Statement of the task of optimizing the process variables of the PSA plant

The task of optimizing the process of hydrogen production using PSA technology can be formulated as follows: for given ranges of disturbing actions  $x = \{y^{in}, T_g^{in}, G^{in}, P^{in}, P_{H_2}^{out}, P_{CO_2}^{out}\}$ , it is required to find a vector of permissible controls  $u = (P_{ads}, \tau_{ads})$ , in which the objective function is the average value of the concentration  $y_1^{out}$  of production hydrogen for a given period  $[0, t_{pr}]$  reaching the extreme value, i.e.

$$I(u^*) = \left( \frac{1}{t_{pr}} \int_0^{t_{pr}} y_1^{out}(u^*) dt \right) = \max_{u=\{P_{ads}, \tau_{ads}\}} \left( \frac{1}{t_{pr}} \int_0^{t_{pr}} y_1^{out}(u) dt \right), \quad (1)$$

to satisfy the connections  $F(x, y'', y', y, u) = 0$  in the form of equations of a mathematical model [1] and constraints on:

$$\begin{aligned} &\text{– concentration of production hydrogen } y_1^{out} \\ &\quad y_1^{out} - y_1^{out} \leq 0; \end{aligned} \quad (2)$$

$$\begin{aligned} &\text{– productivity } Q_{H_2} \text{ of the PSA plant} \\ &\quad \underline{Q} \leq Q \leq \bar{Q}; \end{aligned} \quad (3)$$

$$\begin{aligned} &\text{– the flow rate of the gas mixture } G^{in} \text{ in the feed of the PSA plant} \\ &\quad \underline{G^{in}} \leq G^{in} \leq \bar{G^{in}}; \end{aligned} \quad (4)$$

– adsorption pressure  $P_{ads}$

$$P^{in} \leq P_{ads}, \quad (5)$$

where  $\underline{z}$ ,  $\bar{z}$  are lower and upper limit permissible values of technological variables.

The formulated problem (1)–(5) belongs to the class of problems of nonlinear programming, and for its solution, we will use the penalty methods and sequential quadratic programming [4].

### Automation of the PSA plant for hydrogen production

A 2-level SCADA system for adaptive optimization and control has been developed to solve the optimization problem (1)–(5) and control the hydrogen production process.

The process variables of the hydrogen adsorption plant are subject to random changes during the adsorption process. The disturbance values, which are represented by unregulated variables of the initial gas mixture in the feed of the PSA plant, also vary randomly during the process. In this case, it is necessary to maintain a priori an unknown maximum value of the specified optimization function, which causes the application of the adaptive optimization and control system with variable tasks to automatic regulators operating by the principle of disturbance control with a reference to the process model in the control loop.

In the SCADA system of adaptive optimization and control, the current values of disturbing actions are

continuously controlled, and when the disturbing actions deviate from the nominal values at the upper level, the optimization problem (1)–(5) is promptly solved by the personal computer, and the current optimal tasks  $\hat{u} = (\hat{P}_{ads}, \hat{\tau}_{ads})$  are determined for the automatic control system regulators of the hydrogen production process at the lower level.

Based on the obtained value  $\hat{\tau}_{ads}$ , the cyclogram  $\hat{U}$  of valve operation is recalculated and the programming regulator (PR) is implemented in the 4-adsorption PSA plant for hydrogen production. The current optimum value  $\hat{P}_{ads}$  comes as a reference to the PID controller of a single-loop automatic feedback control system (Fig. 5).

Thus, the SCADA system of adaptive optimization and control of the process modes of the PSA plant provides the following functions:

- search and maintenance of the optimum value of the purity  $y_1^{out}$  of the produced hydrogen;
- calculation of the current optimum time of adsorption step  $\hat{\tau}_{ads}$ ;
- calculation and implementation of the optimum cyclogram of valve operation in 4-adsorption PSA plant for hydrogen production;
- calculation of the current optimum adsorption pressure  $\hat{P}_{ads}$ ;
- calculation and formation of control actions on valve drives [1].

The functional diagram of the automation system of the 4-adsorption PSA plant is shown in Fig. 6.

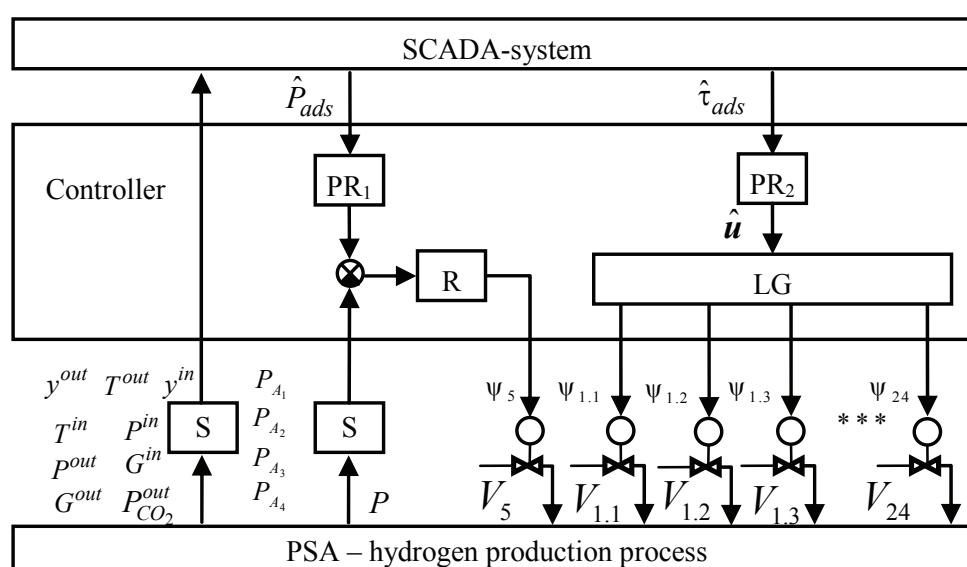


Fig. 5. Block diagram of the control system:

$\Psi$  – the valve opening degree; S – sensor, R – regulator, LG – logic gate, PR – programming regulator



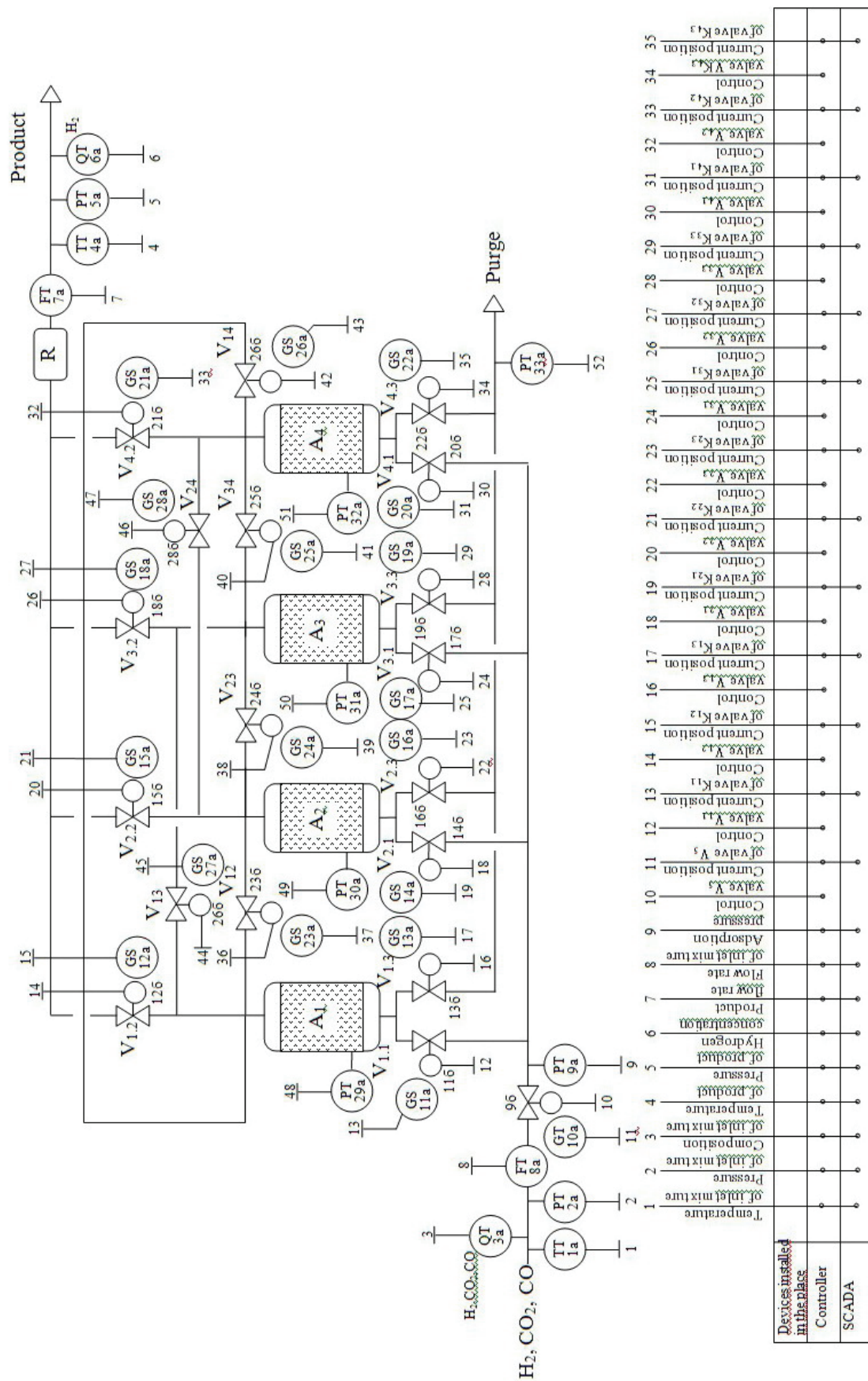


Fig. 6. Functional diagram of automation (start)

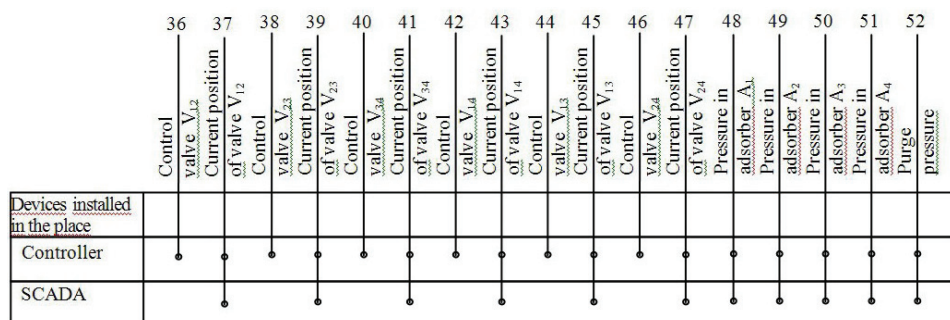


Fig. 6. Functional diagram of automation (end)

### Simulation study of the control system

The results of a comparative analysis of the PSA plant operation with and without adaptive optimization in the SCADA control system are presented in Table 1.

Fig. 7 shows the results of simulation studies of the SCADA control system with a stepwise change in disturbing actions.

The analysis of transient processes in the SCADA system of adaptive optimization and control (Fig. 7 *a*, *b*, *c*) with a stepwise change in the proportion of  $\text{CO}_2$  in the gas mixture flow at the inlet to the PSA plant makes it possible to conclude that in the PSA plant with adaptive optimization and control, a decrease in the concentration of production hydrogen from 99 to 96 vol. % is observed, while in the PSA plant without adaptive optimization it decreases from 94 to 87 vol. % At the same time, the SCADA system maintains the concentration  $y_1^{\text{out}}$  of production

Table 1

### Analysis of PSA plant performance

Distrubing actions – stepwise increase:	$I^*$ , vol. %	$I$ , vol. %	$\Delta I$ , vol. %
$y_2^{\text{in}}$ from 34 to 45 vol. %	95.76	89.46	6.3
$T_g^{\text{in}}$ from 30 to 50 °C	98.92	94.13	4.79
$P_{\text{H}_2}^{\text{out}}$ up to 0.3 MPa	98.92	95.09	3.83
$P_{\text{CO}_2}^{\text{out}}$ up to 0.1 MPa	96.51	90.22	6.29

$I^*$ ,  $I$  are values of the objective function with and without adaptive optimization in the control system, respectively.

hydrogen at the maximum possible level for the changed characteristics of the initial gas mixture in the plant feed and ensures the constraint (3) on the permissible plant capacity.

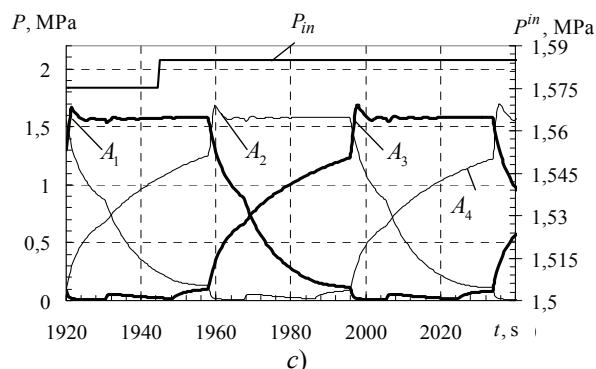
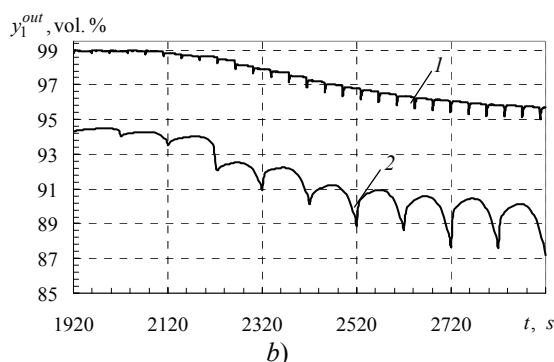
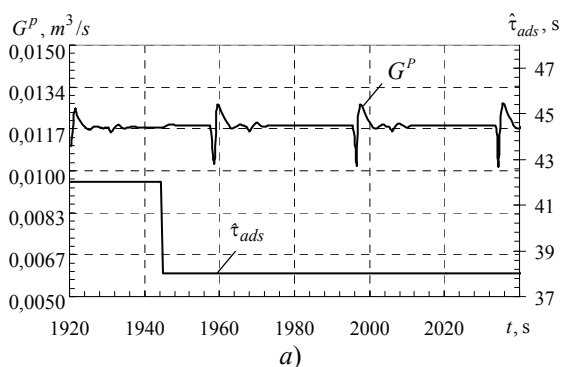
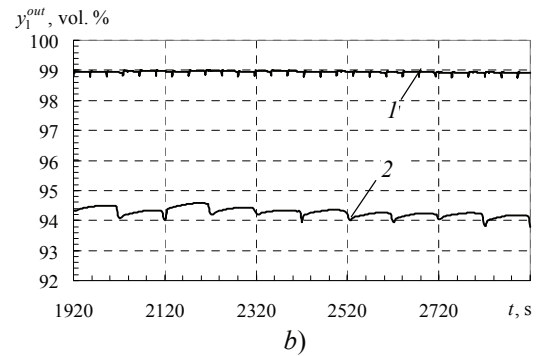
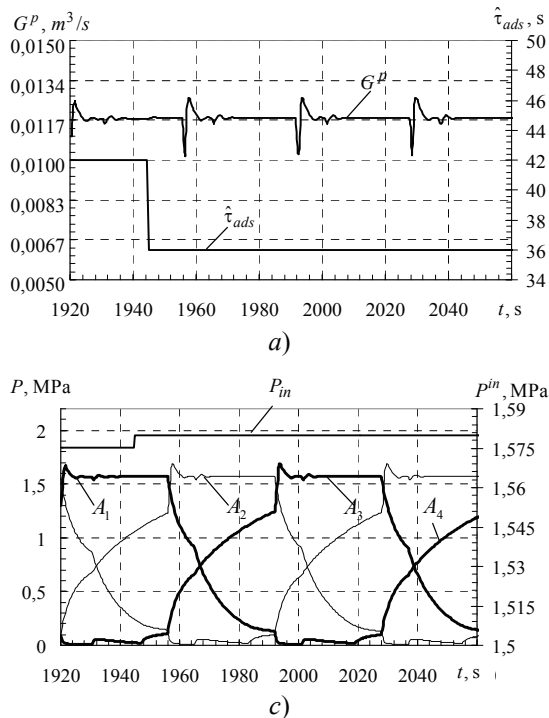


Fig. 7. Transient processes in the SCADA control system with a stepwise increase in the  $\text{CO}_2$  content of the starting mixture from 30 to 45 % vol.:

*a* – the change in the consumption  $G^P$  of production hydrogen and the optimum time of the adsorption cycle  $\tau_{\text{ads}}$  as a function of time; *b* – the change in concentration  $y_1^{\text{out}}$  as a function of time in the presence of adaptive optimization (1) and without adaptive optimization (2); *c* – pressure changes in adsorbers  $A_1$ – $A_4$  of the PSA plant



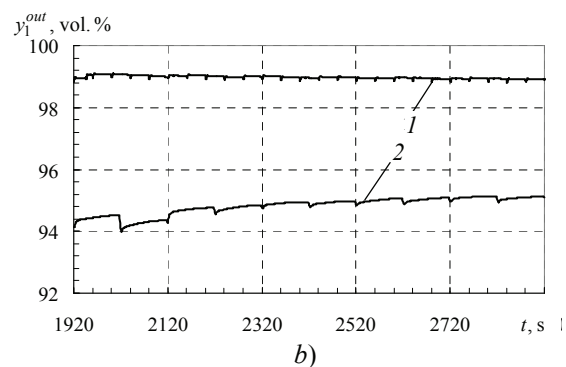
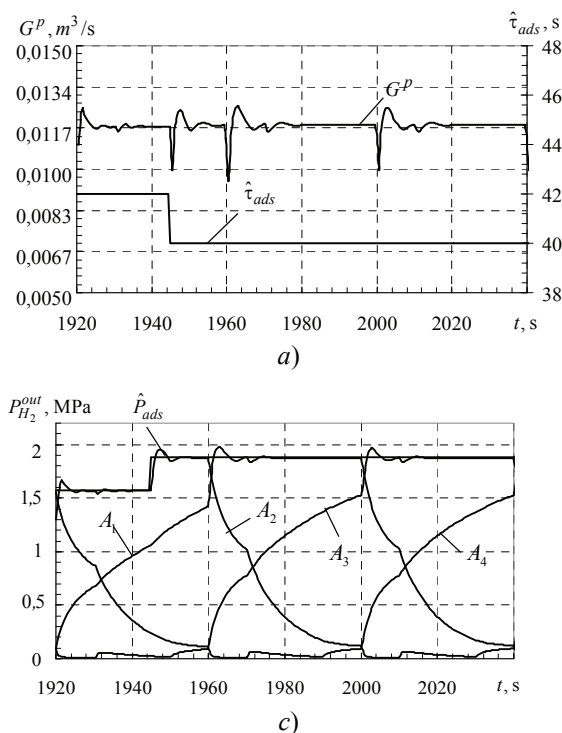
**Fig. 8. Transient processes in the SCADA control system with a stepwise increase in the temperature  $T_g^{in}$  of the initial mixture from 30 to 50 °C:**

*a* – change in the consumption  $G^P$  of production hydrogen and the optimum time of the adsorption cycle  $\hat{\tau}_{ads}$  as a function of time; *b* – the change in concentration  $y_1^{out}$  as a function of time with adaptive optimization (1) and without adaptive optimization (2); *c* – pressure changes in the adsorbers  $A_1$ – $A_4$  of the plant

Fig. 8 shows the graphs of transient processes in the SCADA system of adaptive optimization and control with a stepwise increase in the temperature  $T_g^{in}$  of the initial mixture from 30 to 50 °C. The analysis shows that the SCADA system compensates for the stepwise disturbance by decreasing the duration of the adsorption stage from 42 s to 36 s (Fig. 8 *a*). At the same time, the SCADA system maintains the

concentration of production hydrogen at ~99 % vol. and the consumption  $G^P$  of production hydrogen at the minimum acceptable level (Fig. 8 *a*), while in the PSA plant without adaptive optimization, the concentration of production hydrogen reaches ~94 % vol. (Fig. 8 *b*).

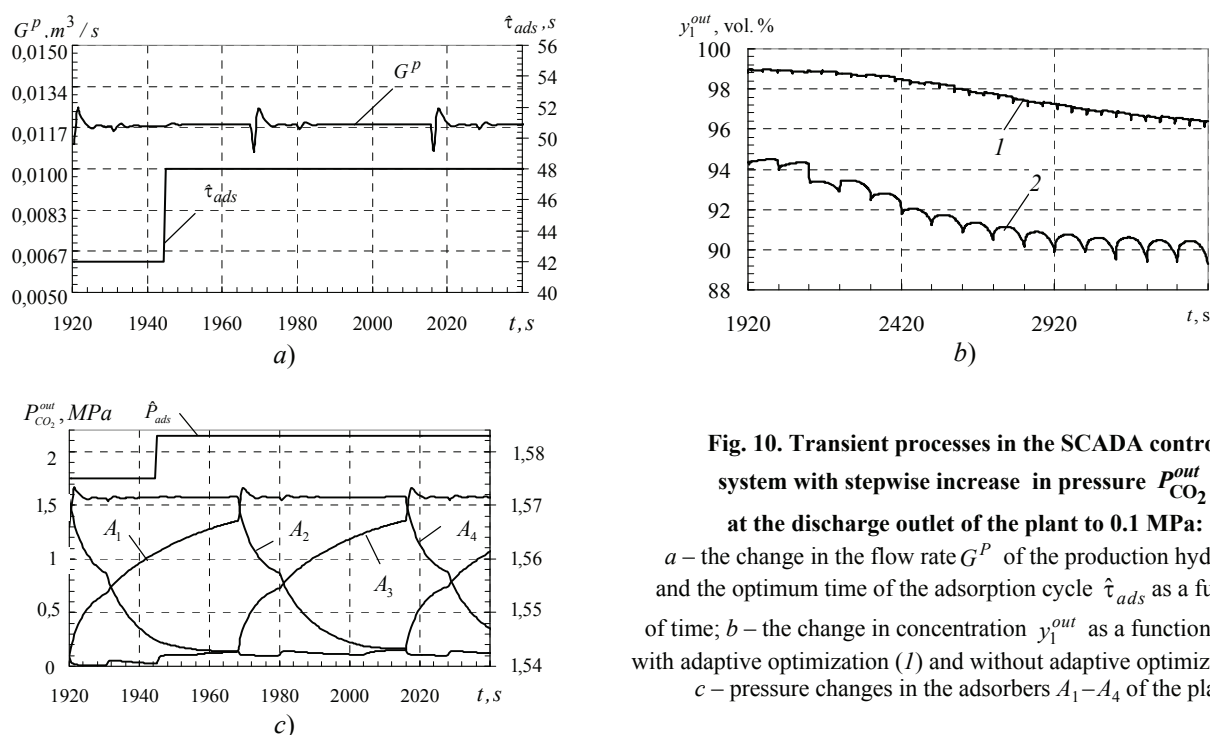
Fig. 9 shows the graphs of the transient processes in the SCADA system with an increase in the pressure  $P_{H_2}^{out}$  at the production outlet of the PSA plant up to



**Fig. 9. Transient processes in the SCADA control system with a stepwise increase in the pressure  $P_{H_2}^{out}$  at the production outlet of the plant up to 0.3 MPa:**

*a* – the change in the consumption  $G^P$  of production hydrogen and the optimum time of the adsorption cycle  $\hat{\tau}_{ads}$  as a function of time; *b* – the change in concentration  $y_1^{out}$  as a function of time with adaptive optimization (1) and without adaptive optimization (2); *c* – pressure changes in the adsorbers  $A_1$ – $A_4$  of the plant





**Fig. 10. Transient processes in the SCADA control system with stepwise increase in pressure  $P_{CO_2}^{out}$  at the discharge outlet of the plant to 0.1 MPa:**

*a* – the change in the flow rate  $G^P$  of the production hydrogen and the optimum time of the adsorption cycle  $\tau_{ads}$  as a function of time; *b* – the change in concentration  $y_1^{out}$  as a function of time with adaptive optimization (1) and without adaptive optimization (2); *c* – pressure changes in the adsorbents  $A_1$ – $A_4$  of the plant

0.3 MPa. In response to this disturbance, the SCADA system increases the adsorption pressure  $P_{ads}$  with an insignificant reduction in the adsorption cycle time  $\tau_{ads}$  (Fig. 8 c), which allows maintaining the production hydrogen flow of the PSA plant within the permissible limits (Fig. 8 b), providing the maximum possible purity of hydrogen  $y_1^{out} = 99\%$  for the changed conditions (Fig. 8 a). In the PSA plant, without adaptive optimization, the purity of production hydrogen is  $y_1^{out} = 95\%$  vol. %.

Fig. 10 a, b, c show the graphs of the transient processes in the SCADA-system with a stepwise increase in pressure at the discharge outlet of the PSA plant up to 0.1 MPa. In the general case, an increase in the discharge pressure  $P_{CO_2}^{out}$  reduces the qualitative indices of the adsorption process (Fig. 10 a, b); the SCADA system of adaptive optimization in this case minimizes the reduction of purity  $y_1^{out}$  of production hydrogen from 99 to 96 vol. %, maintaining the specified limits, while in the PSA plant without adaptive optimization, the purity  $y_1^{out}$  of production hydrogen reduces from 94 to 89 vol. % (Fig. 10 a). The adsorption pressure  $\hat{P}_{ads}$  and the duration of the adsorption stage  $\hat{\tau}_{ads}$  increase in this case, while the consumption  $G^P$  of the production hydrogen remains at the minimum acceptable level (Fig. 10 b, c).

## Conclusion

Using modern methods of systems analysis, mathematical modeling and control theory, new scientific results were obtained for theoretical and practical application of automated processes for adsorption separation of gas mixtures with cyclically varying pressure. We developed mathematical, informational and algorithmic support of a 2-level SCADA-system for adaptive optimization and control over the 4-adsorption PSA plant for hydrogen production with a purity of 96–99 vol. %.

The workability of the developed 2-level SCADA-system of adaptive optimization and control has been confirmed by the simulation method. The efficiency of its functioning under the influence of disturbing actions was verified (the PSA plants with adaptive optimization and without adaptive optimization were compared) for the following conditions: 1) if the proportion of  $CO_2$  in the gas mixture flow and the temperature of the gas mixture at the inlet to the PSA plant increased, the purity of production hydrogen increased by 6.3 and 4.8 %, respectively; 2) if the pressures at the production and discharge outlets of the PSA plant changed, the purity of the target product increased by 3.8 and 6.3 vol. %, respectively.

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## Oxygen-Enriched Air Production System

### Designated purpose, application area.

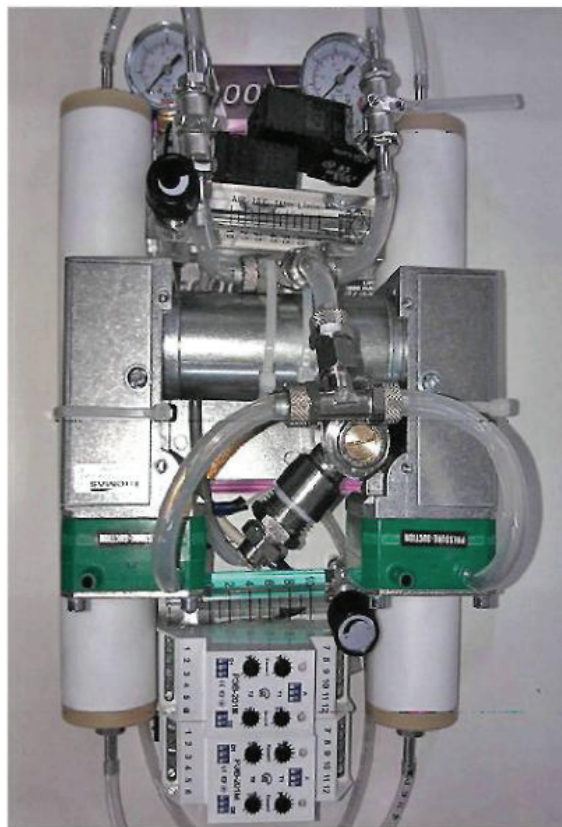
The system allows producing oxygen-enriched air by the pressure swing adsorption method, and simultaneous air drying and cleaning from gaseous contaminants. The oxygen-enriched air production system is intended for industrial dehumidifiers, oxygen generators, medical oxygen concentrators.

### Originality, uniqueness.

The oxygen-enriched air production system in which technology of pressure swing adsorption is applied, allows producing oxygen-enriched air by stepped pressure change in adsorbers without installation of additional heating elements. It provides the possibility of prolonged work (up to 30 000 hours) and reduces electrical energy consumption.

### Specifications.

The implementation of the technology allows producing oxygen-enriched air stream free from gaseous impurities, dried until the dew point of  $-40^{\circ}\text{C}$ , containing from 30 % (at a rate of 10–12 l/min output) to 90 % oxygen (at a rate of 2–3 l/min at the output) with energy consumption 100–150 watts.



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