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WASTE GASES CLEANING AT THE PRODUCTION OF FERRUM OXIDE PIGMENT USING HORIZONTAL APPARATUS WITH BUCKET-LIKE DISPERSERS

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Abstract. The efficiency of horizontal apparatus with bucket-like dispersers (HABD) for waste gases cleaning from Fe_2O_3 dust at the production of red ferrum oxide pigment has been proved by the experiments. It is advisable to carry out the dust trapping in HABD under countercurrent mode. Such mode allows to condense steam from the gas phase and trap small particles of the dust. The positive effect of surface active substances on the dust trapping has been shown. Technologically feasible concentrations of polyacrylamide have been determined. The obtained results may be used for the development of waste gases cleaning technology at the production of ferrum oxide pigment.

Keywords: dust trapping, ferrum oxide pigment.

1. Introduction

Ecological monitoring of red ferrum oxide pigment production at PJSC "Krymsky Titan" exhibits that Fe_2O_3 dust content in the waste gases is considerably higher than actual norms of maximal allowable concentrations (MAC) of polluting compounds from stationary sources. The existing two-staged plant does not meet the modern requirements concerning MAC and is not upgradable. Therefore the problem of waste gases cleaning demands new technological solutions with the application of modern apparatus. On the basis of theoretical foundations for heterogeneous gas systems separation the application of cleaning wet method was grounded using HABD as the main apparatus [1].

Theoretical analysis of HABD operation shows its high efficiency for cleaning of gases emitted from the red ferrum oxide pigment ovens from Fe_2O_3 particles. Previously [2] on the basis of obtained mathematical dependencies and using reference data about physical properties of gas, liquid and solid phases we calculated main indices of the waste gases cleaning of the mentioned process. They were the ground for process simulation and determination of the apparatus optimum mode. A number of assumptions and approximations have been taken so it was necessary to conduct the experiments to clarify the theoretical results.

Taking into account that the investigated waste gases are complex heterogeneous multi-component system with variable parameters, it is impossible to simulate them under the laboratory conditions. Therefore the experiments were carried out in the workshop of JSC "Krymsky Titan" using enlarged experimental setup.

The aim of the work is to study the effect of main operational parameters on the efficiency of Fe_2O_3 dust trapping by HABD.

2. Experimental

2.1. Theoretical Part

The main attribute of purified gas efficiency is practical degree of dust trapping X_p, %, which is determined according to Eq. (1):

$$X_P = \frac{G_i - G_f}{G_i} \cdot 100 \qquad (1)$$

where G_i and G_f – initial and final amount of dust, respectively, kg/s.

Taking into account that $G = z \cdot W_g$, we may represent Eq. (1) as follows:

$$X_P = \frac{z_i - z_f}{z_i} \cdot 100 = \left(1 - \frac{z_f}{z_i}\right) \cdot 100 \qquad (2)$$

where W_g – gas volumetric rate, nm^3/s; z_i and z_f – initial and final dust content, respectively, kg/nm^3.

On the basis of the theoretical analysis of HABD operation [2] the mathematical dependence is derived allowing to calculate the theoretical degree of dust trapping X_T

$$X_T = \left(1 - \frac{z_f}{z_i}\right) \cdot 100 = \left[1 - exp\left(-\frac{3}{2} \cdot \frac{W_l \cdot \Delta R}{W_g \cdot d_d} \cdot h_t\right)\right] \cdot 100 \quad (3)$$

where DR – height of active flushing sector ($\Delta R = R_{app} - R_{dis}$); R_{app} and R_{dis} – radiuses of disperser and apparatus, respectively, m; W_l – liquid volumetric rate, m^3/s; d_d – drop diameter, m; η_t – coefficient of particles trapping.

Physical processes occurred in HABD are complicated, the investigated system is multifactor, and thus the following assumptions were taken: all formed drops have the same size and do not interact between each other; there is no mass-heat exchange between the drop and gas; the drops secondary crushing as a result of knock is neglected. Taking all the mentioned facts into account, the suggested equation is approximate one. However it could help to choose the main factors affecting the apparatus efficiency and conduct purposeful experiments.

One can see from Eq. (3) that cleaning efficiency decreases with the increase in drop diameter d_d and increases with the increase in the height of active flushing sector DR, specific consumption of dispersed liquid W_l/W_g and coefficient of trapping η_t.

The height of active flushing sector is determined by the kinetic energy of dispersed drops and constructive peculiarities of the apparatus. Taking into account the results of previous investigations [3] we used the apparatus with optimum ratio R_{app}/R_{dis}, so there was no purpose to examine the effect of DR on the coefficient of trapping.

The diameter of primary drops formed during HABD operation depends on liquid (water) physical characteristics (σ_l, μ_l, ρ_l), constructive peculiarities of bucket-like disperser (δ_f) and linear rates of its ends (v_l).

$$d_d = 81 \cdot \frac{s_l^{0.46} \cdot d_f^{0.46} \cdot m_l^{0.08}}{n_l \cdot r_l^{0.54}} \quad (4)$$

where σ_l – coefficient of surface tension, N/m; μ_l – dynamic coefficient of viscosity, Pa·s; ρ_l – liquid density, kg/m^3; δ_f – film thickness of dispersed liquid, m; v_l – linear rate of disperser ends [4].

All physical properties of dispersed water (σ_l, μ_l, ρ_l) depend on its temperature. The increase in temperature leads to the decrease in drop average diameter caused by sharp decrease in water viscosity and surface tension and slight change in density. During HABD operation the water temperature (T_l) approximates to the temperature of wet thermometer (T_w), which depends only on gas temperature and its moister content. Therefore, during HABD standard operation mode the water temperature is constant and has no influence on drop size.

The increase in linear rate of disperser ends considerably decreases the average diameter of drops and sharply increases specific power consumption. Moreover, small drops ($d_d < 0.8 \cdot 10^{-3}$ m) are hardly trapped by technological reasons. Therefore the rate of disperser ends is determined by kinetic energy of dispersed liquid providing drop ascent at the height DR and their secondary crushing as a result of collision with apparatus walls. For the industrial devices the advisable linear rate of the disperser ends is within 10–12 m/s [2, 3].

The specific consumption of dispersed liquid W_l/W_g is a parameter having an essential influence on dust trapping that is easy controlled within the wide range. In the technological process the gas volumetric rate is actually constant and linear rate of disperser ends varies within narrow range (10–12 m/s). Therefore the specific consumption of sprayed liquid may be controlled by the number of dispersers and their width.

In HABD the kinematical coagulation of dust particles and liquid drops takes place *via* inertial mechanism. The coefficient of particles inertial trapping is a function of Stokes criterion and for turbulent conditions takes the following view:

$$h_{Stk}^{turb.} = \frac{Stk^2}{(Stk + 0.5)^2}; \; Stk_{cr} = 0.0417 \quad (5)$$

There is a critical minimum value of Stokes number Stk_{cr}, hence dust inertial trapping is possible only on condition that $Stk > Stk_{cr}$.

Stoke criterion describes a ratio between inertia forces of dust particles and medium resistance

$$Stk = \frac{r_p \cdot d_p^2 \cdot n_d \cdot C_k}{18 \cdot m_g \cdot d_d} \quad (6)$$

where ρ_p – dust particle density, kg/m^3; d_p – dust particle diameter, m; C_k – Kaninhem-Millikan correction considering mobility of small particles ($(0.1–2) \cdot 10^{-6}$ m), the size of which is comparable with the length of free path of gas molecules; v_d – velocity of drop motion, m/s [5].

In the first approximation we adopt that $n_d = n_{dis}$. Under constant temperature, drops size and rate of disperser ends the diameter of dust particles is a determinative factor affecting the value of Stokes criterion. With the decrease in dust particles diameter the values Stk and $h_{Stk}^{turb.}$ sharply decrease. To evaluate the efficiency of inertial trapping we made calculations on condition that HABD is used at industrial-scale plant of red ferrum oxide pigment production (T = 373 K, n_d = 10 m/s). So, for the particles of Fe_2O_3 dust with the diameter $d_p = 2.5 \cdot 10^{-6}$ m, which is calculated according to Eqs. (5) and (6), the efficiency of inertial trapping is $h_{Stk}^{turb.} = 0.39$ (39 %), and for $d_p = 1.0 \cdot 10^{-6}$ m – $h_{Stk}^{turb.} = 0.05$

(5 %). The particles with diameter of $0.5 \cdot 10^{-6}$ m are not trapped by drops, because the calculated value $Stk = 0.041$ is less than Stk_{cr}. The calculations show that the inertial mechanism is low-effective to trap dust small particles. Thus, to increase the trapping efficiency it is necessary to realize other effective mechanisms of small particles trapping.

While developing HABD model we admitted that there is no heat and mass exchange between drops and gas. However, these processes proceed in fact. Depending on water steam partial pressure in hot gas, the trapping process may be accompanied by diffusiophoresis – particles motion caused by a gradient of components concentration. During evaporating water from the drops surface the gradient of steam concentration occurs but since the total pressure of the gas phase is constant, the formed hydrodynamic flow of steam-gas mixture is directed along the normal out from the drop surface. During condensation the flow direction is reverse. Such hydrodynamic flow is called Stefan's flow and it has a great influence on the small particles sedimentation. While evaporating drops of the dispersed liquid Stefan's flow prevents from trapping of Fe_2O_3 dust small particles by liquid drops and in case of condensation – promotes this process.

Evaporation cooling takes place if hot gas unsaturated by water steam contacts with water, *i.e.* meets the following conditions:

$$T_g > T_l \quad \text{and} \quad P_g < P_l$$

where T_g and T_l – temperatures of waste gases and water, respectively, K; P_g and P_l – partial pressures of water steam in waste gases and water, respectively, Pa.

After reaching the temperature of wet thermometer (T_w) the water stops to be heated and will be only evaporated at the constant temperature. The heat transferred from gas to water returns back to the gas together with formed water steam, *i.e.* the process proceeds with constant gas enthalpy.

Condensation cooling takes place if hot gas saturated by water steam contacts with cold water ($T_g > T_l$, $P_g > P_l$). A part of water steam in gas is condensed; gas is cooled and dried; water is heated to the temperature of wet thermometer. During the process the enthalpy and water content decrease.

The efficiency of condensation diffusiophoresis may be fully used in the case of countercurrent mode of gas and liquid phases flow. For this purpose at the HABD inlet the hot gas should be washed by water heated to the temperature of wet thermometer. This eliminates the mode of gas evaporation cooling, which worsens solid particles trapping by water drops. At the outlet the condensation cooling of gas by fresh water should be provided allowing gas aftertreatment due to the ordered motion of fine-dispersed gas components to the drops surface. The mentioned mode may be created in HABD if we place some dispersers on the horizontal arbor. The dispersers should provide necessary water concentration and form necessary amount of drops. Thus the apparatus operation approximates the model of multistaged reactor, where controlled countercurrent mode is easy to be realized.

Taking all the mentioned above into account it is advisable to carry out experiments concerning the effect of diffusiophoresis on the efficiency of dust trapping in the waste gases after the production of red ferrum oxide pigment.

Eq. (3) sets aside the efficiency of contact between drops and dust particles. While developing model it was assumed that the contact between dust particle and drop is finished by its trapping (adhesion). Such boundary case is possible if dust particles are wetted by drops. For hydrophilic particles the necessary condition for their trapping is a point contact (touch) between the particle and the drop; for hydrophobic particles the complete penetration of the particle into the drop is required. To regulate wetting process the modern plants use surface-active substances (SAS) [6]. SAS molecules sorbed at the boundary "liquid–solid particle" decrease the surface tension and wet angle. As a result, the wetting process and dust trapping proceed with higher efficiency. Fe_2O_3 particles belong to the hydrophobic ones, therefore it is advisable to exam the effect of SAS concentration on the degree of dust trapping in HABD.

2.2. Practical Part

2.2.1. Experimental setup and procedure

The experimental setup was built directly at the PJSC "Krymsky Titan" (unit of ferrum oxide pigments production) so that the main experiments are carried out using real gases. Such approach allows to obtain the reliable experimental results necessary to develop the recycling technology for waste gases from ferrum oxide pigment production. The scheme of the experimental setup is represented in Fig. 1.

Waste gases from the rotary oven used for the ferrum oxide pigment paste calcinations are fed to HABD (2) through the inlet line (1). Absorber (2) is a horizontal cylindrical apparatus with the internal diameter of 500 and the length of 700 mm (absorption volume is 0.13 m^3). The arbor (3) with three bucket-like dispersers (4) is placed in the lower part of absorber. The sizes of bucket-like disperser, mm: diameter – 200; width – 15 and width of the spraying slits – 2. The apparatus is equipped with fittings for inlet and outlet lines, as well as with sockets for thermometer and sampling. To control the process the absorber is equipped with three observations ports. The arbor is set in motion by electric motor (10) and pulley (5).

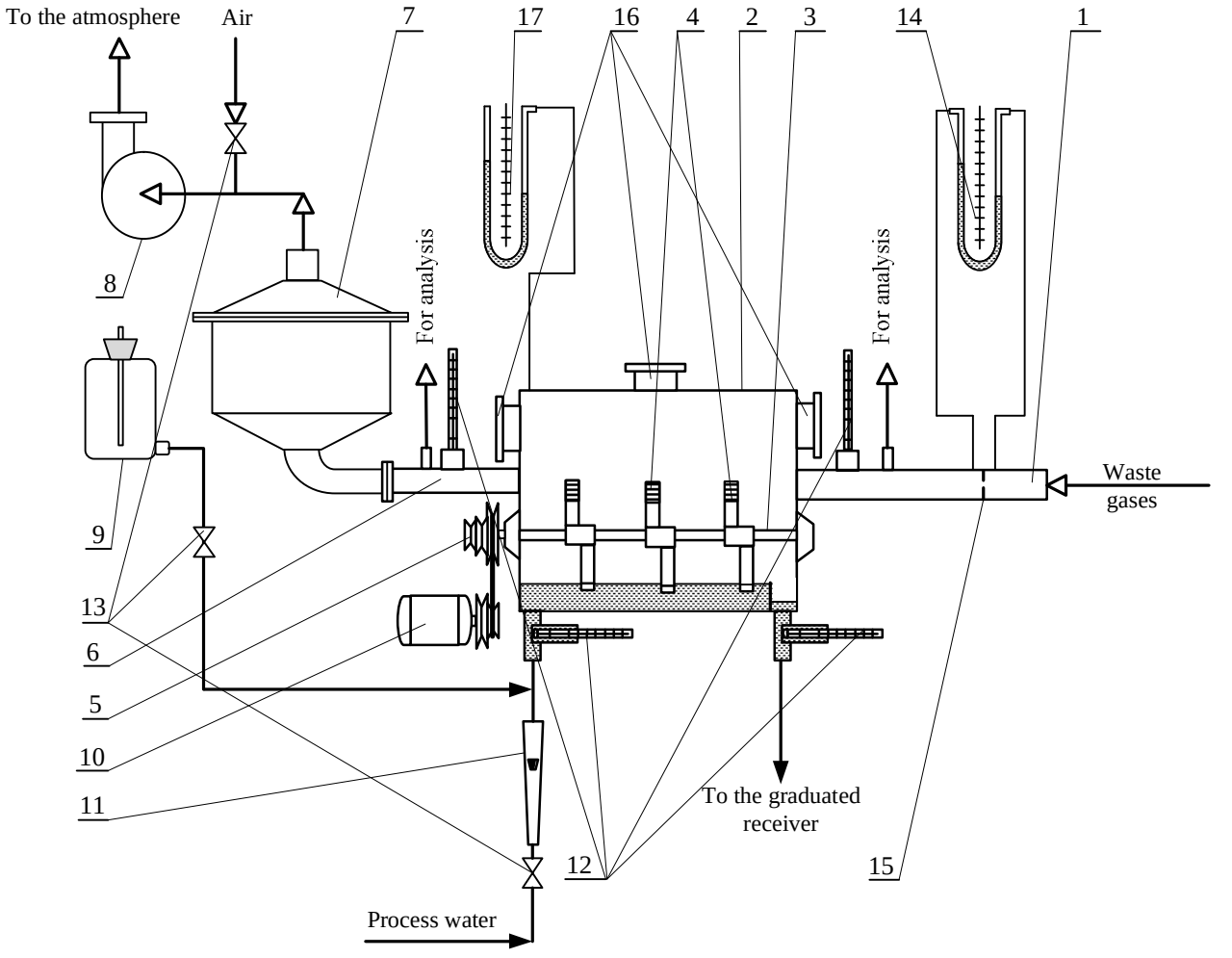

Fig. 1. Scheme of the experimental setup to study waste gases cleaning at the production of ferrum oxide pigment: inlet line (1); horizontal absorber (2); arbor (3); dispersers (4); pulley (5); outlet line (6); drops catchers (7); tailing fan (8); vessel for SAS (9); electric motor (10); rotameter (11); thermometers (12); stop valves (13); differential manometer (14); restriction (15); observation ports (16); and manometer (17)

The purified waste gases are carried off to the atmosphere using outlet line (6), drop catcher (7) and tailing fan (8). To introduce SAS the vessel (9) is used. The waste gases volume is measured using calibrated aperture (15) equipped with differential water manometer (17). The pressure is measured by differential manometer (17) and water consumption – by rotameter (11). All temperatures of the flows are determined by liquid thermometers (12) and all flow rates – by corresponding stop valves (13).

The procedure was as follows. Waste gases from the oven are sucked in the absorber by the fan. Gas flow rate is controlled by stop valve. Then we switch on the arbor drive and set the flow rates of water and polyacrylamide (PAA) solution (0.03 wt %). The latter one was prepared from 6% PAA solution used at the production of ferrum oxide pigment.

The experimental condition are: process water flow rate $(3.3–137)\cdot10^{-6}$ m^3/s; water input temperature 283–295 K; gas flow rate $6.67–48.1)\cdot10^{-3} nm^3/s$; dust concentration in the waste gases $(600–1800)\cdot10^{-6}$ kg/nm^3; number of dispersers 3; linear rate of disperser ends 10 m/s; total efficiency of three dispersers $1.7\cdot10^{-3}$ m^3/s.

W_l/W_g ratio is controlled by change of gas flow rate. The intensity of evaporation and condensation processes is controlled by change of water input temperature and its flow rate.

Samples of water and gas at absorber outlet (to analyze Fe_2O_3 dust concentration) are withdrawn after the moment when steady temperature conditions are set in the absorber. Usually it takes 50–60 min under stable input parameters. Gas sampling is realized by gas sampler Taifun P20-2-2 allowing to control and stabilize rate of gas sampling.

The analysis of gas phase for dust content is carried out by means of "internal filtration" (filter was weighted before and after the experiment). The mentioned procedures are used by workers of PJSC "Krymsky Titan" to control the production. The experimental results were recalculated taking into account gas parameters at the moment of sampling in accordance with standards [7]. The represented results are averaged.

2.2.2. Calculations of experimental results

The main factors characterizing the efficiency of gas cleaning from solid particles are the content of solid particles in the purified gas, degree of dust trapping and intensity of apparatus operation. According to the active standards concerning boundary permissible concentrations (BPC) the dust content cannot exceed $50 \cdot 10^{-6}$ kg/nm^3 [8]. Taking into account that during the experiments maximum content of Fe_2O_3 dust is $1800 \cdot 10^{-6}$ kg/nm^3, the degree of dust trapping is:

$$X_P \geq \frac{z_i - z_p}{z_i} \cdot 100 = \frac{1800 - 50}{1800} \cdot 100 = 97.2\% \qquad (7)$$

Intensity of apparatus operation (I, kg/(s·m^3)) is used to calculate the design apparatus volume. So intensity is the ratio between the weight of trapped dust for contact time and apparatus capacity.

$$I = \frac{G_i \cdot X_P / 100}{V_{app}} = \frac{z_i \cdot W_g \cdot X_P / 100}{V_{app}} \qquad (8)$$

where V_{app} is apparatus capacity, m^3.

The direct investigation of Stefan's flow hydrodynamics effect on the process of dust trapping in HABD is impossible from the technical point of view. Therefore the effect of diffusiophoresis was studied indirectly, introducing such index as condensation intensity (Δg, kg/(s·m^3)). It means the weight of condensed water steam for time unit in the unit of apparatus volume. The intensity was calculated on the basis of the following assumptions: during water intensive dispersion in the apparatus gas achieves saturation relative to water steam; under countercurrent flow of phases in HABD the simultaneous condensation and evaporation cooling of gas are possible. Thus, the condensation intensity is calculated as a sum of condensation and evaporation processes according to Eq. (9):

$$\Delta g = \frac{(x_{in} - x_{out}) \cdot G'_g}{V_{app}} = \frac{(x_{in} - x_{out}) \cdot W'_g \cdot M_g}{V_{app} \cdot 22.4} \qquad (9)$$

where x_{in} and x_{out} – gas water content at HABD inlet and outlet, kg/kg; G'_g – mass flow of dry gas, kg/s; W'_g – volumetric flow of dry gas, nm^3/s; M_g – molecular weight of dry gas, kg/mol.

If condensation is a dominating process in HABD ($x_{in} > x_{out}$), the intensity is a positive value ($\Delta g > 0$). When evaporation predominates ($x_{in} < x_{out}$), the value is negative ($\Delta g < 0$).

Gas water content is calculated according to Eq. (10):

$$x = \frac{p_p \cdot M_{H_2O}}{(p - p_p) \cdot M_g} \qquad (10)$$

where p_p – partial pressure of saturated water steam, Pa; p – total pressure in the apparatus, Pa; M_{H_2O} – water molecular weight, kg/mol. The value of partial pressure at HABD inlet is taken for the temperature of water at the apparatus outlet and the value of partial pressure at HABD outlet – for the temperature of gas at the outlet.

3. Results and Discussion

The results of investigations concerning the effect of dispersed liquid on the dust trapping are represented in Fig. 2. The increase in specific consumption of dispersed liquid W_l/W_g stably increases the theoretical value X_t of dust trapping degree calculated according to Eq. (3) and practical (experimental) one X_p. At the same time, at the water concentration of $50 \cdot 10^{-3}$ m^3/nm^3 the theoretical value approximates to 100 %, the practical one – only to 97 %. Such inequality may be explained by low efficiency of small particles trapping ($d_p \leq 2.5 \cdot 10^{-6}$ m). The intensity I sharply decreases with the increase in water concentration. At stable flow rate of dispersed liquid phase W_l the water concentration takes place due to the decrease in gas volumetric flow rate W_g and amount of dust introduced into the apparatus together with gas, because $G_i = z_i \cdot W_g$. Irregularity of intensity decrease is caused by polydispersion of red pigments dust and dust content in gas depends on particles hovering velocity which, in turn, is proportional to the square of particle diameter d_p. That is, the linear rate of gas decreases as a result of its volumetric flow rate decrease and large particles of dust are not introduced into the apparatus. So, the dust content in gas decreases as well.

The experimental results show the principal possibility of achieving necessary degree of dust trapping (97.2 %), but for achieving this value water concentration should be $\geq 70 \cdot 10^{-3}$ m^3/nm^3. The increase in water concentration needs additional power consumption and is accompanied by sharp decrease in the intensity of dust trapping leading to the increase in HABD capacity.

Taking all the above-mentioned into account we studied the effect of diffusiophoresis on the efficiency of dust trapping at the specific consumption of $\approx 50 \cdot 10^{-3}$ m^3/nm^3. Considering the capacity of experimental HABD of 0.13 m^3, the time of gas stay in the apparatus is 3.8 s. The experimental results are represented in Fig. 3.

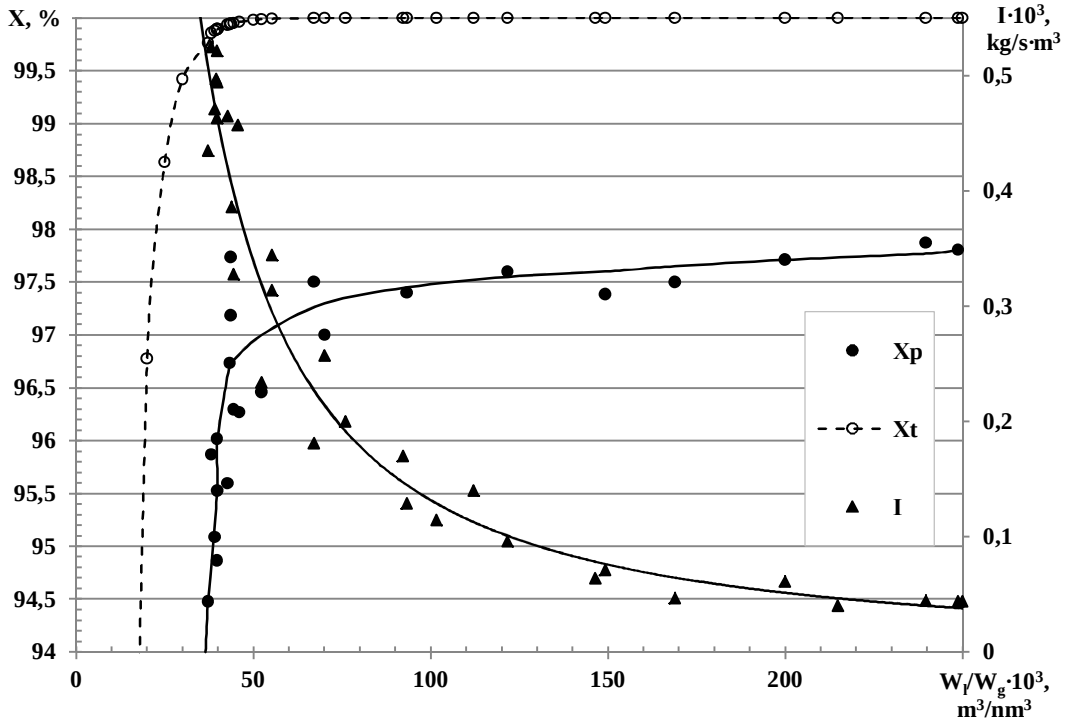

Fig. 2. Dependence of intensity I, theoretical X_t and practical X_p degree of dust trapping on specific consumption W_l/W_g of the dispersed liquid

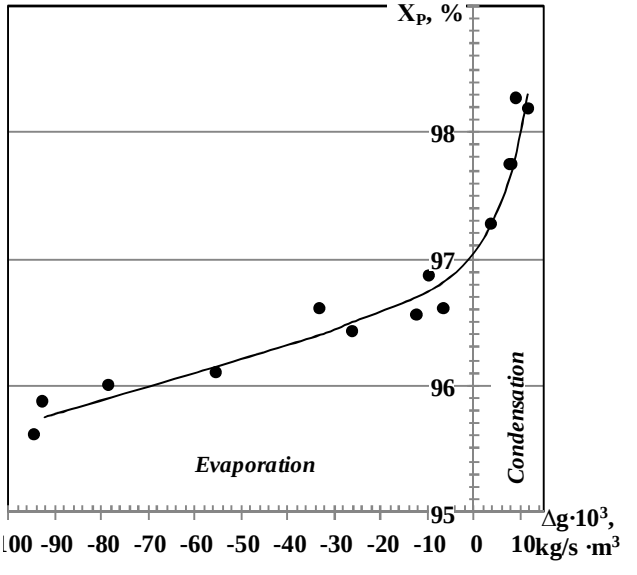

Fig. 3. Degree of dust trapping *vs.* condensation intensity

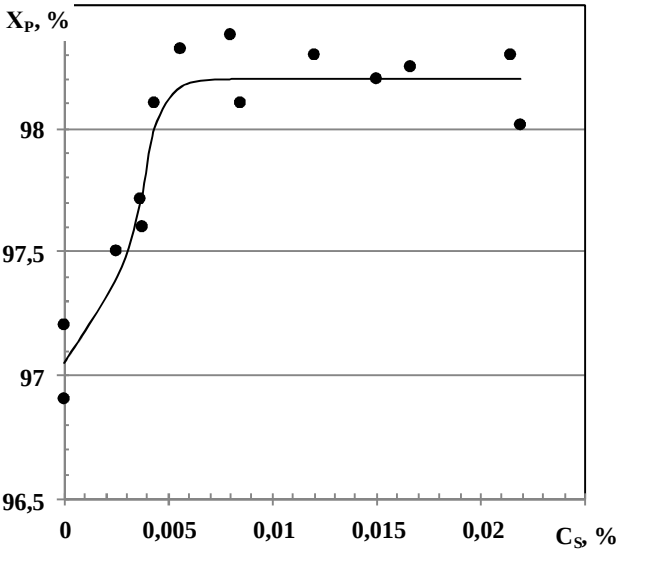

Fig. 4. Degree of dust trapping *vs.* SAS concentration

We proved by the experiments that condensation process has a positive influence on the degree of dust trapping and evaporation has a negative effect. The maximum degree of dust trapping is 98.2 % at the condensation intensity Δg=10·10^{-3} kg/(s·m^3), though the calculated maximum value is $\Delta g \approx 60 \cdot 10^{-3}$kg/(s·m^3). Higher values of condensation intensity were not achieved related to inaccurate modeling of heat flows in the experimental setup and heat loss to the environment. The experimental setup was mounted in an accessible and safe

place for research and long non-insulated gas flue was supplied. The heat losses in the gas flue were significant. So, gas was fed to HABD not with the temperature of 603–653 K (gas temperature at the furnace outlet) but the temperature of 413–463 K. That is why the water temperature at the apparatus outlet was lower (317–328 K) than that designed for the industrial plants (~ 343 K). The water temperature at the outlet approaches to the temperature of wet thermometer and determines water content in gases. Even slight increase in temperature leads to a noticeable increase in water content of gas, and then – to intensification of the condensation process. Moreover, it is difficult to reproduce polythermal mode in a small experimental apparatus where gas evaporation cooling takes place at the first bucket-like disperser, the washing of cooled gas by water heated to the temperature of wet thermometer – at the second one and condensation cooling of gas by fresh water – at the third one. In the industrial unit the amount of dispersers, the distance between them and the time of contact between the gas and liquid drops will increase, so operation mode will approach the regime of multistaged reactor. Therefore there is every reason to expect improved cleaning results.

The following stage of investigations was the determination of SAS concentration effect on waste gases cleaning. The specific consumption of dispersed liquid was $50 \cdot 10^{-3}$ m^3/nm^3, the condensation intensity was 0 $kg/(s \cdot m^3)$. In order to minimize the interference in the existing technological process we used the 6% solution of polyacrylamide (PAA) as SAS, because just this component is used for the production of red ferrum oxide pigment

The results are represented in Fig. 4. The increase in SAS concentration in the liquid phase to 0.006 % increases the gas cleaning efficiency from 97.0 to 98.2 %. The further increase in SAS concentration does not lead to the positive effect. Therefore the technologically advisable SAS concentration is within the range of 0.006–0.008 %. Such character of dependence may be explained as follows: SAS concentration at the phase boundary is few times higher than that in the solution volume, hence even low SAS concentration leads to the significant decrease in water surface tension. The increase in SAS concentration decreases surface tension to some minimum value and then there is no decrease.

The introduction of SAS into the system, apart from the increase in degree of dust trapping, stimulates Fe_2O_3particles sedimentation in the suspension obtained as a result of HABD operation. At SAS concentration of 0.006–0.008 % there is an agglomeration of dispersed particles in the water phase due to which their sedimentation rate is within $(3.3–5.0) \cdot 10^{-3}$ m/s, that allows to separate solid and liquid phases and return them back for the production of red ferrum oxide pigment.

4. Conclusions

1. To trap the small particles of Fe_2O_3 dust and to increase the cleaning efficiency in HABD it is advisable to realize the counterflow of phases with predominant mode of gas condensation cooling.

2. SAS solutions stimulate dust trapping process, in particular using PAA with the concentration of 0.006–0.008 % for HABD flushing provides high cleaning efficiency (98.2 %) and intensifies sedimentation of trapped dust particles in the liquid phase.

3. The adequacy of proposed and developed conception of waste gas cleaning at the production of red ferrum oxide pigment is proved by the experiments and may serve as a base for technological flowsheet development.

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ОЧИЩЕННЯ ВИКИДНИХ ГАЗІВ ВИРОБНИЦТВА ФЕРУМОКСИДНОГО ПІГМЕНТА У ГОРИЗОНТАЛЬНОМУ АПАРАТІ З КОВШОПОДІБНИМИ ДИСПЕРГАТОРАМИ

Анотація. *Експериментально підтверджено ефективність горизонтального апарата з ковшоподібними диспергаторами (ГАКД) для очищення викидних газів виробництва червоного ферумоксидного пігмента від пилу Fe_2O_3. Встановлено, що процес пиловловлення у ГАКД доцільно проводити в протитечійному режимі, що забезпечить конденсацію водяної пари з газової фази та вловлення дрібних частинок пилу. Показано позитивний вплив на процес вловлення пилу поверхнево-активних речовин, встановлено технологічно доцільні концентрації поліакриламіду. Отримані результати можуть бути використані в процесі розроблення технології очищення викидних газів виробництва червоного ферумоксидного пігмента.*

Ключові слова: *пиловловлення, ферумоксидний пігмент.*