

Gas Purification and Quality Control of the End Gas Product

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Abstract

One of the main problems in the flow-through gas purification technologies is related with continuous control of the outlet gas purity. The information concerning purity of the produced gas is on high demand, e.g., for processing systems integrated with gas purifiers. The positive solution of this problem has become possible only now due to the appearance of reactive getters (reactants) that serve as highly efficient sinks for gas impurities and our sorption model of the processes, which take place in gas purifiers with these reactants. According to the given model the appearance of a single valued functional connection between the purity of the gas product and the duration of the treatment of the gas flow by the sorbing powder is typical for any system Me-Y, where Me is a powder reactant and Y is an impurity gas. This strict correlation provides the mathematical justification to a simple method of determining the concentration of the impurity in the gas flow at the exit from the gas purifier. This method comes down to measuring of the quantity of the purified gas by a gas flow meter, the readings of which are graduated in the units of gas concentration.

Keywords

Gas Purifier, Reactants, Sorption Model, Quality Control, Purity Indicator

1. Introduction

In the production of high purity gases an important place belongs to gas purifiers, which in the essence are flow-through tubes by themselves with a sink for capturing impurities from the treated gas. The molecular sieves or getter materials are commonly used for this purpose in powder form or in the form of

sintered porous bodies [1] [2] [3]. The weak points of today's purification methods include low sorption capacity and quality control of the end gas product. The practically achieved sorption capacity of the traditional getter materials is rather low at room temperature [4] [5]. The situation is no better with the purity control where the cost of the precision analytical equipment and measurements by many times exceed the cost of gas purifiers and gas production.

Therefore, the progress in this field can be feasible only with drastic improvements in the capturing capacity of getters and simplification of the methods for continuous control of impurities in the outlet gas.

The main step in the indicated direction is the development of getters with sorption capacity much larger compared to similar materials on the basis of transition metals [6] [7]. In order to approach the other side of the problem, *i.e.* purity control, it is worth noticing that for many users of high purity gases it is not particularly important to know exactly the amount of impurities. More vital thing is to have confidence that the impurities content is within the range of the required purity.

The aim of the present paper is to describe the impurity gas capturing with a powder reactant in the flow-through gas purifier. The theory of the process is further used for searching the methods allowing replacing of costly direct measurements with inexpensive indirect ones.

2. Problem Statement

The dynamic sorption of gases by adsorbents is well studied [8] [9] [10] [11]. This cannot be said about gas capturing by reactants (under reactants we will further understand alkali or alkali-earth metals or their alloys [12]). Unlike adsorbents, reactants (henceforth Me) provide the entire volume of the material for capturing gas Y by forming a non-volatile chemical compound MeY (**Figure 1**).

While the surface of adsorbents rapidly passivates in gas medium and gas sorption by adsorbents is limited at room temperature by diffusion and low values of the ultimate solubility of gases in solids, reactants continuously capture gases till the material is entirely exhausted in the chemical reaction. At the limit sorption capacity of reactants is determined by the ratio Me:Y = 1:1, *i.e.* for capturing of one gas atom one metal atom is "spent", which is unique for such application field as gas purification.

The difference between adsorbents and reactants is also significant in respect to the sorption kinetics. At initial stages, when active surface sites are abundant, adsorbents and reactants are similar in their activity. However, the saturation of the surface with impurity adatoms results in practical termination of the sorption process while in the case of reactants it is accompanied only with a change in the intake mechanism. The quantity m of the captured impurity Y at constant concentration in the gas phase may change with time either as $m = k_0 t$

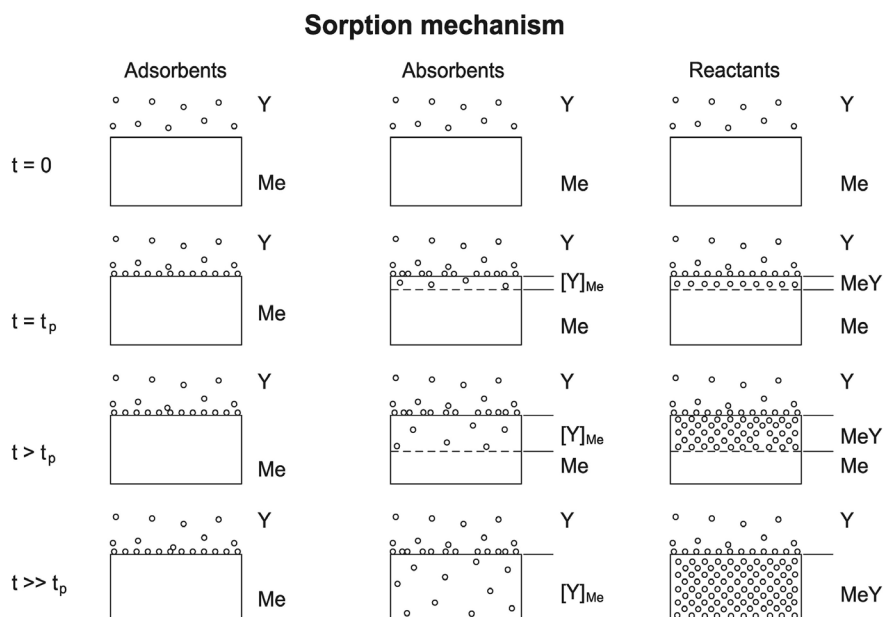


Figure 1. Three getter classes: adsorbents, absorbents and reactants. Me—metal, Y—gas, MeY—chemical compound, $[Y]_{Me}$ —solid solution of Y in Me; $t = 0$, fresh surface of Me, $t = t_p$, passivation time of the adsorbent; $t > t_p$, solubilizing or formation of the compounds in the case of absorbents and reactants, $t \gg t_p$, the state of equilibrium between adsorbents and reactants with gas Y.

or $m = k_p t^{1/2}$, where k_0 and k_p are kinetic constants depending on the reactant nature.

Now we can start solving the problem of gas purification in a flow-through tube with the powder reactant Me. Let's consider an elementary layer dx containing n particles of the reactant limited by coordinates x_1 and $x_2 = x_1 + dx$ along the tube of the length L and radius R . We suppose that the reactant particles are spheres with radius r_0 , and the porosity coefficient of the powder mass is ε . Then the layer volume $V = \pi R^2 dx$ can be presented as a sum $V = V_{Me} + V_Y$, where $V_{Me} = 4\pi r_0^3 n / 3$ is the volume of the reactant and $V_Y = \varepsilon V$ is the volume of voids within the layer, **Figure 2**.

Hence, we find that $n = 3(1 - \varepsilon)R^2 dx / 4r_0^3$ and the total surface of the reactant particles in the elementary layer is $S_{Me} = 3\pi R^2 (1 - \varepsilon) dx / r_0$.

The gas enters the layer with the impurity concentration $c(t, x_1)$ and leaves it with a smaller concentration $c(t, x_2)$. The decrease in the concentration is due to the intake of the impurity by the reactant Me. Let's define the concentration of the impurity in Me as $q(x, t) = m(t, x) / V_{Me}$, where $m(t, x)$ is the amount of the captured gas in the layer dx at time t .

The material balance for the sink of impurities is

$$\varepsilon \frac{\partial c}{\partial t} + v \frac{\partial c}{\partial x} = \frac{\partial q}{\partial t}, \quad (1)$$

where v is the gas velocity in the tube till the entrance into the powder mass, c is the concentration $c(t, x)$ of the gas in voids that separate the particles of

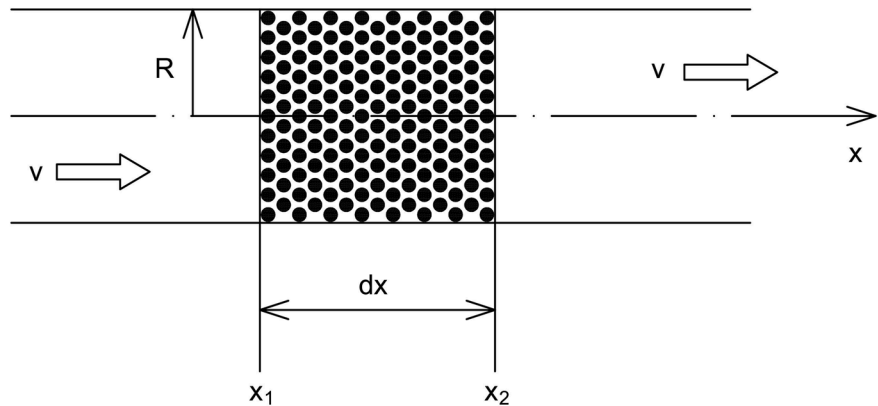


Figure 2. Elementary layer of powder mix in a flow tube.

Me. The right hand part $\partial q/\partial t$ of this equation is related with the intake kinetics.

Let us assume that we deal with a linear sorption law and a chemical reaction of first order. Then the increment of the amount of gas Y that sank in the layer dx during the time dt is to be equal $dm(t, x) = (k_0 S_{Me}(t, x) c(t, x)/c_0) dt$. From here we get that

$$\frac{\partial q}{\partial t} = \frac{3k_0(1-\varepsilon)}{c_0 r_0^3} c(t, x) r(t, x)^2, \quad (2)$$

where $r(t, x)$ is the radius of the active core of the particle at time t , which runs from r_0 to 0 in the intake process.

The value $r(t, x)$, which is a boundary between the reacted and not yet reacted parts of the reactant particle, according to Appendix I can be calculated as

$$r(t, x) = r_0 - \frac{k_0 M_{Me}}{\rho_{Me} M_Y c_0} \int_0^t c(t', x) dt', \quad (3)$$

where ρ_{Me} is the density of the reactant while M_{Me} and M_Y designate molar mass of Me and Y, respectively. Substituting the found value $r(t, x)$ to (2) and then substituting (2) to (1) we come to the main equation of the problem about concentration $c(t, x)$ of gas in the flow tube with powder reactant Me

$$\frac{\partial c}{\partial t} + \frac{v}{\varepsilon} \frac{\partial c}{\partial x} = -\alpha c \left(1 - \frac{\delta}{c_0} \int_0^t c(t', x) dt' \right)^2. \quad (4)$$

Here, $\alpha = 3k_0(1-\varepsilon)/r_0 c_0 \varepsilon$, $\delta = k_0 M_{Me}/\rho_{Me} M_Y r_0$, $t > 0$, and $0 > x > L$.

Equation (4) is to be supplemented with the obvious boundary conditions

$$c(0, x) = 0, \quad (5)$$

$$c(t, 0) = c_0. \quad (6)$$

3. The Model

Let us introduce dimensionless parameters:

$\xi = x/L$: the longitudinal coordinate, $0 < \xi < 1$,

$\tau = tv/\varepsilon L$: the time,

$u = c/c_0$: the gas concentration, $0 < u(\tau, \xi) < 1$,

$A = \alpha \varepsilon L/v$: the sorption parameter,

$B = \delta \varepsilon L/v$: the exhaust parameter.

In this case the mathematical model is transformed to the form:

$$\frac{\partial u}{\partial \tau} + \frac{\partial u}{\partial \xi} = -AuF(\tau, \xi), \quad \tau > 0, 0 < \xi < 1, \quad (7)$$

$$u(0, \xi) = 0, \quad (8)$$

$$u(\tau, 0) = 1, \quad (9)$$

where

$$F(\tau, \xi) = \left[1 - B \int_0^\tau u(\tau', \xi) d\tau' \right]^2 \equiv F[u] \quad (10)$$

is a functional of the dimensionless concentration $u(\tau, \xi)$.

The solution of the problem (7)-(10) was found with the help of an iterative method (see Appendix II). The problem has two controlling parameters: A and B , where A determines the rate of the impurity intake from the gas and B determines the lifespan of the powder reactant. At the stage of the analysis an issue of the time scale of the discussed process arises. As it follows from the model (7)-(10) the solution at the first iteration (formula II.1, Appendix II) describes the steady-state impurity concentration distribution in the conditions of “inexhaustible sink” when function $F(\tau, \xi) = 1$. However in a real sorption process the share of the exhausted powder in different cross sections of the tube increases with τ and this is accompanied with the decrease of function $F(\tau, \xi)$ and decrease of the impurity sink. Characteristic time scale of this decrease is determined by the form of function $F(\tau, \xi)$ and has a value of $\tau = 1/B$.

The calculated time dependent changes for the gas concentration are presented in **Figure 3**, where curve $u_1(t=10, \xi)$ represents the case of “inexhaustible sink”.

Particularly illustrative is the curve $u_2(10, \xi)$, which shows the concentration profile at $\tau = 1/B = 10$. The moment of time $\tau = 10$ is the moment, when all the powder particles at the inlet cross-section $\xi = 0$ occur entirely spent in the reaction with the impurity gas Y. Right after this the mentioned cross section with completely exhausted particles starts its movement inward the tube in the form of a flat front $\Xi(\tau)$, which divides the powder mass into two areas: the area of the completely exhausted powder mass taking the region of the tube $[0, \Xi(\tau)]$ and the area of the capable to react with gases powder mass, which takes the region $(\Xi(\tau), 1]$. As this front approximates the end of the tube the share of the exhausted powder increases and the purity of the exiting from the tube gas decreases: values $u(\tau, 1)$ as seen from **Figure 3** increases together with τ . This behavior is expected and this raises the question about the dependence of the purity of the outlet gas on time.

The plots in **Figure 4** illustrate the convergence of the results in the adopted iterative calculation procedure.

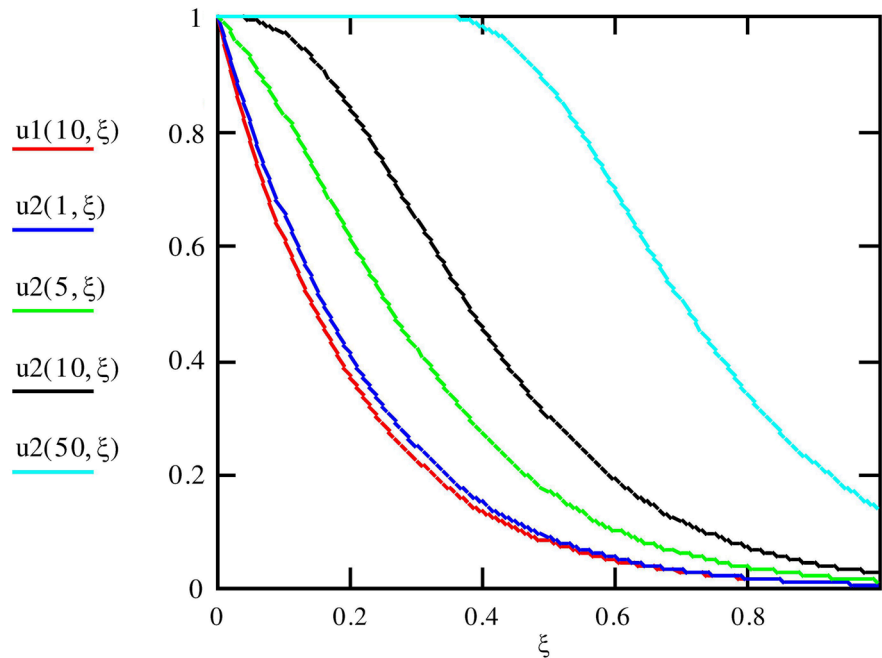


Figure 3. Distribution of gas concentration along the tube at different values of τ . u_1 —the first iteration, u_2 —the second iteration; the initial data: $A = 5$, $B = 10^{-1}$.

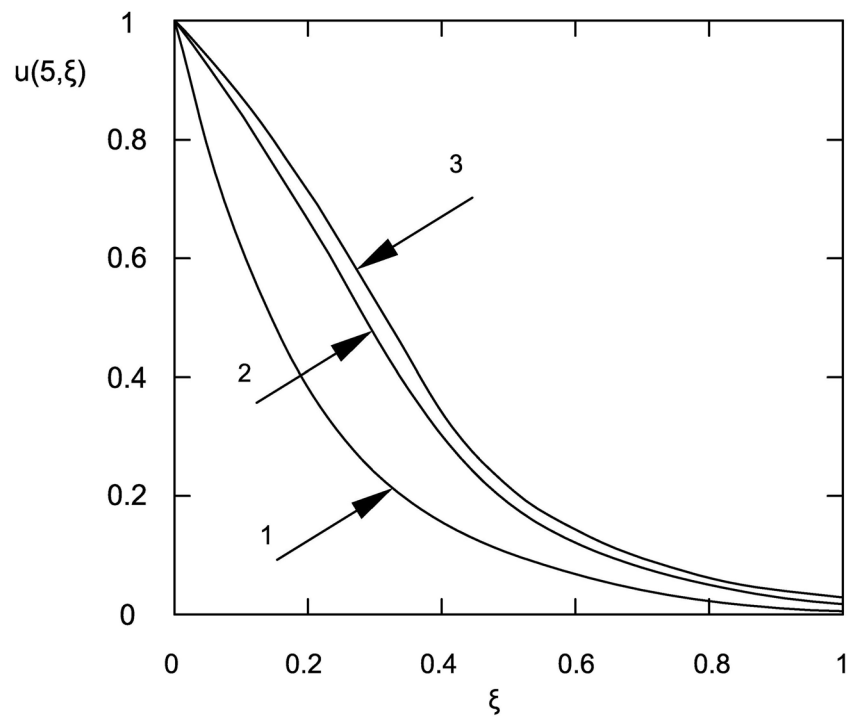


Figure 4. Concentration profile of the gas flow of the tube in three iterations. 1, 2 and 3—the first, the second and the third iterations accordingly; the initial data: $A = 5$, $B = 10^{-1}$.

In fact, it can be seen that curve u_3 is clearly closer to curve u_2 than the latter is to curve u_1 . Let us also have in mind that qualitative behavior of curves repeats for all the iterations. That is, there are all the reasons to believe that the presented here approximation is correct and can be used for searching for correlations between the purity of the end gas product and one or another measurable parameter of the sorption process.

The question about the dependence of concentration Y in the flow of gas in the outlet from the gas purifier on time is the question about the behavior of function $u(\tau, 1)$. **Figure 5** contains the results of calculation of this function according to the formula (II, 2 in Appendix II) for a simple scheme when a tube gas purifier with a powder reactant is installed in a gas line between a CVD chamber and a hydrogen generator. The following data was used for the calculations: $c_0 = 0.05 \text{ mol/m}^3$, $\varepsilon = 0.33$, $r_0 = 10^{-4} \text{ m}$, $L = 0.1 \text{ m}$, $k_0 = 0.14 \text{ mol/m}^2 \cdot \text{s}$ and $v = 0.02 \text{ m/s}$.

The plot for $u = u(\tau, 1)$ is shown in the concentration range that covers usual requirements for the purity of technological gases. If we assume, e.g., that the red line in **Figure 5** corresponds to maximum permissible concentration u_c then the projection of the intersection point of the red line with curve $u(\tau, 1)$ onto the axis τ will determine the lifetime τ_c of the given gas purification tube. That is, the problem of tracking the quality of gases treated by gas purifiers is in principle solvable and the solution follows directly from the described sorption model.

As regards the degree of purification of the gas flow, its maximal value according to Appendix II is limited by e^{-A} . From here for the mentioned above case of the CVD chamber with a hydrogen generator we get at $A = 10$ that

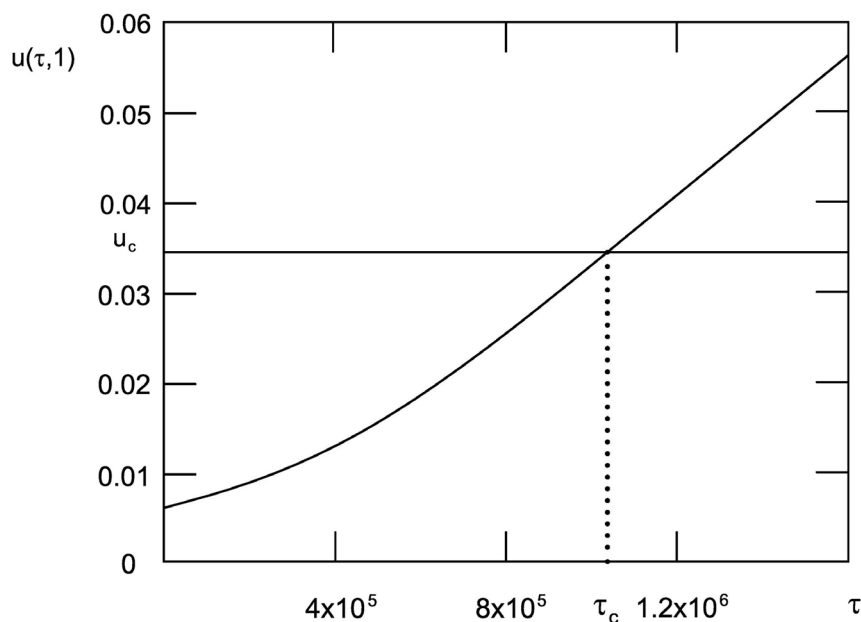


Figure 5. Dependence of the purity of gas product on time τ . The initial data: $A = 10$, $B = 2 \times 10^{-6}$.

the first portions of purified by the reactant gas will have the purity of about 99.9999995%.

For better understanding let us go back to $u_2(10, \xi)$ to show in a vivid form how the sorption process is realized in powder reactants (**Figure 6**).

In the given figure a chain of situated along axis ξ spherical particles in contact with each other is depicted below curve u_2 (**Figure 6(b)**). Here the fact of the correspondence between the structural phase relations in sorbent particles and the values of gas concentration presented by curve $u_2(10, \xi)$ deserves attention.

4. Discussion

Function $u = u(\tau, 1)$ depends on the nature of Me and Y, and the curve $u(\tau, 1)$ is strictly reproducible when the process is standardized. In other words,

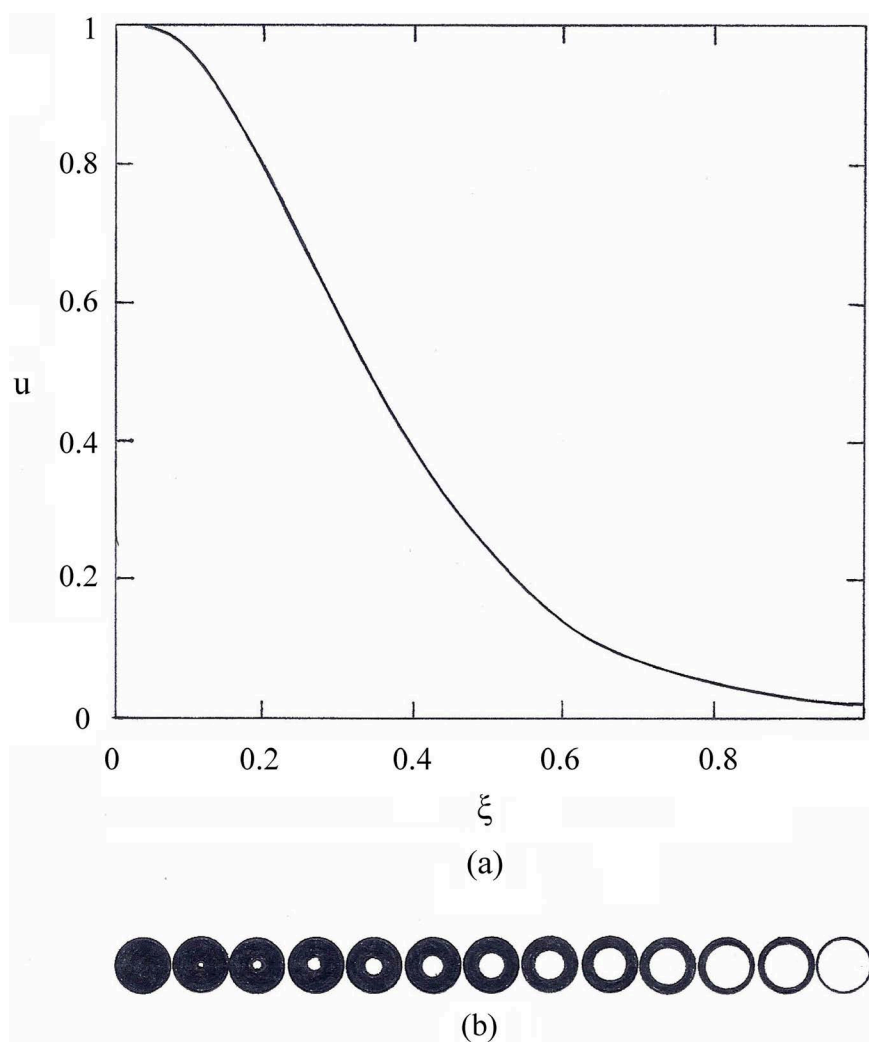


Figure 6. Structural—phase evolution of the sorbent particles and concentration of gas impurity. (a) Dependence of gas concentration on ξ at $\tau = 10$ according to the second iteration; (b) The phase structure of the sorbent particles: light areas—Me, dark areas—MeY; the initial data: $A = 5$, $B = 10^{-1}$.

under the condition that the characteristics of the system Me-Y as well as the technical data of the equipment and the process parameters retain their values with time, curve $u(\tau, 1)$ can serve a reliable source of information about the concentration of Y in the products of purification. It is important, therefore, to find an analytical expression for this function.

The total amount of gas that passed through the tube at time τ is $N = (\pi R^2 \varepsilon^2 p L / k_B T) \tau$ and the total amount of the impurity Y is $N_Y = c_0 N$, where p is the gas pressure, k_B is the Boltzmann constant and T is absolute temperature. The amount of the captured impurity Y at time τ is $\Delta N_Y(\tau) = N_A \Delta m_{Me}(\tau) / M_{Me}$, where $\Delta m_{Me}(\tau)$ is the mass of the spent reactant Me and N_A is the Avogadro constant. Then the impurity concentration in the outlet gas is $c = (N_Y - \Delta N_Y) / N$ or

$$u(\tau, 1) = 1 - (N_A / c_0 N M_{Me}) \Delta m_{Me}(\tau), \quad (11)$$

where $\Delta m_{Me}(\tau)$ is to be calculated. In order to do so we can use Equation (3) in the form

$$r/r_0 = 1 - B \int_0^\tau u(\tau', \xi) d\tau'.$$

Then the share of the reactant spent at time τ and coordinate ξ is

$$1 - \left[1 - B \int_0^\tau u(\tau', \xi) d\tau' \right]^3,$$

while the total share of the reactant that has entered into reaction with impurity Y at time τ (see **Figure 6**) is

$$\int_0^1 \left\{ 1 - \left[1 - B \int_0^\tau u(\tau', \xi) d\tau' \right]^3 \right\} d\xi.$$

The graphical form of this function ($D(\tau)$ henceforth) is given in **Figure 7**.

On the other hand, the same share $D(\tau)$ can be expressed as $\Delta m_{Me}(\tau) / m_{Me}$, where $m_{Me} = \pi R^2 L (1 - \varepsilon) \rho_{Me}$ is the initial mass of powder reactant. Therefore, we have

$$\Delta m_{Me}(\tau) = m_{Me} D(\tau). \quad (12)$$

Substituting (12) in (11) we finally arrive to dependence of the purity of outlet gas on τ

$$u(\tau, 1) = 1 - QD(\tau) / \tau, \quad (13)$$

where $Q = [R_B T (1 - \varepsilon) \rho_{Me} / c_0 \rho \varepsilon^2 M_{Me}]$ and R_B is the universal gas constant.

Formulas (12) and (13) are the contribution of the sorption theory of reactants, which we are developing, in the technology of gas purification. Formula (12) allows estimating in the efficiency of purification according to the cost/quality criterion. For this it is enough to assume in (12) $\tau = \tau_c$ as it is done below

$$\Delta m_{Me}(\tau_c) / m_{Me} = \int_0^1 \left\{ 1 - \left[1 - B \int_0^{\tau_c} u(\tau', \xi) d\tau' \right]^3 \right\} d\xi, \quad (14)$$

and to repeat these estimations, if required, for other purity levels u_c of the end product.

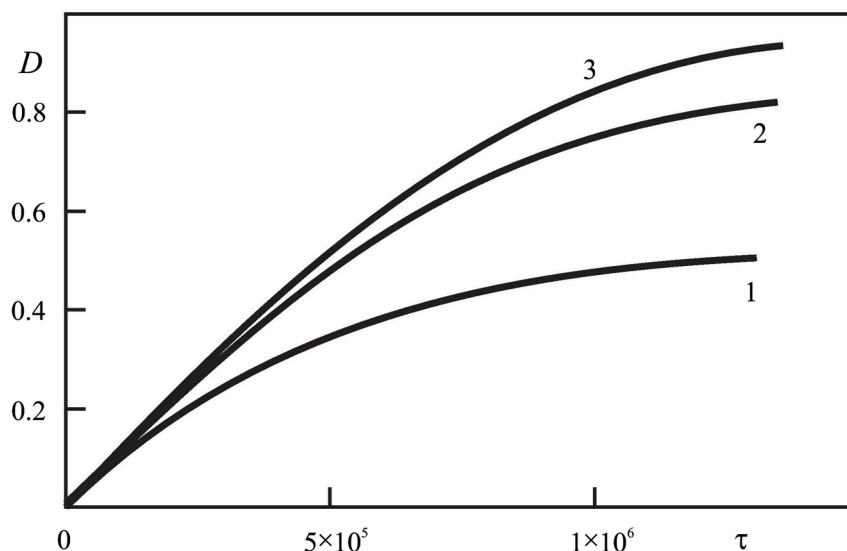


Figure 7. Change of share D of sorption waste with time. 1, 2 and 3—the first, the second and the third iterations accordingly; the initial data: $A = 10$, $B = 2 \times 10^{-6}$.

In its turn formula (13) can be considered as a theoretical grounding of the method of determination of the purity of the outlet gas according to purification time. At engineering level this method is realized by using a timer with a display graduated according to the results of the analysis of the gas samples taken in specified time intervals at the outlet of the flow tube. This supposes organization of a preliminary testing procedure with the involvement of precision analytical equipment according to the data of which experimental curve $c(t)$ vs t is built. The readings of the display are adjusted to this curve.

In industrial conditions it is more convenient to use not t but the amount of purified gas as the value to be measured. The impurity concentration at the entrance from the flow tube is determined not by time itself but by the depending on time gas quantity N , which passed through the tube. The purity of the gas product is the lower the less active sorbent remains in the tube; and there is the less active gas sorbent the larger amount of gas passed through it. That is, it is clear from the physical considerations that the value N can be converted into the values of concentration of outlet gas.

If in expression for N we pass over from τ to real time t we get $N = z(pvt)$, where z is a constant and (pvt) is a variable, which with the accuracy of a constant factor is equal to the amount of passing through the tube gas. The advantage of the complex parameter (pvt) over t as a measures value is obvious. Any variation of values p and/or v , which is possible and sometimes even desirable in the operation of gas purifiers, will distort the values of concentration c shown by the purity indicator, in which the argument to be measured is t . So, for tracking the purity of the gas product it is worthwhile to create a device on the basis of the tool, which is targeted at measuring gas flows, *i.e.* on the basis of a flow meter.

In this case in much the same way as an experimental curve $c(t)$ vs t a curve $[N(t)]$ vs $N(t)$ is built up and then with the help of the obtained graphical data a program for converting the readings of the flow meter into concentration units of the purified gas is created. A flow meter turns into an indicator of gas purity (patent pending).

The reliability of such an indicator will be very high if in its graduating the results of several tests performed with the help of Atmospheric Pressure Chemical Ionization Mass Spectrometer (APCI-MS) are used. The existing gas analyzers for gas stream monitoring are capable of detecting only a very limited set of gas species. For this reason the indicator, which we are describing here, should be tuned for determining the concentration of the main, *i.e.* the target gas and in this case the APCI-MS is indispensable.

So, the analysis of the sorption phenomena in powder reactants shows the possibility of solving the problem of economical security of the process systems, which use in their technologies gases fed from gas purifiers. The authors realize the importance of this problem for gas industry and are planning to describe in their next publication how the idea of the indicator of purity of the gas product is modified depending on the conditions of the sorption process. What is understood here is the substitution in gas purifiers the reactants following the linear sorption law with the reactants following parabolic law as well as the transfer from the sorption processes in motionless powders to the processes with the participation of tribological effects.

5. Conclusions

- 1) A mathematical model is developed for the process impurity gas capturing by powder reactants in flow tubes targeted for finishing gas purification.
- 2) The solutions of the model show the existence of the single valued functional connection $c = c(t)$ between the duration of the purification process and the purity of the exiting the tube gas.
- 3) The mentioned function provides the theoretical basis for the attempts of creating indirect methods of measuring purity of the gas product.
- 4) Any attempt of this kind by necessity includes certain practical actions aimed at building experimental curves $c(t)$ vs t or $[N(t)]$ vs $N(t)$ on the results of which an indicator of purity of the gas product is designed.
- 5) The simplest variant of this kind of an indicator is a gas flow meter calibrated for the units of gas concentration.

On the whole, it can be expected that the appearance of gas purifiers with metallic reactants and purity indicators of the outlet gas will considerably improve the efficiency of gas purification and the level of economical security for the customers of high purity gases.

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References

- [1] Zeller, R. and Vroman, C. (2006) Porous Sintered Composite Materials. US Patent No. 7112237.
- [2] Kerry, F.G. (2007) Industrial Gas Handbook: Gas Separation and Purification. CRC Press, Taylor & Francis Group, New York.
- [3] Rouquerol, J., Rouquerol, F., Llewellyn, P., Maurin, G. and Sing, K.S.W. (2013) Adsorption by Powders and Porous Solids: Principle, Methodology and Applications. 2nd Edition, Academic Press, New York.
- [4] Chuntonov, K.A. and Yatsenko, S.P. (2013) Getter Films for Small Vacuum Chambers. *Recent Patents on Materials Science*, **6**, 29-39. <https://doi.org/10.2174/1874464811306010029>
- [5] Chuntonov, K., Setina, J. and Douglass, G. (2015) The Newest Getter Technologies: Materials, Processes, Equipment. *Journal of Material Science and Chemical Engineering*, **3**, 57-67. <https://doi.org/10.4236/msce.2015.39008>
- [6] Chuntonov, K. and Setina, J. (2016) Reactive Getters for MEMS Applications. *Vacuum*, **123**, 42-48. <https://doi.org/10.1016/j.vacuum.2015.10.012>
- [7] Chuntonov, K. and Setina, J. (2017) Activationless Gas Purifiers with High Sorption Capacity. US Patent No. 9586173.
- [8] Adamson, A.W. (1982) Physical Chemistry of Surfaces. 4th Edition, John Wiley & Sons, New York.
- [9] Masel, R.L. (1996) Principles of Adsorption and Reaction on Solid Surfaces. John Wiley & Sons, New York.
- [10] Toth, J. (2002) Adsorption: Theory, Modeling, and Analysis (Surfactant Science Series, Volume 107), Marcel Dekker, New York.
- [11] Yang, R.T. (2003) Adsorbents: Fundamentals and Applications. John Wiley & Sons, Hoboken. <https://doi.org/10.1002/047144409X>
- [12] Chuntonov, K., Atlas, A., Setina, J. and Douglass, G. (2016) Getters: From Classification to Materials Design. *Journal of Materials Science and Chemical Engineering*, **4**, 23-34. <https://doi.org/10.4236/msce.2016.43004>

Appendix I

The current radius $r(t, x)$ of the powder particle is the distance from the center of this particle to the reactive boundary MeY/Me. Let us show how to calculate the value $r(t, x)$ in the case when the particle follows the linear law. In the case of reactant Me with plane surface S (**Figure 8**) located in gas medium with impurity Y, during time Δt a layer of product MeY with thickness Δh_{MeY} is formed (**Figure 8(b)**).

The consumption of the metal Me in an act like this one is measured by a layer of thickness Δh_{Me} (**Figure 8(a)**). Linearity of the sorption law becomes apparent here in the fact that during each interval of time Δt the boundary MeY/Me keeping its size and shape deepens into the volume of Me by one and the same value Δh_{Me} .

If the reactant has a shape of a ball particle of radius r (**Figure 8(c)**) then during time Δt the interphase boundary approximates the center of the particle by the value Δr . As far as at the molecular level the mass transfer processes are determined by the chemical nature of the pair Me-Y, it is natural to assume that the movement rate of the boundary MeY/Me along the normal to the surface of the solid will be the same for bodies of any geometrical shape and will be equal to dh_{Me}/dt . For this reason we postulate that $dr(t, x)/dt = dh_{\text{Me}}/dt$.

Basing on the above said and taking into account the data of Figure 8 we get that

$$dr(t, x)/dt = -k_0 M_{\text{Me}} c(t, x) / \rho_{\text{Me}} M_{\text{Y}} c_0, \quad (\text{I},1)$$

where ρ_{Me} is the density of substance Me, M_{Me} and M_{Y} are molar masses of Me and Y respectively. From here we find

$$r(t, x) = r_0 - (k_0 M_{\text{Me}} / \rho_{\text{Me}} M_{\text{Y}} c_0) \int_0^t c(t', x) dt', \quad (\text{I},2)$$

taking into consideration that $r(t=0) = r_0$.

One more remark. The thickness of the layer Δh_{MeY} in **Figure 8(b)** is set too high on purpose to make things more vivid. In reality $\Delta h_{\text{MeY}} \approx \Delta h_{\text{Me}}$: although the formation of the layer MeY takes place due to the arrival of atoms Y this

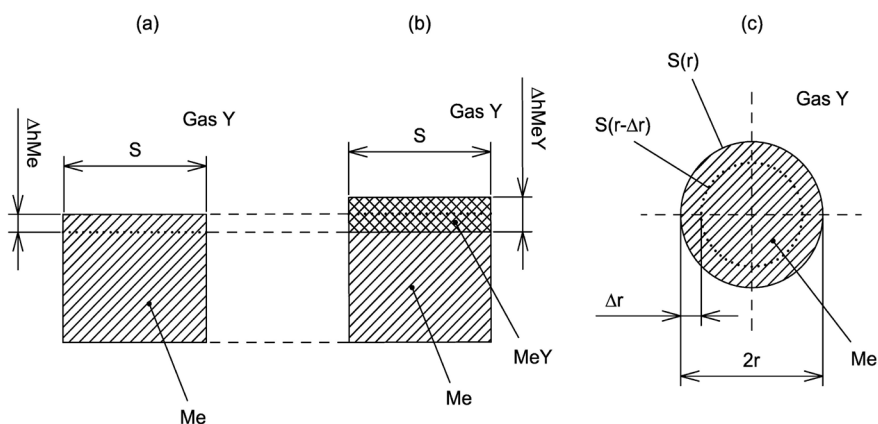


Figure 8. Linear sorption law. (a) and (b) flat material surface; (c) spherical body.

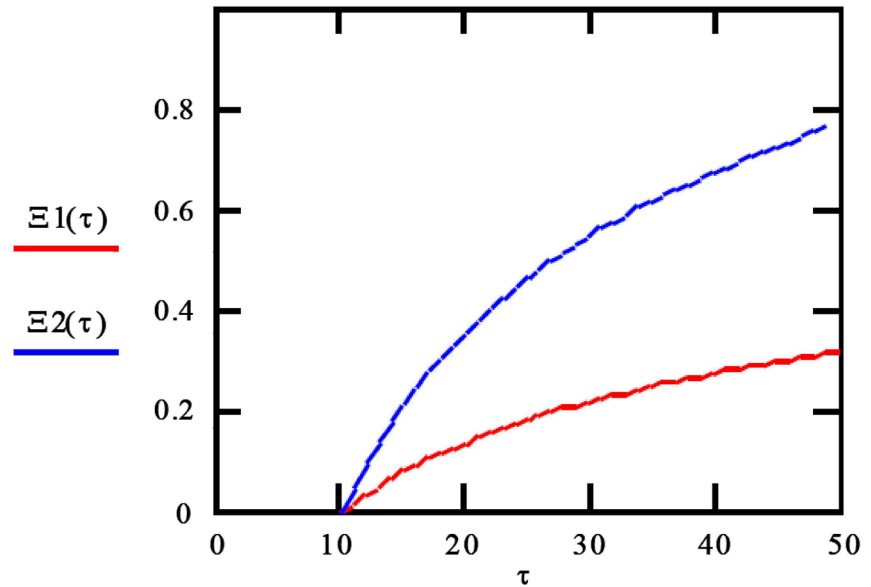


Figure 9. Movement of front Ξ as a function of time. Ξ_1 —the first iteration, Ξ_2 —the second iteration; the initial data: $A = 5$, $B = 10^{-1}$.

process is accompanied with volume contraction of the product. The given fact together with the equality $\Delta h_{\text{Me}} = \Delta r$ is the basis for derivation of formula (I,1).

Appendix II

Equation (7) with boundary conditions (8) and (9) can be solved with the help of an iterative procedure, which in the essence is building up disturbances according to the small value of parameter B .

The first iteration gives

$$\frac{\partial u_1}{\partial \tau} + \frac{\partial u_1}{\partial \xi} = -Au_1$$

with the solution

$$u_1(\tau, \xi) = \begin{cases} \exp(-A\xi), & 0 \leq \xi \leq \tau \\ 0, & \xi \geq \tau. \end{cases} \quad (\text{II},1)$$

The second iteration

$$\frac{\partial u_2}{\partial \tau} + \frac{\partial u_2}{\partial \xi} = -Au_2 F_1(\tau, \xi),$$

results in the solution

$$u_2(\tau, \xi) = 0, \quad \xi > \tau$$

$$u_2(\tau, \xi) = \exp(-A\xi) \exp \left\{ 2B(\tau - \xi) [1 - \exp(-A\xi)] - \frac{B^2}{2} (\tau - \xi)^2 [1 - \exp(-2A\xi)] \right\}, \quad 0 \leq \xi \leq \tau \leq \frac{1}{B}$$

$$u_2(\tau, \xi) = \exp \left\{ -A[\xi - \Xi(\tau)] + 2B(\tau - \xi) \left[\exp(-A\Xi(\tau)) - \exp(-A\xi) \right] - \frac{B^2}{2}(\tau - \xi)^2 \left[\exp(-2A\Xi(\tau)) - \exp(-2A\xi) \right] \right\}, \frac{1}{B} \leq \tau \leq T_1(\xi) \quad (\text{II}, 2)$$

$$u_2(\tau, \xi) = 1, \tau > T_1(\xi).$$

Here $T_1(\xi)$ is the moment of time starting from which all particles of the section with the coordinate (ξ) completely lose reactivity and $\Xi(\tau)$ describes the time law of movement of such a layer, which divides the tube with the powder into two areas, an area of the waste and an area with the still active sorbent. **Figure 9** shows the movement of front $\Xi(\tau)$ on the example of two iterations, the first and the second ones.

The third iteration is so bulky that it is presented only in a graphical form.

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