# Analytical Evaluation of the Amount of Extracted Quantities of Sorbate and Sorbent Dynamic Capacity under Sorption Purification of Flow Solutions

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**Abstract**

A calculation method for the evaluation of the amount of adsorbate (N) and the dynamic capacity of the sorbent (Q) under conditions of purification from pollutants of flow solutions is proposed. The considered approaches are valid when using sorbents and removing sorbates of any nature from flow media, regardless of the linear flow rate and, respectively, volume flow rates of the eluate under laminar flow conditions.

The classical kinetic curve of sorption in its most general form includes three time intervals from the beginning of the process:

– the time interval in which the adsorbate is absent in the solution flowing from the adsorber;

– the time interval in which the sorption coefficient $\rho_{\tau i}$ equal to $(C_0-C_{\tau i})/C_0$, where $C_0$ and $C_{\tau i}$, respectively, are the pollutant concentrations in the initial solution, and by the time $\tau_i$ is a function of $\tau_i$, and the value $\rho_{\tau i}$ decreases systematically with increasing $\tau_i$ from the beginning of the process. Accordingly, the concentration of pollutants in solutions leaving the adsorber increases.

– time interval with $\rho_{\tau i}$ equal to zero, when the dynamic capacity of the sorbent at a given linear flow rate through the adsorber is completely saturated.

The approach proposed in the article is possible to use regardless of the type and complexity of the function $\rho_{\tau i} = f(\tau_i)$ at a constant linear flow rate and other process conditions.

The method allows:

– calculating the values of N and Q at any time from the beginning of the sorption in the absence of an experimental estimate of the value $\rho_{\tau i}$;

– evaluating the calculated time of continuous operation of the sorbent before its replacement or regeneration.

It is based on the use of β-spline curves, which in turn is based on the properties of geometric continuity, used in solving problems of geometric modeling by means of cubic polynomials.

**Keywords**

Adsorption capacity, pollutants, laminar flow of solutions, β-spline curves.

The adsorption capacity of natural clay minerals [1–20] and bioproducts [21] is systematically investigated for the purpose of using them as sorbents for purification of drinking, process and waste waters and bioindication of biosphere purity [21].

However, in the overwhelming majority of cases, the sorption of pollutants is studied under static conditions that allows one to evaluate the equilibrium depth of the process, its thermodynamic characteristics, and the most probable type of adsorption isotherm [1–17].

Studies of the sorption of pollutants from flowing solutions are much less frequent [18–20], although the regularities of the process in both cases differ significantly.

For practice, the most important is the dynamic mode because it allows creating a continuous technological process.

Earlier [22], we considered the case of calculating the amount of pollutant material and the specific dynamic capacity of the sorbent, when the sorption process in the particular form was transferred by a kinetic curve expressing the dependence $\rho = f(\tau)$ shown in Fig. 1 (curve *1*).

The value of $\rho$ in terms of physical meaning is the sorption coefficient, which is equal to the ratio of the difference of concentrations of sorbate in the initial solution $C_0$ and at the given current moment $\tau_i$ ($C_{\tau,i}$) to its initial value. The value of $\tau_i$ is the time from the beginning of the sorption.

$$\rho_{\tau,i} = (C_0 - C_{\tau,i})/C_0 \qquad (1)$$

or

$$C_\tau = C_0(1 - \rho_{\tau,i})$$

Thus, in terms of physical meaning, $\rho_{\tau,i}$ is the fraction of pollutant extracted by the adsorbent from the solution at the time moment $\tau_i$ from the beginning of the process

The kinetic curve in the most general form in this examination includes three distinct regions.

A complete sorption removal of the sorbate takes place on the AC region of the kinetic curve. When the time $\tau_C$ is reached, its breakthrough begins. The *MN* line, parallel to the abscissa axis, characterizes the value of $\tau_r$, i.e. the time to which the concentration of pollutant in the solution leaving the adsorber does not exceed the regulatory requirements.

The principal feature of the kinetic curve 1 (Fig. 1) is that in the *CD* region the value of $\rho_{\tau,i}$ is a linear function of $\tau_i$. A similar picture is observed relatively often [18–20], but still has a private character. If the coordinates of the characteristic points $C$ ($\rho = 1$) and $D$ ($\rho = 0$) are taken into account, one can obtain the expression for (1) [22], and then calculate the amount of substance ($N$) of the absorbed pollutant and the dynamic capacity of the sorbent $Q$ by the time point $\tau_i$ in the *CD* region from the beginning of the process, including to the $\tau_r$.
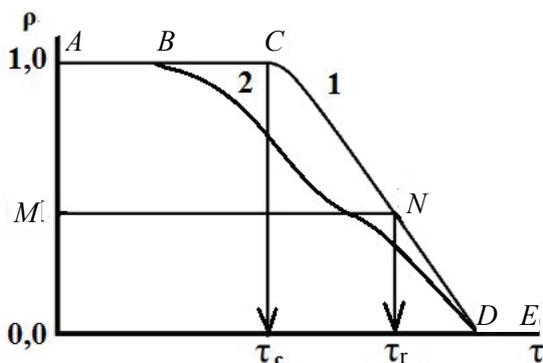


**Fig. 1. Schematic representation of kinetic curves of sorption with linear (*1*) and nonlinear (*2*) dependence $\rho = f(\tau)$ in the *CD* region**

There is no sorption in the DE region, since the dynamic capacity is saturated. Then the total value of $N$ and $Q$ by the time moment $\tau_D$ are found by summing the corresponding values of $N$ and $Q$ of the AC and CD regions, in particular:

$$N_\Sigma = N_{AC} + N_{CD}$$

$$Q_\Sigma = Q_{AC} + Q_{CD}$$

However, in the general case, the dependence $\rho = f(\tau)$ in the AC region can have a more complex form (curve *2*, Fig. 1). Its character is sometimes even more complicated. In this case, the proposed approach is inappropriate.

We consider a more general case of the form of the dependence

$$\rho_{\tau,i} = f(\tau_i),$$

for example, represented by curve 2, Fig. 1. Further complication of the form of the kinetic curve of sorption will not result in substantial changes.

It is of fundamental importance to estimate the value of $\rho_r$, which can be found by calculation using Eq. (1). It should be noted here that $\rho_r$ is not a constant for the pollutant in question, but depends on its initial concentration $C_0$. In other words, it should be written

$$\rho_r = (C_0 - MPC_w)/C_0,$$

where $MPC_w$ is maximum permissible concentration of pollutant in water. Then we have

$$\rho_r = 1 - MPC_w / C_0.$$

Consequently, the higher $C_0$, the greater is $\rho_r$.

The amount of substance (N) sorbed by the adsorbent for a certain time interval from $\tau_1$ to $\tau_2$ can be obtained from the dependence:

$$N = C_0 V \int_{\tau_1}^{\tau_2} d\left(1 - \rho(\tau)\right), \qquad (2)$$

where $C_0$ is determined earlier in the appropriate units, for example, mol/L, and $V$ is the volume of the solution that passed through the adsorber in the considered time interval. Taking into account the properties of the integrals in integrating equation (2), we have:

$$N = -C_0 V(\rho_{\tau,2} - \rho_{\tau,1}). \qquad (3)$$

Since there is always the dependence

$$\rho_{\tau,2} \leq \rho_{\tau,1},$$

then the value $N$ can only be positive and/or equal to zero, which corresponds to its physical meaning. In the latter case, there is no adsorption. The following discussion is related to the kinetic curve of sorption shown in Fig. 2.

Let the points $A$, $B$, $C$, $D$, $E$, $F$ and $M$ correspond to the times from the beginning of the sorption $\tau_A$, $\tau_B$, $\tau_C$, $\tau_D$, $\tau_E$, $\tau_F$, $\tau_M$, shown on the abscissa, and sorption coefficients $\rho_A$, $\rho_B$, $\rho_C$, $\rho_D$, $\rho_E$, $\rho_F$, $\rho_M$. In this case, $\rho_A = \rho_B = 1$ is complete sorption, no breakthrough. $\rho_M = 0$ – sorption discontinued due to the dynamic saturation capacity. Consequently, inequality $1 < \rho_B$, $\rho_C$, $\rho_D$, $\rho_E$, $\rho_F < 0$ is observed, and in the direction from $\rho_B$ to $\rho_F$ the value of $\rho_i$ decreases. The amount of absorbed sorbate by the sorbent material in the time interval from the start of the sorption to $\tau_B$ is equal to

$$N_{AB} = C_0 V_{AB},$$

in time intervals $\tau_B - \tau_C$, $\tau_C - \tau_D$, $\tau_D - \tau_E$, $\tau_E - \tau_F$, $\tau_F - \tau_M$, according to (2), it is respectively equal to:

$$N_{BC} = -C_0 V_{BC}(\rho_C - \rho_B);$$
$$N_{CD} = -C_0 V_{CD}(\rho_D - \rho_C);$$
$$N_{DE} = -C_0 V_{DE}(\rho_E - \rho_D);$$
$$N_{EF} = -C_0 V_{EF}(\rho_F - \rho_E);$$
$$N_{FM} = -C_0 V_{FM}(\rho_M - \rho_F);$$

Suppose that for a constant linear flow rate, the time intervals $\tau_i - \tau_{i+1}$ are equal, then $V_i$ is a constant value.

If this condition is satisfied $N_\Sigma = N_{AM}$ is equal to

$N_\Sigma = C_0 V - C_0 V(\rho_C - \rho_B) - C_0 V(\rho_D - \rho_C) - C_0 V(\rho_E - \rho_D) - C_0 V(\rho_F - \rho_E) - C_0 V(\rho_M - \rho_F).$

$$N_\Sigma = C_0 V(1 + \rho_B - \rho_M). \tag{4}$$

Since $\rho_M = 0$, then finally we have

$$N_\Sigma = C_0 V(1 + \rho_B).$$

Total specific dynamic capacity $Q_\Sigma$ is

$$Q_\Sigma = N_\Sigma / m.$$

and

$$Q_\Sigma = \frac{C_0 V}{m}(1 + \rho_B).$$

If the value $V$ in the time interval under consideration is not constant, then to find $Q_\Sigma$, it is necessary to sum $Q_i$ of all time intervals, i.e.:

$$Q_\Sigma = \sum_{i=1}^{\Pi} Q_i. \tag{5}$$

Since for each pollutant there is a certain normative-permissible concentration in the solution flowing from the adsorber, which is convenient to express as $\rho_r$ (Fig. 1), it is expedient to calculate the specific dynamic capacitance of the sorbent up to the value of $\rho_r$.

Then equation (4) is transformed into (6) ($V = \text{const}$):

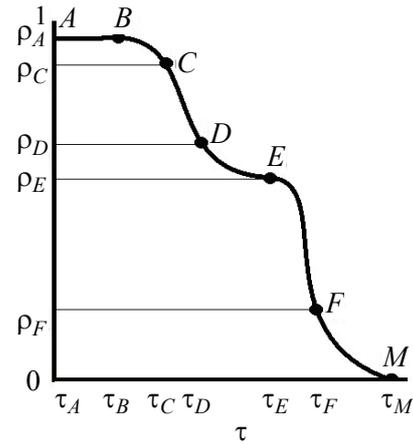$$N_\Sigma = C_0 V(1 + \rho_B - \rho_r) \tag{6}$$



**Fig. 2. Schematic view of the kinetic curve of sorption**

In a number of process problems, it is necessary to know the specific dynamic capacity of the sorbent at a certain given time point, for example $\tau_r$. Knowledge of such quantities determines the duration of continuous operation of adsorbers before the replacement or regeneration of the sorbent, the cyclic control of the eluate flowing from it. In this case, it is necessary to know either the form of the kinematic curve function for the entire sorption period, which is practically impossible in the case of complex dependences of $\rho_i = f(\tau_i)$, or know it at different time intervals. If it is known to which interval of time $\tau_i - \tau_{i+1}$ refers the time of interest of the adsorber work and the form of the function $\rho_i = f(\tau_i)$ in this region, then we can estimate $\rho_i$ to a given moment from the beginning of the sorption.

Let us consider a similar approach, when $\rho_i$ is known for more than four values of $\tau_i$. The technique is based on the use of β-spline curves, which in turn is based on the properties of geometric continuity and smoothness of the used solutions of geometric modeling tasks by cubic polynomials [23].

To this end, a curve passing through all points (the interpolation task) is constructed from a given array of experimental points, in our case with the coordinates $\rho_i$ and $\tau_i$ (Table 1).

The technique uses β-splines, while the use of composite β-spline curves is based on the property of geometric continuity and smoothness. In the construction of a composite regular curve, an important role is played by the conjugation condition at the points of contact of segments of regular curves smoothed by it.

Typically, interpolation uses cubic or linear splines. As further calculations showed, the functional dependence on each time interval is close to linear: the coefficients for the second and third powers of the variable $\tau$ are relatively small. However, to ensure the smoothness of interpolants (continuity of the first and second derivatives), it is expedient to use a cubic spline.

Table 1

**The coordinates of the experimentally obtained points**

| No | $\tau$, min, from the beginning of sorption | $\rho$ |
|----|----|----|
| 1 | 0.0 | 1.00 |
| 2 | 10.0 | 1.00 |
| 3 | 20.0 | 1.00 |
| 4 | 32.5 | 0.92 |
| 5 | 40.0 | 0.78 |
| 6 | 45.0 | 0.63 |
| 7 | 50.0 | 0.43 |
| 8 | 55.0 | 0.10 |
| 9 | 58.0 | 0.00 |
| 10 | 60.0 | 0.00 |
| 11 | 65.0 | 0.00 |
| 12 | 70.0 | 0.00 |

In the first two time intervals, $\rho$ remains constant and equal to 1, i.e. adsorbate is extracted completely, at the last two intervals $\rho = 0$, since sorption ceases due to the saturation of the dynamic capacity of the sorbent. In the remaining intervals of time, it is necessary to interpolate the regions of dependence by a cubic spline, which is a twice continuously differentiable function in the time interval from 20 to 60 min of sorption.

At each elementary time interval (Table 1) $[\tau_i - \tau_{i+1}]$, where $i = 3, \ldots, 9$, the empirical functional dependence is interpolated by a cubic parabola

$$\tilde{\rho} = y_i = a_i(\tau - \tau_i)^3 + b_i(\tau - \tau_i)^2 + c_i(\tau - \tau_i) + d_i. \quad (7)$$

Parabola coefficients $a_i, b_i, c_i, d_i$ (7) at each time interval are determined from the following conditions:
1) interpolation, i.e. matches in nodes

$$y_i(\tau_i) = \rho_i, \; y_i(\tau_{i+1}) = \rho_{i+1}, \; i \text{ – from 3 to 9};$$

2) smooth joining

$$y_i'(\tau_i) = y_{i-1}'(\tau_i), \; i \text{ – from 4 to 9};$$

3) continuity of the second derivative at the interpolation nodes (experimental points)

$$y_i''(\tau_i) = y_{i-1}''(\tau_i), \; i \text{ – from 4 to 9};$$

4) boundary conditions determined by the nature of the experimental problem

$$y_1'(\tau_1) = 0, \; y_9'(\tau_{10}) = 0.$$

Substituting the data of the second and third columns of Table 1 in the analytical expressions of the four conditions listed above, we make up four equations for seven time intervals from $[\tau_3; \tau_4]$ to $[\tau_9; \tau_{10}]$, and arrive at a system of 28 linear algebraic equations with 28 unknowns, which we solve by the Gauss method, using the Gauss-Jordan Elimination Calculator [25].

To write the equations corresponding to the intervals of the interpolation curve, the calculated values of the constants $a_i$, $b_i$, $c_i$ and $d_i$ are substituted into equation (7).

The obtained equations for the time intervals of the interpolation curve for the task solved in this case are collected in Table 2.

The form of the kinetic curve of sorption ($\beta$-spline) corresponding to its interpolation equations generalized in Table 2 is shown in Fig. 3.

Table 2

**The time intervals from the beginning of the sorption and the corresponding equations of the interpolation curve**

| No | $\tau$, min | $\rho$, unit fraction | Time interval $[\tau_i; \tau_{i+1}]$ | Equation of interpolation curve |
|----|----|----|----|----|
| 1 | 0 | 1.00 | [0; 10] | $\rho = 1$ |
| 2 | 10 | 1.00 | [10; 20] | $\rho = 1$ |
| 3 | 20 | 1.00 | [20; 32.5] | $\rho = -0.0005(\tau - 20)^2 + 1$ |
| 4 | 32.5 | 0.92 | [32.5; 40] | $\rho = 0.00053(\tau - 32.5)^2 - 0.0129(\tau - 32.5) + 0.92$ |
| 5 | 40 | 0.78 | [40; 45] | $\rho = 0.0001(\tau - 40)^3 - 0.0013(\tau - 40)^2 - 0.00263(\tau - 40) + 0.78$ |
| 6 | 45 | 0.63 | [45; 50] | $\rho = -0.0004(\tau - 45)^3 + 0.0003(\tau - 45)^2 - 0.0311(\tau - 45) + 0.63$ |
| 7 | 50 | 0.43 | [50; 55] | $\rho = 0.0009(\tau - 50)^3 - 0.0058(\tau - 50)^2 - 0.0591(\tau - 50) + 0.43$ |
| 8 | 55 | 0.1 | [55; 58] | $\rho = -0.0003(\tau - 55)^3 + 0.0077(\tau - 55)^2 - 0.0502(\tau - 55) + 0.1$ |
| 9 | 58 | 0.01 | [58; 60] | $\rho = -0.0006(\tau - 58)^3 + 0.0049(\tau - 58)^2 - 0.0124(\tau - 58) + 0.01$ |
| 10 | 60 | 0.00 | [60; 65] | $\rho = 0$ |
| 11 | 65 | 0.00 | [65; 70] | $\rho = 0$ |
| 12 | 70 | 0.00 | | |

Earlier it was shown that the amount of the substance ($N$) of the sorbed pollutant for a certain time interval from $\tau_1$ to $\tau_2$ can be obtained from the dependence (2), which is easily transformed upon integration into (3). We consider an example of calculation of value $\rho$ to the specified moment of time. Let it be necessary to find the specific dynamic sorption capacity used for 53 minutes of operation of the adsorber with sorbent mass m in accordance with the data of Table 2. The specified time corresponds to a time interval with interpolation equation

$$\rho_{spec} = 0.0009(\tau - 50)^3 - 0.0058(\tau - 50)^2 - \\ - 0.0591(\tau - 50) + 0.43.$$

We substituting in it the specified time

$$\rho_{spec} = 0.0009(53 - 50)^3 - 0.0058(53 - 50)^2 - \\ - 0.0591(53 - 50) + 0.43;$$

$$\rho_{spec} = 0.225.$$

The specified time in accordance with the curve in Fig. 3 is in the time interval $\tau_F - \tau_M$. If the value $V$ in all time intervals is constant, $Q_{initial}$ is calculated from a dependence of the type (4), where $\rho_{spec}$ is used instead of $\rho_M$:

$$Q_{spec} = \frac{C_0 V}{m}(1 + \rho_B - \rho_{spec}).$$

In this particular case (Fig. 2) we have

$$Q_{spec} = \frac{C_0 V}{m}(2 - \rho_{spec}),$$

$$Q_{spec} = 1.775 C_0 V / m.$$

If V is not a constant, equation (5) is used.

Thus, using the β-spline method, we can calculate $\rho_i$ and $Q_i$ at any time from the beginning of the sorption.

The proposed approaches are not related to the nature of the sorbate and sorbent, its mass, the number of pollutants simultaneously sorbed from the solution, and its linear rate.

The total dynamic capacity of the sorbent of several sorbed pollutants at a specified time is found by summing $Q_i$ of each i-sorbate.

At the same time, we note the following. When solving real technological problems, it is necessary to distinguish between the influence of the linear flow rate on at least two characteristic parameters of the sorbent in relation to the pollutants under consideration. First, the sorption capacity of ecotoxicants corresponding to their detection, even in trace amounts, in solutions emerging from the adsorber, which is determined by the sensitivity of the method of analysis used.

Secondly, it is necessary to estimate the specific dynamic capacity to the level of the breakthrough corresponding to the concentration of pollutants permitted by regulatory documents. The second value, as shown above, depends not only on the physical, chemical and physical-chemical characteristics of ecotoxicants, but also on their toxicological activity.

In addition, as noted above, the proposed method is valid both for sorption purification from one and at the same time several ecotoxicants, the concentrations of which in the initial solution and solution leaving the adsorber can be determined by independent methods or in a single analysis, for example, by X-ray fluorescence spectroscopy. It is applicable to the sorption removal of pollutants of any nature, using both natural and synthetic sorbents for the laminar flow of solutions passing through the adsorber.

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