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FORMULATION OF A MATHEMATICAL MODEL FOR THE COAGULATION-ADSORPTION PROCESS IN AN UP-FLOW DIRECT FILTRATION, GRANULAR ACTIVATED CARBON BED

The mathematical model of coagulation-adsorption process performed in granular activated carbon has been presented. It has been shown that suspended alum-sludge has the ability to adsorb dissolved compounds. In consequence the real run time of GAC is extended. Application of a model has been shown for prepared water with colour intensity of 30 and 60 g Pt/m³ and phenol concentration of 4 g/m³. Activated carbon produced by Chemviron Carbon Co., type HKW1, with a sufficient mechanical durability has been applied.

NOTATION

- B – colour of water [g Pt/m³],
 b – Langmuir isotherm constant,
 C – concentration of suspended solids [g/m³],
 C_0 – adsorbate concentration in the inflow to the column [g/m³],
 C_{os}^p – initial concentration of the sludge in the bed [g_{os}/m³],
 C_{os}^k – final concentration of sludge in the bed, which is equal to the maximum capacity of the bed for suspended solids q_{max} [g_{os}/m³],
 C_{zaw} – concentration of suspended solids in raw water [g/m³],
 D – coefficient of molecular adsorbate diffusion [m²/s],
 D_k – coagulant dose [g/m³],
 d_a – diameter of adsorbate particle [m],
 d_w – diameter of the grains in the bed [m],
 F – area of section of diffusion stream [m²],
 H – height of the filter bed [m],
 K – Boltzman constant 1.38×10^{-23} [J/K],
 k – coefficient depending on the type and quality of coagulant; for pure alum this figure is 0.55,
 L_0 – initial concentration of suspended alum-sludge [g/m³],

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- M – adsorption mass [g],
 N – amount of suspended compounds in coagulant [g/m^3],
 Q – filtration flow rate [m^3/s],
 R – direction of diffusion [m],
 S_0 – surface area of sludge [m^2/g],
 T – temperature [K],
 t – time of process [h],
 u_p – initial water content [%],
 u_k – final water content [%],
 V_p – linear filtration velocity [m/s],
 x – quantity adsorbed on one unit of adsorbent [g/kg],
 x_m – quantity adsorbed in monolayer on one unit of adsorbent [g/kg],
 x_{os} – adsorption capacity of sludge [g/g_{os}],
 Δr – time-dependent change of diameter of grains in the bed as a consequence of the retention of suspended solids in the bed [m],
 ΔV – differential unit of volume [m^3],
 ε – porosity of bed,
 $\varepsilon(t)$ – average porosity of the bed changing in time,
 ψ – spherical surface of the grains in the bed,
 η – coefficient of the dynamic viscosity of the medium where diffusion takes place [$\text{N}\cdot\text{m}^2/\text{s}$],
 ρ_w – density of hydrated activated carbon [kg/m^3].

1. INTRODUCTION

The main steps involved in a present-day water treatment plant are coagulation, sedimentation, filtration, adsorption and disinfection. These processes are usually run in series with each step further improving the quality of the water.

The inverse filter bed is a departure from the normal water treatment methods, by combining the coagulation, sedimentation, filtration and adsorption processes to obtain one viable unit. The reasons behind this concept are that by combining the steps, the cost of manufacture and day to day running costs are greatly reduced. The inverse filter bed utilises granular activated carbon (GAC) as its filter media. It is also a departure from traditional operations, where due to gravity water passes through the bed, to a process whereby water is forced to flow upwards through the bed. When influent is introduced to the bottom of the bed, the up-flow allows the bed to expand, increasing the inter-granular spacing between the activated carbon particles. This allows more floc particles to be held in the larger spaces between the grains of GAC. The increased levels of flocs augment the adsorption capacity of the bed, as the alum sludge also adsorbs dissolved organic compounds [1].

2. AIM AND SCOPE OF THE STUDY

The objectives of this study may be itemized as follows:

1. Formulation of a mathematical model for the coagulation-adsorption process in a bed of activated carbon.

2. Determination and calculation of the parameters involved in the coagulation-adsorption process taking place in an up-flow direct filtration bed.
3. Verification of the model through application of research data.

3. MODEL OF THE COAGULATION-ADSORPTION PROCESS IN A BED OF ACTIVATED CARBON

The formulation of the mathematical model allows the calculation of the run time per cycle of filtration and the number of filtration runs until the adsorption bed capacity is exhausted in the coagulation-adsorption process.

The limiting factor in the process of contact coagulation in the filter bed is the level of deposits of suspended solids, which together with suspensions from the coagulation process should not exceed 150 g/m^3 [2]. In terms of this process it has a better application for water which has intense colour and a low turbidity content. The filtration cycles are considerably shorter in comparison to the cycles of conventional filters. However, the cycles should not be shorter than 6 hours. The recommended speed of filtration should not exceed 6 m/h . The height of the bed is higher compared to traditional filter beds, in general $2\text{--}2.5 \text{ m}$. Diameters d_{10} of the grains of sand should range from 0.55 to 0.65 mm , and d_{80}/d_{10} should not be greater than 2.5 mm .

The model that describes the process of contact coagulation in the bed of activated carbon, in terms of the geometry of the bed and the adsorption abilities of the alum-coagulation sludge, used Flicks' first law of diffusion in a steady state [3]–[5]. The process of filtration is described using the equation of mass balance in a unit of volume of the bed [6] and the filtration constant value [7]. The equation of mass balance in a unit of volume of the filter layer is described as:

$$\frac{\partial q}{\partial t} \Delta V + \varepsilon(t) \frac{\partial C}{\partial t} \Delta V = QC|_H - QC|_{H+\Delta H}, \quad (1)$$

where:

$\frac{\partial q}{\partial t}$ – the speed of change in the quantity of solid particles deposited inside the filter [$\text{g/m}^3\text{s}$],

$\frac{\partial C}{\partial t}$ – change in the average concentration of solid particles in the porous space, in time [$\text{g/m}^3\text{s}$].

When defining $\Delta V = F \times \Delta H$ (F is a transverse section of the bed), taking into account that $Q = F \times V_p$, (V_p is the linear filtration velocity) and accepting $\Delta H \rightarrow 0$, one obtains:

$$-V_p \frac{\partial C}{\partial H} = \frac{\partial q}{\partial t} + \varepsilon(t) \frac{\partial C}{\partial t}. \quad (2)$$

Since the volume of solution in the bed is relatively small, the above equation can be simplified to:

$$-V_p \frac{\partial C}{\partial H} = \frac{\partial q}{\partial t} \quad (3)$$

The above form of equation is well documented in literature [6]. In order to solve this equation, the function describing the changes in the concentration of suspended solids, in this depth of bed $\frac{\partial C}{\partial H}$ is required. The limiting factors that affect the changing quantities of suspended solids $\frac{\partial q}{\partial t}$ retained in the filter are also required. These functions are well documented in literature [7]. Function

$$-\frac{\partial C}{\partial H} = \lambda C \quad (4)$$

is based on the changes in the concentration of suspended solids during their flow through the elementary layer of the bed, and is proportional to the initial concentration. In equation (4), λ is the filtration coefficient that varies over time and depends on the geometry of the layer, the hydrodynamic conditions in the layer, the physico-chemical properties of water and the value of the variable q , which relates to the quantities of suspended solids deposited inside the bed. The value λ depends on the force of adhesion, which retains suspended solids in the bed, and hydrodynamic forces, which counteract this retention.

The formula for the filtration constant used to analyse this process is expressed in the following form [7]:

$$\lambda = \frac{6(1-\varepsilon) \Delta r}{\varepsilon \psi d_w \Delta H} \quad (5)$$

Equation (3) can be modified with regard to the above equation and depending on equation (4) and expressed in the following form.

$$V_p \frac{6(1-\varepsilon) \Delta r}{\varepsilon \psi d_w \Delta H} C = \frac{dq}{dt} \quad (6)$$

The value Δr changes from 0 at the beginning of the filtration cycle to the value representing the radius of the intergranular capillary (r/z). The defined variables are integrated by the values ranging from $t = 0$ to $t = t_c$ (where t_c is the time of the filtration cycle), and by the values ranging from $q = 0$ to $q = q_{\max}$ (where q_{\max} is the maximum capacity of the bed for pollutant). After taking into consideration the formula for the radius of intergranular capillary we obtain [8]:

$$r_c = \frac{\varepsilon d_w}{6(1-\varepsilon)}. \quad (7)$$

Therefore:

$$t_c = \frac{z\psi H}{V_p} \ln \frac{q_{\max}}{c_0}. \quad (8)$$

Accepting the filtration mechanism, the equation can be analysed by certain analogies to the process of sludge thickening, in which flocs colliding with each other squeeze out water from their structures, and simultaneously to the hydraulics of the flow through the layer which causes compression of the sludge. Accepting that the initial concentration of suspended alum-sludge (L_0) increases in the bed to a similar level as that in a condensed sludge at the end of filtration process, the final concentration of alum-sludge is a measure of the capacity of the bed for suspended solids (q_{\max}).

Accepting the initial water content (u_p) and the final water content (u_k) of the sludge, the capacity of the bed for suspended solids can be expressed as:

$$q_{\max} = L_0 \frac{100 - u_k}{100 - u_p}. \quad (9)$$

The final form of the equation of the filtration cycle is described as:

$$t_c = \frac{z\psi H}{V_p} \ln \frac{100 - u_k}{100 - u_p}. \quad (10)$$

The values u_p and u_k , given in literature [9], [10], are based on the levels of 99.7% and 98.5 %, respectively.

Fick's equation is expressed as:

$$\frac{dm}{dt} = -DF \frac{dC}{dR}. \quad (11)$$

The above equation for the adsorption column of the height H and intergrain radius capillary r_c after accepting suitable parameters was transformed to allow the possible qualification of the effects of the process C_e/C_0 for the time of contact with the bed equal to H/V_p :

$$\frac{C_e}{C_0} = \exp \left(- \frac{H}{V_p} \frac{144(1-\varepsilon)^2 D}{\varepsilon d_w^2 [6(1-\varepsilon) + \varepsilon]} \right). \quad (12)$$

The process of adsorption in a granular activated carbon bed proceeds in several stages. At the beginning of cycle, dissolved particles of adsorbate diffuse directly to

the grains of activated carbon, which can be described by the Stokes–Einstein equation [11] defining the coefficient of molecular diffusion:

$$D = \frac{KT}{3\pi\eta d_a} \quad (13)$$

With an increase in the concentration of suspended solids in the bed, the molecular diffusion coefficient decreases due to an increase in viscosity of the medium. Therefore the efficiency of the adsorption on activated carbon decreases. Simultaneously, on the surface of the flocs the process of adsorption begins.

At the end of the filtration cycle, the access to active adsorption sites on the activated carbon decreases as a consequence of the compressed sludge, and as a result, adsorption takes place, first of all, on the active sites of the compressed sludge. After the time t_c , defined by equation (10), the filtration cycle is completed and the bed has to be washed with clean water.

As the hydrodynamic flow conditions do not change during this cycle, the contact time with the bed is stable. So the molecular diffusion coefficient changes as a consequence of decreasing viscosity, which results in a decrease of the adsorption effect.

The initial concentration of outflow from the bed equals:

$$C_e^p = C_0 \exp(-AD), \quad (14)$$

where:

$$A = \frac{H}{v_p} \frac{144(1-\varepsilon)^2}{\varepsilon d_w^2 [6(1-\varepsilon) + \varepsilon]}, \quad (15)$$

and $D = f(\eta)$ decreases to the value:

$$C_e^k = C_0 \exp(-AD_z), \quad (16)$$

where:

$$D_z = f(\eta_z).$$

Viscosity of alum-sludge (η_z) depends on the water content in the sludge (u), water viscosity (η) and the flocs surface area (s_0). This viscosity can be found from a modified Einstein equation [12]:

$$\eta_z = \eta [1 + 1.92 \cdot 10^{-3} S_0^{1.46} (100 - u)]. \quad (17)$$

Properties of the sludge structure, made during the process of coagulation, as well as the level of its development depend on the composition of the raw water, the dose of coagulant, the types of coagulant and flocculent, and on the conditions of the flocculation [12].

The average value of molecular diffusion coefficient for dissolved organic compounds, measured with TOC in water after the process of coagulation has a range $D = (5.8-6.0) \times 10^{-10} \text{ m}^2/\text{s}$ [13].

Decrease in the efficiency of adsorption by activated carbon is compensated to a certain degree by adsorption on the flocs generated.

During one filtration cycle (t_c), activated carbon is able to adsorb on one unit of weight:

$$x_1 = \frac{(2C'_0 - C_e^p - C_e^k)V_p t_c}{2H\rho_w(1-\varepsilon)}. \quad (18)$$

After introducing the definition of one time cycle t_c (equation (10)) and accepting the value of particle sphere $\psi = 0.85$ we arrive at

$$x_1 = \frac{(2C_0 - C_e^p - C_e^k)z\psi}{2\rho_w(1-\varepsilon)} \ln \frac{100 - u_k}{100 - u_p}. \quad (19)$$

The number of filtration cycles necessary to exhaust the adsorption capacity of carbon (N) is determined by equation:

$$N = \frac{2x_m b (C_e^p + C_e^k) \rho_w (1-\varepsilon)}{[2 + b(C_e^p + C_e^k)](2C_0 - C_e^p - C_e^k)z\psi \ln \frac{100 - u_k}{100 - u_p}}. \quad (20)$$

In reality, the concentration of adsorbate, which changes in time, decides the driving force of the adsorption process on activated carbon. At the beginning of the filtration cycle this concentration is:

$$C_0^p = C_0 - x_{os} C_{os}^p. \quad (21)$$

At the end of the filtration cycle the concentration of adsorbate amounts to:

$$C_0^k = C_0 - x_{os} C_{os}^k. \quad (22)$$

The concentration of suspended solids (C_{os}^p) in the inflow to the column can be calculated using [9]:

$$C_{os}^p = C_{zaw} + kD_k + 0.25B + N. \quad (23)$$

Alum dose as a function of water colour can be described by the empirical formula [2], [9]:

$$D_k = 7\sqrt{B}. \quad (24)$$

Taking into account that contact coagulation can be used for water with low turbidity ($C_{zaw} = 0$) and that $N = 0$ we arrive at:

$$C_{os}^p = 0.55D_k + 0.25B. \quad (25)$$

Combining equation (9) with (25) results in the equation that describes the maximum concentration of sludge in the bed:

$$C_{os}^k = (0.55D_k + 0.25B) \frac{100 - u_k}{100 - u_p}. \quad (26)$$

The concentration of adsorbate in the outflow from the column at the beginning of the filtration cycle, taking into consideration the adsorption ability of alum sludge, is equal to:

$$(C_e^p)' = C_0^p \exp(-AD). \quad (27)$$

And taking into consideration the water content in the sludge, the final concentration will be:

$$(C_e^k)' = C_0^k \exp(-AD_z). \quad (28)$$

Because the real adsorbate concentration does not change during the filtration cycle (equations (26) and (27)), the amount adsorbed on one unit of adsorbent $(x_1)'$ has to change:

$$(x_1)' = \frac{[C_0 - (C_e^p)' + C_0^k - (C_e^k)'] z \psi}{2\rho_w(1 - \varepsilon)} \ln \frac{100 - u_k}{100 - u_p}. \quad (29)$$

So the number of filtration cycles necessary for exhausting the carbon adsorption capacity $(N)'$ is described by the following equation:

$$(N)' = \frac{x'}{(x_1)'} = \frac{4 x_m b [(C_e^p)' + (C_e^k)'] \rho_w (1 - \varepsilon)}{\{2 + b [(C_e^p)' + (C_e^k)']\} [C_0^p + (C_e^p)' + C_0^k - (C_e^k)'] z \psi \ln \left(\frac{100 - u_k}{100 - u_p} \right)}. \quad (30)$$

4. METHODS

Column tests were carried out with a filter bed, whose height was 0.5 m. The filter medium was granular activated carbon, type HKW1, produced from bituminous coal by the Chemviron Company. Table 1 lists the GAC and column test parameters. The filtration linear velocity of 5.5 m/h was in the range of the standard velocity used for sand filter media.

Table 1

Column test and GAC parameters

Parameter	Notation	Unit	Value
Height of bed	H	m	0.5
Velocity	V_p	m/h	5.5
Spherical surface of the grains in the bed	ψ	-	0.85
Density of GAC (wet)	ρ_w	kg/m ³	1900
Diameter of the grains in the bed	d_w	m	0.002
Quantity adsorbed in monolayer on one unit of adsorbent	x_m	g/kg	20
Langmuir isotherm constant	b	m ³ /g	0.9

The column test was conducted with model solution that consisted of dechlorinated tap water and analytical grade humic acid and phenol. Humic acid represents all colloids, and phenol represents all dissolved organic compounds.

As the colour of the model solution was derived solely from the humic acid, the research was conducted with two solutions, colour 30 and colour 60, 30 g Pt/m³ and 60 g Pt/m³, respectively. The concentration of phenol was 4 g/m³.

All investigations were carried out according to the standard methods for examination of water and wastewater [15], under natural pH.

Research was conducted to determine the characteristics of the sludge formed during the filtration process (table 2).

Table 2

Sludge parameters

Parameter	Notation	Unit	Colour 30	Colour 60
Surface area	S_0	m ² /g	126	156
Initial water content in sludge	u_p	%	99.962	99.955
Final water content in sludge	u_k	%	99.387	99.384
Viscosity at the beginning of filtration cycle	η_p	N·m ² /s	0.9919	1.037
Viscosity at the end of filtration cycle	η_k	N·m ² /s	2.17	2.646
Absorption ability of sludge for phenol	x_{os}	g/g	0.0035	0.004
Molecular diffusion coefficient of phenol at the beginning of a cycle	D	m ²	$5.4 \cdot 10^{-10}$	$5.2 \cdot 10^{-10}$
Molecular diffusion coefficient of phenol at the end of a cycle	D_z	m ²	$2.48 \cdot 10^{-10}$	$2.04 \cdot 10^{-10}$

5. APPLICATION OF THE MATHEMATICAL MODEL

The results obtained from the research conducted with colour 30 were applied to the mathematical model to determine the number of filtration cycles in one adsorption cycle and as a consequence the length of adsorption cycle for a full scale up-flow filtration bed of the height of 2 m. The coagulant dose used was 0.5 of the theoretical coagulant dose. This figure was determined by previous research as the optimum dose, in terms of performance, for the up-flow filtration bed.

The coagulant dose was determined by:

$$D_k(0.5) = 7\sqrt{B} = 0.5 \cdot 7\sqrt{30} = 19.2 \text{ g/m}^3. \quad (31)$$

The length of the filtration cycle was 12 hours, which determined the variable z :

$$t_c = 12 \text{ h} = \frac{z \cdot 0.85 \cdot 0.5}{5.5} \ln \left(\frac{100 - 99.387}{100 - 99.962} \right), \quad z = 55.84. \quad (32)$$

The concentration of the sludge at the beginning of the filtration cycle was determined by:

$$C_{os}^p = 0.55 \cdot 19.2 + 0.25 \cdot 30 = 18.06 \text{ g/m}^3. \quad (33)$$

The concentration of the sludge at the end of the filtration cycle was determined by:

$$C_{os}^k = 18.06 \frac{100 - 99.387}{100 - 99.962} = 290.53 \text{ g/m}^3. \quad (34)$$

Adsorbate concentration in the outflow from the column at the beginning of filtration cycle was:

$$(C_e^p)' = 3.97 \exp(-2.178 \cdot 10^9 \cdot 5.4 \cdot 10^{-10}) = 1.22 \text{ g/m}^3 \quad (35)$$

Adsorbate concentration in the outflow from the bed at the end of the filtration cycle was:

$$(C_e^k)' = 2.98 \exp(-2.178 \cdot 10^9 \cdot 2.48 \cdot 10^{-10}) = 1.73 \text{ g/m}^3. \quad (36)$$

Number of filtration cycles in one adsorption cycle

$$(N)' = \frac{x'}{(x_1)'} \quad (37)$$

$$= \frac{4 \cdot 20 \cdot 0.9(1.22 + 1.73) \cdot 1900 \cdot 0.55}{[2 + 0.9(1.22 + 1.73)][3.97 - 1.22 + 2.98 - 1.73] \cdot 55.84 \cdot 0.85 \cdot \ln 16.13} = 90,$$

Length of adsorption cycle t_A recalculated for a filter bed of $H = 2$ m:

$$t_A = 180 \text{ d.} \quad (38)$$

Table 3 presents the comparison of the results obtained for colour 30 and colour 60.

Table 3

Comparison of colour 30 and colour 60

Parameter	Notation	Unit	Colour 30 (g Pt/m ³)	Colour 60 (g Pt/m ³)
Coagulant dose	D_k	g/m ³	19.2	27.1
Length of filtration cycle	t_c	hours	12	8
Concentration of sludge at the beginning of filtration cycle	C_{os}^p	g/m ³	18.06	29.9
Concentration of sludge at the end of filtration cycle	C_{os}^k	g/m ³	290.53	409
Adsorbate concentration in outflow at the beginning of filtration cycle	$(C_e^p)^y$	g/m ³	1.22	1.25
Adsorbate concentration in outflow at the end of filtration cycle	$(C_e^k)^y$	g/m ³	1.73	2.25
Number of filtration cycles in one adsorption cycle	$(N)'$	—	90	212
Length of adsorption cycle (bed height 2 m)	t_a	days	180	282

The results indicate that a higher concentration of sludge is obtained for colour 60 compared to colour 30 (409 g/m³ and 290 g/m³, respectively). This increase in sludge concentration has a significant effect on the number of filtration cycles, as well as the length of adsorption cycle for the filtration bed.

The increase in sludge concentration extends the bed life of the GAC from 180 days for colour 30 to 282 days for colour 60.

These results show adsorption of dissolved organic compounds on the grains of GAC as well as adsorption of dissolved organic compounds on the sludge formed. This adsorption by sludge reduces the load on the GAC, and as a consequence extends the life of the activated carbon bed.

6. CONCLUSIONS

The mathematical model presented allows the calculation of filtration bed life based on the results obtained from pilot-scale studies. The adsorption by the sludge formed by coagulation is important as it affects the length of the adsorption cycle.

This is important as it allows study of the economical factors involved in a present-day water treatment plant. The cost of replacing the activated carbon against the cost of the increase in coagulant used.

This research is ongoing with further analysis of the process on modelling solutions with greater colour, as well as with the ability of the up-flow filtration bed to treat natural water.

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MATEMATYCZNY MODEL KOAGULACJI KONTAKTOWEJ W ZŁOŻU WĘGLA AKTYWNEGO

Przedstawiono matematyczny model procesu koagulacji kontaktowej w warstwie granulowanego węgla aktywnego. Wykazano adsorpcyjne właściwości osadu pokoagulacyjnego i jego wpływ na wydłużenie czasu pracy kolumny GWA do wyczerpania pojemności sorpcyjnej węgla dla rozpuszczonych związków organicznych. Zastosowanie modelu przedstawiono dla układów: roztwory modelowe o barwie 30 i 60 g Pt/m³ oraz stężeniu fenolu 4 g/m³—węgiel aktywny Chemviron Carbon typu HKW1.