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ONE-DIMENSIONAL DIFFUSION EQUATION FOR THE PARTICLE SIZE DISTRIBUTION OF PERLITE FILTER GRANULATION

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Preliminary communication
Many filtration systems employing perlite granulations have been designed so far. Size distribution of perlite particles directly influences the retention properties of filter media. The information on the size distribution of perlite particles, used in each specific dead-end filtration process (the flow of fluid being filtered is perpendicular to the surface of filter medium), is crucial for the adequate design of filter medium. In order to facilitate the design of filter systems possessing filter media of this kind, a new and particular mathematical model has been developed for this present study. It is based on an appropriate partial differential equation and additional mathematical conditions, whose solution is an exponential function describing the probability density distribution of perlite particle sizes. The formulated model was experimentally verified by measuring the particle sizes of a perlite granulation using the morphometric method, based on the application of a standard light microscope and digital image analysis software. The fitting procedure of experimental data gave acceptable values of accuracy parameters - high R-square factor (R^2 = 0,905) and small value of the root-mean square error (MSE = 0,490).

Keywords: *filter granulation; morphometric method; partial differential equation; particle size distribution; perlite*

Jednodimenzijska difuzijska jednadžba za raspodjelu veličine čestica granulacije perlita za pročišćavanje

Prethodno priopćenje
Mnoštvo pročistačkih sustava do sada je načinjeno s uporabom različitih granulacija perlita. Raspodjela veličina čestica perlita izravno utječe na svojstva retencije pročistačkih medija. Informacije o raspodjeli veličine granulacija perlita, koji se koriste u svakom konkretnom postupku dead-end filtracije (filtracija koja se odvija u pravcu okomitom na ravninu pročistača), od presudne je važnosti za odgovarajuću tvorbu pročistaćkog medija. Kako bi se olakšala tvorba pročistaćkog medija koji posjeduje pročistaćke sposobnosti ove vrste, novi i specifični matematički model je razvijen za ovo istraživanje. Temelji se na odgovarajućoj parcijalnoj diferencijalnoj jednadžbi i dodanim matematičkim uvjetima, čije rješenje je eksponencijalna funkcija koja opisuje vjerojatnost raspodjele gustoće različitih veličina čestica perlita. Formulirani model eksperimentalno je potvrđen mjerenjem veličine čestica perlita pomoću morfometrijske metode, koja se temelji na primjeni standardnog svjetlosnog mikroskopa i softvera za digitalnu analizu slika. Postupak pronalaženja najtočnije funkcije za rezultate istraživanja ostvaruje zadovoljavajuću točnost - visoki R^2 čimbenik (R^2 = 0,905) i mala vrijednost srednje kvadratne pogreše (MSE = 0,490).

Ključne riječi: *filter granulacija; morfometrijska metoda; parcijalna diferencijalna jednadžba; perlit; raspodjela veličine čestica*

1 Introduction

Perlite is volcanic glass mainly composed of silica, aluminium, potassium, sodium and 2÷5 % of constitutive water. The commercial sense of the term perlite assumes any kind of volcanic silicates that will expand and form lightweight material when is heated quickly. During the heating process at temperatures between 760 and 1200 °C, water trapped in the structure of the material vaporizes causing the expansion of the material volume from about 7 to 16 times with respect to its original value. Consequently, the bulk density of expanded perlite is 30÷150 kg/m³, cca. 1100 kg/m³ for unexpanded (raw) material. The expanded material is a brilliant white, due to the reflectivity of the trapped bubbles.

Some specific properties of perlite, such as: extremely high water retention, low thermal conductivity, high adsorption of sound, low volume density and low price makes this material very suitable for application in different technical areas. Consequently, many applications of perlite have been developed, including applications in: petro-chemical industry for cementing of oil wells [1]; industrial filtration [2]; industry of manufacturing insulation materials [3], engineering geology for drainage [1]; horticultural industry for potting mixes [4]; adsorption of heavy metals like Cd, Cu (II) and Cr(III) [5÷10]; removal of toxic heavy metals like Pb [11], as well as simultaneous removal of Pb(II), Co(II) [12], Zn [13] and arsenate As(V) adsorption [14].

Many of these technical challenges are related to the particle size distributions of applied perlite granulations. For example, immobilization of pathogenic *Acinetobacter junnii* using perlite depends on particle size [15]. As an analogue, preparation of micro-filtration membranes is based on the crushed natural perlite powder of appropriate size, mixed with adequate organic additives and water [16].

Perlite size distribution directly influences retention properties of filter media. Therefore, information on the particle size distribution of perlite granulation used in each specific filtration process is important for adequate design of filter medium. In this way, the particle size distribution of the perlite granulation is analysed in present study. The mathematical model, based on appropriate partial differential equation, is formulated and experimentally verified.

2 Materials and methods

This paper presents a mathematical model, which describes the probability of density distributions of perlite filter granulations. The model is based on a partial differential equation of the parabolic type, governed by specific additional mathematical conditions. According to the authors' knowledge, the application model of this kind (analytical approximation of size distribution of perlite granulation particles) has not been noted as yet.

Having in mind that this model is in the focus of interest of the present study, and represents the main contribution of this paper, it is presented in section 2 ("Results and discussion") in detail. However, experimental details, statistical characterisation and fitting of experimental data used for model verification are described in the present section ("Material and methods").

2.1 Experimental setup and conditions

In the paper is analysed and discussed particle size distribution of a perlite granulation Harborlite 900 (Harborlite®, World Minerals Company 130, CA - USA), used in beverage filtration, Fig. 1.

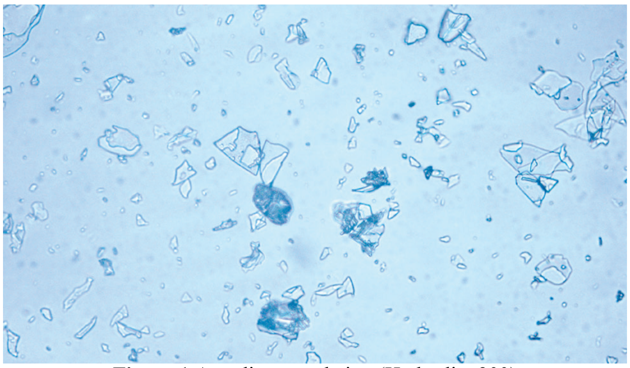

Figure 1 A perlite granulation (Harborlite 900)

Particle sizes are measured by the morphometric method (magnification 400×), commonly used in the microbiology for bacteriological analysis for measurement and parameterisation of microscopic objects e.g. sizes and shapes of cells and their parts [17]. The morphometric method is an accurate low-cost and very simple method, based on the application of classic light microscope (Fig. 2).

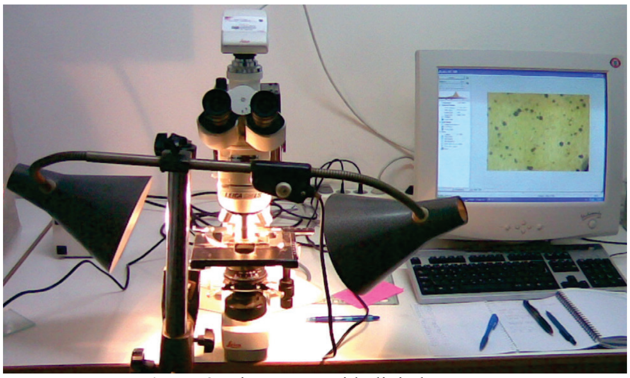

Figure 2 Microscope with digital camera

To provide optimal experimental conditions for digital image analysis, a tested sample of perlite granulation is mixed with distilled water to achieve the "dilution ratio" of 10^{-3}, giving a suspension of 1 g of perlite and 1000 ml of distilled water. Digital images of perlite particles, acquired by *Leica DMLS* microscope, (equipped with objectives providing magnification ratios of 50×, 100×, 200×, 400×, 1000×) with a built-in digital camera (*Leica DC 300*), are analyzed by software *Leica IM 1000* (specified for digital image analysis). More precisely, the image area A of each scanned particle was digitally measured, and the particle effective diameter d was calculated using a well-known formula:

$$d=\sqrt{\frac{4\cdot A}{\pi}}, \tag{1}$$

As a result, particle size distribution of analyzed perlite granules is formulated, providing an experimental data base for mathematical model verification.

2.1 Statistical characterisation and data fitting

Measured particle diameters of investigated sample are allotted to intervals of uniform width (Δd = 5 μm), starting from 2,5 μm and up to 67,5 μm. Number of particles having diameters smaller than 2,5 μm took 3,6 % of total sample, while the number of particles larger than 67,5 μm was 0,3 %. In order to increase the fitting accuracy, these particles were trimmed from the original sample (weak influence on the filtration process). Consequently m = 13 size classes of perlite granulation particles were established (2,5 ÷ 7,5 μm) up to (62,5 ÷ 67,5 μm). Each size-class is represented by the interval midpoint d_i (i = 1, 2, ... , m), starting from 5 μm (for the smallest class of particles) and up to 65 μm (for the largest particles). The absolute frequency n_i (i = 1, 2, ... , m) is evaluated for each class of particle sizes. An empirical discrete frequency distribution table, containing m = 13 data pairs (d_i, n_i) was established. Consequently, the sum of absolute frequencies equals total number of particles in the analyzed sample, i.e.

$$n=\sum_{i=1}^{m} n_i, \tag{2}$$

The relative frequency (%) of particles is expressed from:

$$f_i=\frac{n_i}{n}\cdot 100,\quad (i=1,2,\ldots,m). \tag{3}$$

On the basis of frequency distribution, the common statistic parameters are evaluated, as well as the probability density function (*pdf* in further text), given by:

$$pdf(d_i)=\frac{f_i}{\Delta d}\left(\frac{\%}{\mu\mathrm{m}}\right),\quad (i=1,2,\ldots,m), \tag{4}$$

pdf values of particles with diameters d_i (i = 1, 2,..., m) are fitted following the iterative algorithm of Levenberg-Marquardt [18, 19] to adjust the parameter values of exponential model function:

$$y=a_1\,\mathrm{e}^{bx}, \tag{5}$$

based on two fitting parameters (a_1 and b).

The acceptance of non-linear fit is estimated by two commonly used parameters. The first is the coefficient of determination:

$$R^2 = 1 - \frac{\sum_{i=1}^{m} \left(y_i - \hat{y}_i\right)^2}{\sum_{i=1}^{m} \left(y_i - \overline{y}_i\right)^2}, \tag{6}$$

where y_i denotes experimental *pdfs*, defined by (4), $\hat{y}_i$ are fitted values and $\overline{y}$ denotes the mean *pdf* value.

The second parameter for estimation of fitting accuracy is the root mean square error, or standard error of estimation:

$$RMS_e = \sqrt{\frac{\sum_{i=1}^{m} \left(y_i - \hat{y}_i\right)^2}{m-k}} \tag{7}$$

where m = 13 is the number of particles size-classes and k = 2 is the number of constants in the fitting function (5).

3 Results and discussion

3.1 Formulation of the mathematical model

An original mathematical model, based on a parabolic partial differential equation and additional conditions, has been developed and presented in this work. It is shown that the problem under consideration can be described by the following homogeneous diffusion equation:

$$y_t = \left[C(x)\, y_x\right]_x , \tag{8}$$

having its particular solution:

$$y(x,t)\big|_{t=1} = \mathrm{e}^{a+bx} = a_1 \mathrm{e}^{bx} \;\Rightarrow\; a_1 = \mathrm{e}^{a}, \tag{9}$$

in the form of expression (5). The function $y = y(x, t)$ represents *pdf* of perlite granulation particle sizes, while x and $t > 0$ are space coordinates and time, respectively. The function $C = C(x)$ is unknown and b and a_1, resp. a are given constants.

To get an idea which kind of functions $y = y(x, t)$ and $C = C(x)$ could be, a simplification of this problem will be made by introducing the following additional condition: The simplest case is if $C(x) = c^2 = const$. Then the problem is reduced to solve the diffusion equation:

$$y_t = c^2 y_{xx}. \tag{10}$$

It is well known that:

$$y(x,t) = \frac{1}{2c\sqrt{\pi t}}\, \mathrm{e}^{-\frac{x^2}{4c^2 t}}, \tag{11}$$

represents a solution of (10) [19]. This gives the idea that for not constant $C = C(x)$ as in (8) the desired solution of (8) has the form:

$$y(x,t) = \frac{A}{t^{\alpha}}\, \mathrm{e}^{\frac{g(x)}{t^{\beta}}}, \tag{12}$$

where A, α and β are unknown constants. Also $g(x)$ has to be determined. Taking t=1 in (12) and equating this to (9) implies:

$$A = 1,\; g(x) = a + bx . \tag{13}$$

Hence (12) becomes:

$$y(x,t) = \frac{1}{t^{\alpha}}\, \mathrm{e}^{\frac{a+bx}{t^{\beta}}} . \tag{14}$$

Substituting (14) in (8) yields the differential equation:

$$C'(x) + \frac{b}{t^{\beta}} C(x) = -\frac{\alpha}{b} t^{\beta-1} - \frac{a\beta}{b}\frac{1}{t} - \beta\frac{x}{t}, \tag{15}$$

with three unknowns, α, β and $C = C(x)$ and two independent variables x and t. If one equation with three unknown functions is given, the solution is not unique. But in equation (15) only such C are desired which does not include t, because $C(x)$ does not possess the variable t. This fact reduces the set of $C(x)$ and determines the constants α and β.

To obtain the solution of (15) the following additional condition is introduced, assuming $C(x)$ to be of the form:

$$C(x) = \sum_{n=0}^{\infty} c_n x^n, \tag{16}$$

with unknown constant coefficients c_n. Inserting the infinite power series (16) in (15) gives a series equation. This series equation is satisfied for all values of x, only if the coefficients of distinct powers of x are zero. Thus:

$$c_1 + \frac{b}{t^{\beta}} c_0 = -\frac{\alpha}{b} t^{\beta-1} - \frac{\alpha\beta}{b}\frac{1}{t}, \tag{17}$$

$$2c_2 + \frac{b}{t^{\beta}} c_1 = -\frac{\beta}{t}, \tag{18}$$

$$(n+1)c_{n+1} + \frac{b}{t^{\beta}} c_n = 0,\quad n \geq 2. \tag{19}$$

These are three recursion relations for the coefficients c_n. The c_n do not include t powers. e.g., α and β must be determined in this way that c_n is constant. Looking at the t powers in (17), (18) and (19) it can be seen that three cases related to unknown constant β in expression (12): $\beta = 0$, $\beta \neq 1$ and $\beta = 1$ have to be distinguished.

3.1.1 Case 1: $\beta = 0$

If $\beta = 0$, (17) becomes:

$$c_1 + b c_0 = -\frac{\alpha}{b}\frac{1}{t}, \tag{20}$$

comparison of coefficients in this relation with respect to t^0 and t^{-1} gives:

$$c_1 = -bc_0,\ \alpha = 0. \tag{21}$$

Thus relation (18) becomes:

$$c_2 = \frac{b^2}{2} c_0, \tag{22}$$

and (19) has the form:

$$c_n = -\frac{b}{n} c_{n-1},\ n \geq 3. \tag{23}$$

By using (22) this recursion relation yields:

$$c_n = (-1)^n \frac{b}{n!} c_0,\ \ n \geq 0. \tag{24}$$

Then, (16) becomes:

$$C(x) = c_0 \sum_{n=0}^{\infty} (-1)^n \frac{b^n}{n\,!} x^n = c_0 \mathrm{e}^{-bx}. \tag{25}$$

Taking solution (14) with $\alpha = \beta = 0$ above gives:

$$y(x,t) \equiv y(x) = \mathrm{e}^{a+bx}. \tag{26}$$

The function $y(x)$ is a solution of the stationary form of the diffusion equation:

$$\left[C(x)\, y_x\right]_x = 0, \tag{27}$$

and cannot be used.

3.1.2 Case 2: $\beta \neq 1$

For $\beta \neq 1$ and using $\beta \neq 0$ above, the comparison of coefficients in (18) yields:

$$c_2 = 0,\ c_1 = 0 \text{ and } \beta = 0. \tag{28}$$

The last equation in (28) is a contradiction to precondition $\beta = 1$. Another way for proving that $\beta \neq 1$ never can be true is given by using (19). Thus the recurrence formula (19) gives $c_n = 0$, $n \geq 2$, by equating to zero the coefficients of distinct powers of t. Therefore, $C(x) = c_0 = const.$ because $c_1 = c_2 = 0$ in (28). In this case the diffusion eq. (8) becomes (10) with $c^2 = c_0$ and possesses the solution:

$$y(x,t) = \frac{1}{2\sqrt{c_0 \pi t}} \mathrm{e}^{-\frac{x^2}{4c_0 t}}, \tag{29}$$

which contradicts to (14) resp. (9).

3.1.3 Case 3: $\beta = 1$

So the remaining alternative is $\beta = 1$. In this case Eqs. (17), (18) and (19) take the form:

$$c_1 + \frac{b}{t} c_0 = -\frac{\alpha}{b} - \frac{a}{bt}, \tag{30}$$

$$2c_2 + \frac{b}{t} c_1 = -\frac{1}{t}, \tag{31}$$

$$c_{n+1} = -\frac{b}{(n+1)\cdot t} c_n,\ n \geq 2. \tag{32}$$

Equating the coefficients of t powers to zero in (32) gives:

$$c_2 = c_3 = \ldots = c_n = 0,\ n \geq 2. \tag{33}$$

So the infinite series (16) reduces to the linear function:

$$C(x) = c_0 + c_1 x. \tag{34}$$

Taking $c_2 = 0$ in (33) and inserting it in (31) implies:

$$c_1 = -\frac{1}{b}. \tag{35}$$

Expression (31) also determines $c_2 = 0$ independent of (32). Substituting (35) in relation (30) and using the comparison of coefficients of t^0 and t^{-1} concludes:

$$\alpha = 1,\ \ c_0 = -\frac{a}{b^2}. \tag{36}$$

Therefore (34) is:

$$C(x) = -\frac{a + b\,x}{b^2}. \tag{37}$$

Hence the diffusion Eq. (8) becomes:

$$y_t + \frac{1}{b} y_x + \frac{a+bx}{b^2} y_{xx} = 0. \tag{38}$$

Taking (14) with $\alpha = \beta = 1$, the solution of (38) is:

$$y(x,t) = \frac{1}{t} \mathrm{e}^{\frac{a+bx}{t}}, \tag{39}$$

and includes the particular solution (9) of Eq. (8) when $t = 1$.

For normalization of parameter $\mathrm{e}^a = a_1$ normalization factor D and constants β_1 and β_2 have to be calculated in a way to satisfy the following condition:

$$D\int_{\beta_1}^{\beta_2} y(x, t=1)\mathrm{d}x = \frac{D}{b}(\mathrm{e}^{a+b\beta_2} - \mathrm{e}^{a+b\beta_1}) = 1, \tag{40}$$

$a, b < 0$.

If $-\infty \triangleleft \beta_1 \triangleleft \beta_2 \triangleleft \infty$ resp. $\beta_1 = -\infty$, $\beta_1 \triangleleft \beta_2 \triangleleft \infty$, then

$$D = b\,\mathrm{e}^{-a}(\mathrm{e}^{b\beta_2} - \mathrm{e}^{b\beta_1})^{-1} \text{ resp. } D = b\,\mathrm{e}^{-(a+b\beta_2)}. \qquad (41)$$

It is proven in chapters 3.1.1 and 3.1.2 that cases 1 and 2 cannot be used. It follows that only case 3 gives the acceptable solution (39) of partial differential Eq (8). For t = 1, solution (39) gets the desired form of (9).

3.2 Experimental verification of the model

Basic statistical parameters of particle size distribution of analyzed perlite granulation are given in Tab. 1. The mean diameter is 18,37 µm, while the maximum value is 65 µm. Coefficient of variation C_V = 66,42 % shows that size distribution of this perlite granulation is fairly weakly concentrated around the mean diameter.

Table 1 Descriptive statistics of the particle size distribution of perlite granulation Harborlite 900

Parameter	Value
Mean / µm	18,37
Min / µm	→ 0
Max / µm	65
RMS / µm	12,2
Coeffient of Variation – C_V / -	66,42
Skewness *S* / -	1,18
Flatness *F* / -	4,17

Particle size distribution of filtration medium shows significant deviation from the normal distribution: skewness factor is 1,18, while the corresponding value for the normal distribution is 0. According to the flatness factor, having a value of 4,17, the size distribution of this perlite granulation also deviates from the medium flattened normal distribution characterized by flatness factor value of 3.

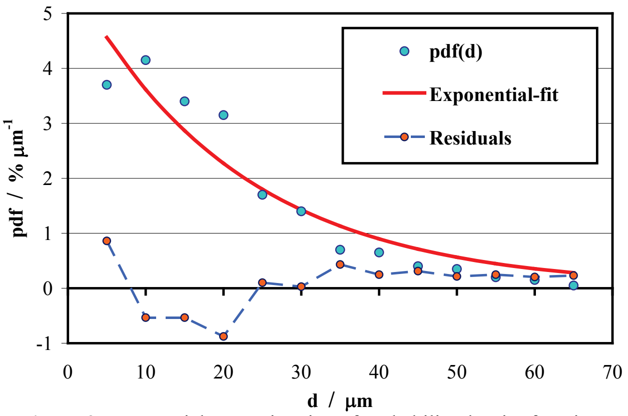

Figure 3 Exponential approximation of probability density function (*pdf*) of perlite size distribution (diameter *d*)

The approximation of perlite granulation size *pdf* by exponential function (5) is illustrated in Fig. 3, together with residuals (fitting errors). The fitting procedure gave coefficients:

a_1 = 5,75355993885832,
b = ÷0,04644545597440,

and acceptable values of accuracy parameters, i.e. high *R*-square factor R^2 = 0,905 and a fairly small value of the root-mean square error - *Root MSE* = 0,490. This way, the applicability of the formulated mathematical model, whose solution is the exponential function (5), has been experimentally verified.

4 Conclusion

The presented approach enables detailed analysis of perlite granulations as non-stable matrix filtration materials. Particle size distribution has a profound effect on constitutive properties of the filter material and filter performances. The primary reason for measuring the sizes of particles is to predict behaviour of perlite filter material in a separation process and to specify the performance of a filter medium in terms of its ability to retain particles of different sizes. Based on the known particle sizes distributions of several perlite granulations, it is possible to establish various additional filter granulation mixtures, whose particle size distributions are optimized for any specific filtration requirements. Thus, a variety of filtration mediums can be produced from just a few granulations of known particle size distributions [20].

However, pure raw experimental data on the size distribution are not suitable enough for a sophisticated analysis and design of contemporary highly efficient filtration systems and their components. Advanced methods, based on the mathematical modelling including those employing the partial differential equation models can significantly enhance and facilitate the design procedure. Therefore, in the present study a new mathematical model has been formulated and verified, based on hyperbolic diffusion partial differential equation, whose only acceptable solution is the exponential function (5), i.e. (9). It has been verified by successful fitting of the experimental data acquired from existing perlite granulation. Presented model could be applied in beverage industry for analysis of perlite filter aids specified to separate solid particles from liquids.

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