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APPLICATION OF THE STOCHASTIC MODEL FOR ANALYSIS OF FLOTATION KINETICS WITH COAL AS AN EXAMPLE

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Flotation as a random process, in which the random variable representing the number of particles raised to the froth layer during a fixed time t depends on time, is a stochastic process. As a result of turbulence of the medium in the flotation chamber, apart from the process of particles adhesion to bubbles, a reverse process occurs whose intensity depends on the ash content. This is the process of detachment the particles from bubbles. Such a situation is described best by the stochastic process of birth and death.

The paper briefly presents the assumption and differential equation of the model as well as its solution in the form of the equation of flotation kinetics. The authors have presented the interpretation of equation parameters. According to the empirical dependences of recovery of floating particles in the froth product on time for several coal samples, differing by the ash content, the following parameters of the kinetics equation were calculated: the resultant adhesion rate constant and permanent adhesion rate constants. The resultant adhesion rate constant, being the sum of permanent adhesion and detachment rate constants, is independent on the ash content whereas the permanent adhesion rate constant decreases with the increase of ash content.

Key words: flotation kinetics, stochastic process, model of birth and death, adhesion rate constant, detachment rate constant

INTRODUCTION

Flotation, as any technological process in which the results are determined by many random factors, is the process occurring in time. In order to form a permanent attachment between a particle and a bubble there must be, first of all, a collision between a particle with a bubble and the kinetic energy must be contained in a certain range of values, on the one hand large enough to overcome the barrier of the potential

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of the particle-bubble interaction and, on the other, little enough to make this connection stable. In other words, not to detach the particle from the bubble. Both the particle-bubble collision and the value of particle kinetic energy are of random character.

As it can be seen, the permanent particle-bubble connection is determined by a set of random events whose probabilities affect the velocity of the process course. The higher probability, the faster is the flotation process.

Kinetics, i.e. the course of the process in time, results not only from the statistical character of phenomena occurring on the phase boundaries but also from the successive inflow of free surface into the flotation system on which the adhesion of mineral particles in the form of air bubbles occurs and which, among others, limits the velocity of the process course.

Many authors dealt with the problem of flotation kinetics: (Zuniga 1935, Schuhmann 1942, Sutherland 1947, Beloglazov 1947, Pogorelyj 1961a,b, 1962, Bushell 1962, Imaizumi and Inoue 1963, Melkitch 1963a, 1963b, 1964, Tomlinson and Fleming 1963, Bogdanov et al., 1964, Bogdanov et al., 1964, Volin and Swami 1964, Zeidenberg et al., 1964, Harris and Rimmer 1966, Loveday 1966, Tille and Panu 1968, Inoue and Imaizumi 1968, Kapur and Mehrotra 1973, 1974, Mehrotra and Kapur 1974, Mehrotra and Kapur 1975, Trahar and Warren 1976, Collins and Jameson 1976, Harris 1978, Maksimov and Jemelianov 1983, Xu Changlian 1985, Szatkowski and Freyberger 1985, Vanangamudi and Rao 1986, Laskowski et al., 1991, Varbanov et al., 1993, Lazic and Calic 2000).

In a way, there is an analogy between the mechanism of chemical reaction and the mineralization of the air bubble in the flotation process. Therefore, the flotation kinetics is described by the equation analogical to the equation of kinetics of chemical reaction.

Zuniga (1935) was the first who applied the differential equation of kinetics of chemical reaction to the description of kinetics of batch flotation. It can, in its general form, be written as follows:

$$\frac{d C}{dt} = - k C^n \quad (1)$$

where: $C(t)$ – concentration of floating particles remaining in the flotation chamber up to the moment t ; k – flotation rate constant; n – constant characterizing the order character of the process (order of flotation kinetics).

The flotation rate constant, occurring in this equation, is a macroscopic parameter which should contain information about the process affecting factors. This information is recorded by the models of flotation rate constant which determines the process kinetics.

Equation (1) is the equation of flotation kinetics of particles which are homogeneous in their surface properties, i.e. equal-floating particles which possess the

same value of the flotation rate constant. After solving equation (1) for $n = 1$ and calculating the recovery of floatable particles to the froth product, the following expression is obtained:

$$\varepsilon(t) = 1 - \exp(-k t) \quad (2)$$

As it results from equation (2), after long flotation time (theoretically infinitely long), all floating particles in the given physicochemical conditions will have been floated out. It is known from experience that flotation is a kinetic and thermodynamic phenomenon (Drzymala, 2001). It means that the value of recovery after a long time of flotation, especially for larger and difficult to float particles, is smaller than 1. There are no analogical cases in the kinetics of chemical reaction.

After imposing the following boundary condition upon equation (2):

$$\lim_{t \rightarrow \infty} \varepsilon(t) = \varepsilon_\infty \quad (3)$$

the dependence of recovery on time is expressed by the function:

$$\varepsilon(t) = \varepsilon_\infty [1 - \exp(-k t)] \quad (4)$$

Formula (4) represents a general form of kinetics equation of the 1st order. In such a concept this is a determinist model. The parameters of this model are empirical constants. The notion of equilibrium recovery ε_∞ was introduced into the model of chemical reaction as a second parameter to readjust the model to empirical dependences. There were no theoretical premises in the construction of the basic equation of kinetics which could justify such a step. The equation of the process of birth and death is such an equation of kinetics, known in the theory of stochastic processes (Bailey 1964), the solution of which comprises all the subprocesses present in mineralization of bubbles.

STOCHASTIC MODEL OF KINETICS OF BATCH FLOTATION

In the stochastic model the number of particles $N(t)$ raised to the froth level up to time t is the random variable depending on time. From the point of view of theory of stochastic processes the model based on the kinetics of chemical reaction is a pure birth process. The particles which were subjected to adhesion to bubbles are not able to return to the phase of suspension.

As a result of turbulence of medium in the flotation chamber, despite the process of adhesion of particles to bubbles, there is a reverse process of lower intensity, i.e. the process of detachment the particles from the air bubbles (Mika and Fuerstenau 1968, Schulze 1977, Woodburn et al., 1971, Schulze 1992, Maksimov and Emelianov 1983,

Geidel 1985, Honaker and Ozsever 2003). The process of birth and death is a model which describes such a process.

The model of the process of birth and death was applied and solved mathematically by Litwiniszyn (1966) for the description of kinetics of the colmatage process. In this process the particles of solids, present in the liquid, are being caught in the porous medium as a result of surface interactions of this medium with particles. Here two sub-processes may appear: adhesion to the porous body and detachment resulting from interactions with the flowing liquid.

The process of ion flotation (Stachurski 1970) and the process of wet magnetic separation (Siwiec 1982) run according to the model of birth and death. If the state of system in which l particles are attached to bubbles is marked as E_l and the state in which $l+1$ particles are attached is marked as E_{l+1} , in the process of birth and death, the transitions from E_l to E_{l+1} and E_l to E_{l-1} are possible.

For the transition from E_l to E_{l+1} the function of intensity of the adhesion process $\lambda(l)$ is formed whereas for the transition from the state E_l to E_{l-1} the function of intensity of the detachment process $\mu(l)$ is formed, according to the following formulas:

$$\lambda(l) = \lambda_o(n_o - l) \quad (6a)$$

$$\mu(l) = \mu_o l \quad (6b)$$

where: n_o – number of particles floating in the floatation chamber in the moment $t = 0$; λ_o and μ_o – certain constants. Thus the intensity of the adhesion process is proportional to the number of particles remaining in the free state ($n_o - l$), while the intensity of the process of detachment is proportional to the number of particles attached to bubbles (l). Probability $P_l(t)$ that at the moment t there are l particles attached to bubbles is fulfilled by the following system of differential equations (Litwiniszyn 1966):

$$\frac{dP_l(t)}{dt} = -[\lambda_o(n_o - l) + \mu_o l]P_l(t) + \lambda_o(n_o - l + 1)P_{l-1}(t) + \mu_o(l + 1)P_{l+1}(t) \quad (7)$$

while $l = 1, 2, \dots, n_o$.

Equations (7) has the following solution:

$$P_l(t) = \left(\frac{n_o}{l} \right) \frac{l}{(\lambda_o + \mu_o)^{n_o}} \left[(\mu_o + \lambda_o) e^{-(\lambda_o + \mu_o)t} \right]^{n_o - l} \left[1 - e^{-(\lambda_o + \mu_o)t} \right]^l \quad (8)$$

The average value of the random variable $N(t)$ is:

$$E[N(t)] = \sum_{l=0}^{n_o} l P_l(t) = \frac{n_o \lambda_o}{\lambda_o + \mu_o} [1 - e^{-(\lambda_o + \mu_o)t}] \quad (9)$$

Accordingly, the recovery of particles raised to the froth layer is expressed by the formula:

$$\varepsilon(t) = \frac{E[N(t)]}{n_o} = \frac{\lambda_o}{\lambda_o + \mu_o} [1 - e^{-(\lambda_o + \mu_o)t}] \quad (10)$$

The form of this formula is analogical to formula (4) while $\varepsilon_\infty = \frac{\lambda_o}{\lambda_o + \mu_o}$ and $k = \lambda_o + \mu_o$. It can be said that the constants λ_o and μ_o are, respectively, the constants of the process of adhesion and detachment of particles from the air bubbles. These constants can be calculated from formula (10) because:

$$\lim_{t \rightarrow \infty} \varepsilon(t) = \frac{\lambda_o}{\lambda_o + \mu_o} = \varepsilon_\infty \quad (11)$$

$$\left. \frac{\partial \varepsilon(t)}{\partial t} \right|_{t \rightarrow 0} = \lambda_o \quad (12)$$

Matching the empirical dependence to the model one it is possible to evaluate the values of the processes of adhesion and detachment under given physicochemical and hydrodynamic conditions in the flotation chamber.

A PHYSICAL INTERPRETATION OF THE BIRTH AND DEATH MODEL CONSTANTS

It results from formula (4) of recovery of a useful mineral in the froth product for $\varepsilon_\infty = 1$ that after the appropriately long time (theoretically infinitely long) the whole floatable mineral will be transferred to the froth product (all particles connected with the air bubbles will find their way to the froth product). The flotation rate constant is:

$$\frac{d\varepsilon}{dt}(t=0) = k \quad (13)$$

A comparison of this formula to formula (12) leads to the conclusion that in the case of flotation without detachment the flotation rate constant is equivalent to the resultant adhesion rate constant. All the particles attached to the air bubbles will pass to the froth product. None of them will be detached from the flotation aggregate.

From formula (4) for $\varepsilon_\infty = 1$ the flotation rate constant and, at the same time, the resultant adhesion rate constant is:

$$k = \frac{1}{1-\varepsilon} \frac{d\varepsilon}{dt} \approx \frac{1}{1-\varepsilon} \frac{\Delta\varepsilon}{\Delta t} \quad (14)$$

Since

$$\varepsilon = \frac{l}{n_o} \quad \text{and} \quad \Delta\varepsilon = \frac{\Delta l}{n_o} \quad (15)$$

then

$$k = \frac{\Delta l}{\Delta t(n_o - l)} = S_b \frac{l_c}{\Delta t(n_o - l)} \frac{\Delta l}{S_b l_c} = S_b P_c P_a \quad (16)$$

where : n_o – initial number of floatable particles in the flotation chamber; l - number of particles attached to the air bubbles up to the time t ; Δl - number of particles attached to the bubbles in the time Δt ; l_c - number of particles colliding with the bubble in the unit time; S_b - total surface of bubbles passing through the surface unit of the cross-section of the flotation machine in the time unit; P_c and P_a – probabilities of collision and adhesion, respectively,:

$$P_c = \frac{l_c}{\Delta t(n_o - l)} \quad P_a = \frac{\Delta l}{S_b l_c} \quad (17)$$

The condition of adhesion to occur is the collision between a particle and a bubble. Thus, the resultant adhesion rate constant is the product of probability of collision and probability of adhesion on the surface of air bubbles flowing through the surface unit of the cross-section of the flotation chamber in the time unit. Resultant adhesion is therefore a number of all particles which were instantaneously or permanently attached to the bubbles surfaces. A part of them will be detached as a result of turbulent movements of the medium. Consequently, the number of particles attached permanently to the bubble surface is the difference between the total number of particles attached to bubbles and the number of particles detached from them. The particles attached permanently will be raised to the froth product. Therefore, in the case of the model of flotation, with respect to the process of particle detachment from the bubble surfaces, the following scheme of the process of adhesion (in the sense of balance of the number of particles) can be assumed:

$$\text{Permanent adhesion} = \text{resultant adhesion} - \text{detachment} \quad (18)$$

As it results from the above scheme and formula (10), the constant λ_o denotes the permanent adhesion rate constant whereas the flotation rate constant k is the resultant adhesion rate constant.

It is an obvious fact that such particles are detached which were previously subjected to adhesion to the bubble. Therefore the detachment rate constant will be the product of the resultant adhesion rate constant k and probability of detachment, P_d :

$$\mu_o = k P_d = S_b P_c P_a P_d \quad (19)$$

On the other hand, according to scheme (18), the constant of permanent adhesion rate constant is :

$$\lambda_o = k - \mu_o = S_b P_c P_a - S_b P_c P_a P_d = S_b P_c P_a (1 - P_d) \quad (20)$$

Applying formulas (20) and (19) it is possible to calculate the value of equilibrium recovery,

$$\varepsilon_\infty = \frac{\lambda_o}{\lambda_o + \mu_o} = \frac{S_b P_c P_a (1 - P_d)}{S_b P_c P_a} = 1 - P_d \quad (21)$$

Thus, P_d , λ_o and μ_o can be determined from the empirical dependence $\varepsilon(t)$. This fact creates an additional tool for studying the basis of the process of mineralization of air bubbles in the mass process.

EXPERIMENTAL

METHOD OF SAMPLES PREPARATION

Coal of the special seam (type 33) of the Piast Mine was used which was crushed in the roller crusher below 0.5 mm. The wet screen analysis was applied, obtaining the particle size fractions (0.5-0.4), (0.4-0.315), (0.315-0.2), (0.2-0.1) mm. After drying, each size fraction was subjected of float and sink analyses. The analysis was carried out in the solution of zinc chloride. Narrow density fractions (-1.35), (1.35-1.5), (1.5-1.7), (1.7-1.8), (1.8-2.0) and (+2.0) Mg/m³ of size fraction were obtained as products of separation. The narrow size fraction-density fractions were stored in vacuum to limit the oxidation of coal surface and next then were used for research. Ash content was determined in every sample.

DETERMINING THE FLOTATION KINETICS

Flotation experiments for coal were performed at room temperature in the Denver laboratory apparatus of 1 dm³ capacity with the constant rotor speed 2020 rpm and the fixed air flow-rate (Mlynarczykowska, 2004). The content of solids was the same in all experiments and was 80 g/dm³. Such conditions ensured the constant amount and size of air bubbles in the chamber with limited turbulence of flotation pulp caused by the rotor. Low concentration of flotation pulp was used for practical reasons because

at low pulp density it is possible to obtain favourable results of coal flotation (Sablik 1998).

The weighed portion was wetted for 15 min in the solution of the flotation agent of predetermined concentration, next stirred for 5 min without air. The aqueous solution of butanol was used as a collecting and frothing agent which does not change pH of pulp but only decreases the surface tension at the gas – liquid interface which causes the air dispersion in the suspension to increase. Also its adsorption on the surface of air bubbles follows which ensures their stabilization and prevents coalescence (Malysa 2000a,b, Krzan and Malysa 2002). On the basis of initial investigations, proper concentrations of the reagent were selected ensuring the formation of froth of appropriate structure and durability.

In the performed series of investigations the fractionated flotation was carried out in which the concentrates were collected in the following time intervals: 15, 15, 30, 30, 30 s and every 1 min. The time of carrying out the flotation depended on particle sizes and fraction density. As a rule the last froth product was collected after 6 minutes of flotation. The samples were weighed after drying and the ash content was determined.

RESULTS AND DISCUSSION

The investigations of flotation kinetics of respective densimetric fractions of size fraction 0.2-0.315 mm were performed at butanol concentration of $2 \cdot 10^{-3}$ mol/dm³ and the corresponding surface tension 68.5 mN/m. Its measurements were made by means of the ring method with the digital K9 KRUESS tensiometer at room temperature (about 22°C). The measurement accuracy was 0.1 mN/m.

The curves of flotation kinetics were drawn according to the results of empirical tests of this sample. The empirical dependences were approximated by model equations of flotation kinetics of the 1st order (Eq. (4)). The parameters of this equation, ε_∞ and k , were calculated in the following way. For the given value ε_∞ equation (4) can be transformed to the form:

$$\ln \frac{\varepsilon_\infty}{\varepsilon_\infty - \varepsilon} = k t = y \quad (22)$$

Next, the constant k was calculated by the least square method. The rate of agreement of the model dependence with empirical courses was evaluated by means of the curvilinear correlation coefficient R :

$$R = \sqrt{1 - \frac{\sum_{i=1}^n (\varepsilon_i - \hat{\varepsilon}_i)^2}{\sum_{i=1}^n (\varepsilon_i - \bar{\varepsilon})^2}} \quad (23)$$

in which: $\bar{\varepsilon}$ – average value of recovery; $\hat{\varepsilon}_i$ - value of recovery calculated from the model in the i -th point; ε_i - experimental value of recovery in the i -th point; n – number of experimental points. The value ε_∞ was changed every 0.01 to reach the maximum value of the curvilinear correlation coefficient.

Figure 1 shows the model curves of flotation kinetics. Empirical values are marked by circles. In all cases the curvilinear correlation coefficient is larger than 0.96. Model dependences $\varepsilon(t)$ are given by each figure. The permanent adhesion rate constant was calculated on the basis of model dependences $\varepsilon(t)$, according to formulas (12), (11) and (4):

$$\lambda_0 = \lim_{t \rightarrow 0} \frac{d\varepsilon(t)}{dt} = k \varepsilon_\infty \quad (24)$$

The calculated values of the permanent adhesion rate constant are given by each model dependence. As it can be seen in the quoted results, the resultant adhesion rate constant k is practically independent from the ash content.

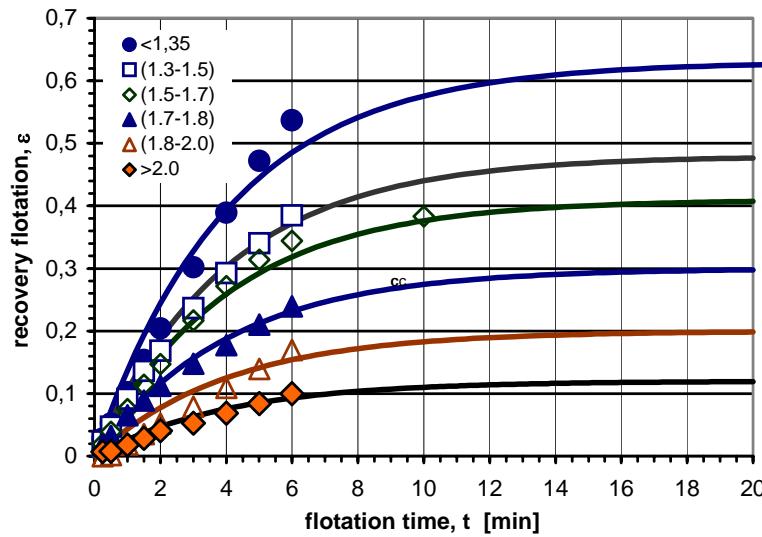


Fig.1. Flotation kinetics:

density fraction <1.35 [Mg/m³], $A=1.06\%$, $\varepsilon=0.63(1-e^{-0.245t})$, $\lambda_0=0.154$ [1/min],
 density fraction (1.35-1.5) [Mg/m³], $A=7.91\%$, $\varepsilon=0.48(1-e^{-0.249t})$, $\lambda_0=0.12$ [1/min],
 density fraction (1.5-1.7) [Mg/m³], $A=23.18\%$, $\varepsilon=0.41(1-e^{-0.25t})$, $\lambda_0=0.102$ [1/min],
 density fraction (1.7-1.8) [Mg/m³], $A=36.32\%$, $\varepsilon=0.30(1-e^{-0.246t})$, $\lambda_0=0.074$ [1/min],
 density fraction (1.8-2.0) [Mg/m³], $A=48.71\%$, $\varepsilon=0.20(1-e^{-0.245t})$, $\lambda_0=0.079$ [1/min],
 density fraction >2.0 [Mg/m³], $A=77.63\%$, $\varepsilon=0.12(1-e^{-0.249t})$, $\lambda_0=0.03$ [1/min].

This constant, according to formula (16), is proportional to the product $P_c P_a$. The probability of adhesion is expressed by the following formula (Yoon and Mao 1966, Schimmoler et al. 1993).

$$P_a = \exp\left(-\frac{E}{E_k}\right) \quad (25)$$

where: E – size of energy barrier of the particle-bubble interaction; E_k – kinetics energy of the particle, necessary to form the bubble-particle aggregate. Energy barrier E grows with the increase of ash content. Also the particle kinetic energy increases because is proportional to particle density which grows with the increase of ash content. Accordingly, the value of the expression under the exponent may change slightly (i.e. increase) with the growth of ash content. If we take into consideration the fact that with the growth of particle density also the probability of collision increases (Nguyen Van and Kmet 1992), then the product of both probabilities may remain constant. On the other hand, the constant of permanent adhesion decreases with the growth of ash content.

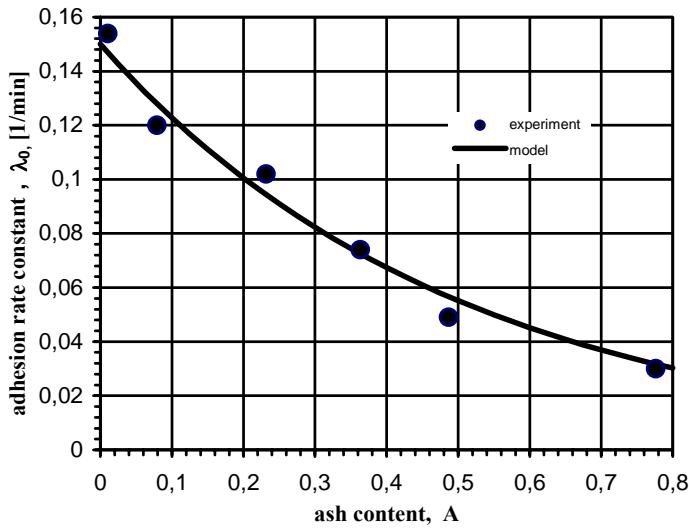


Fig. 2. The permanent adhesion rate constant as a function of ash content in the sample of particle size distribution (0.2-0.315) mm; $\lambda_0 = 0.15 e^{-2A}$

Figure 2 presents the dependence of permanent adhesion rate constant on ash content in the sample under flotation. The following empirical model was proposed:

$$\lambda_o(A) = a e^{-b A} \quad (26)$$

where: a and b – empirical constants while $a = \lambda_o(0)$ is the permanent adhesion rate constant for the ash free coal. It will depend on the coal type, its petrological composition, surface oxidation rate, particle size, etc. and physicochemical and dynamic conditions in the flotation chamber. The constant b , with the above factors, will depend first of all on the composition of coal mineral matter. For the sample tested here, the dependence $\lambda_o(A)$ is expressed by the formula:

$$\lambda_o(A) = 0.15 e^{-2A} \quad (27)$$

The index of curvilinear correlation is 0.99990 which proves good compatibility of the proposed model dependence with experimental values.

CONCLUSIONS

The deterministic model of kinetics of batch flotation of coal of the 1st order with 2 parameters (Eq. 4) does not comprise quantitatively all sub-processes present in bubbles mineralization. The experiment showed that the resultant flotation rate constant in this model is practically independent from the ash content. This fact can be explained by means of the stochastic model of birth and death. The flotation rate constant in the stochastic model achieves the interpretation of the resultant adhesion rate constant which is the sum of the permanent adhesion rate constant and detachment rate constant. With the growth of ash content the permanent adhesion rate constant decreases and the detachment rate constant increases so that their sum remains constant. It can be assumed that all particles of the non-zero rate of exposition of the coal substance on the particle surface are subjected to adhesion to bubbles. Yet, on the surface of particles of low ash content, i.e. high content of the coal substance, the three-phase contact is formed with a large perimeter and it will be, consequently, a permanent connection. At a low content of coal substance and a low rate of exposition, this connection will be less permanent because of the smaller perimeter of the three-phase contact. Therefore the process of particles detachment will be more intense.

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Flotacja jako proces losowy, w którym zmienna losowa przedstawiająca liczbę ziaren wynoszących do warstwy piany do określonego czasu t zależy od czasu, jest procesem stochastycznym. Na skutek turbulencji ośrodka w komorze flotacyjnej oprócz procesu adhezji ziaren do pęcherzyków zachodzi proces odwrotny o intensywności zależnej od zawartości popiołu, czyli proces odrywania ziaren od pęcherzyków. Tego typu sytuację najpełniej opisuje stochastyczny model procesu narodzin i ginięcia. W artykule przedstawiono pokrótkie założenia oraz równanie różniczkowe modelu jak również jego rozwiązanie w postaci równania kinetyki flotacji. Podana została interpretacja parametrów równania. Na podstawie empirycznych zależności uzysku ziaren flotujących w produkcie pianowym od czasu dla kilku próbek węgla, różniących się zawartością popiołu, wyliczono parametry równania kinetyki: stałą prędkość adhezji wypadkowej oraz stałe prędkości adhezji trwałej. Stała prędkość adhezji wypadkowej będąca sumą stałych prędkości adhezji trwałej i prędkości odrywania jest niezależna od zawartości popiołu, natomiast stała prędkość adhezji trwałej maleje ze wzrostem zawartości popiołu.