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THE METHOD OF DENSITY DETERMINATION OF POST-COAGULATION SEDIMENT WITH THE USE OF X-RAY MICROTOMOGRAPHY

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In this paper computed tomography was applied to determine the density of sediments from the water treatment process. Coagulation process caused the destabilisation of the dispersion, and particles had different shapes, sizes and structures. The density of particles had an impact on the settling velocity and is very difficult to be described mathematically. This article presents the method of X-ray microtomography (XMT) application for dried and pressed sediment. The sample volume was obtained through X-ray and image processing. The correct determination of the sample volume depends on a correct number of thresholds. The image processing was performed with the use of the LabVIEW environment and the Vision module. The paper presents the results of tests and boundary parameters for the pressing and image process.
Keywords: coagulation, density determination, X-ray tomography.

В цій статті комп'ютерна томографія була застосована для визначення густини осадів в процесі очищення води. Процес коагуляції викликав дестабілізацію дисперсії, частинки мали різну форму, розміри та структуру. Густина частинок мала вплив на швидкість осідання, цей процес дуже складно описати математично. Ця стаття презентує методику застосування рентгенівської мікротомографії для висушеного та спресованого осаду. Об'ємний взірець був досліджений рентгенівськими променями, після чого було проведено обробку зображення. Коректне визначення параметрів взірця залежить від правильного числа порогів. Обробка зображення була виконана з використанням середовища LabVIEW та модуля Vision. В статті представлено результати випробувань та граничні параметри процесу стиснення та зображення.
Ключові слова: коагуляція, визначення густини, рентгенівська томографія.

В представленной статье компьютерная томогафия была применена для определения плотности осадков в процессе очистки воды. Процесс коагуляции вызвал дестабилизацию дисперсии, частицы имели разныые формы, размеры и структуры. Плотность ч астиц влияла на скорость оседания, этот процесс очень сложно описать математически. Данная статья представляет методику применения рентгеновской микротомографии для высушенного и спрессованного осадка. Объёмный образец был исследован рентгеновскими лучами с последующей обработкой изображения. Корректное определения параметров образца зависит от правильного числа порогов. Обработка изображения была проведена с использованием прогаммного продукта LabVIEW и модуля Vision. В статье представлены результаты испытаний и предельные параметры процес сов сжатия и изображения.
Ключевые слова: коагуляция, определение плотности, рентгеновская томография.

Introduction. The coagulation process is commonly used in water and wastewater treatment processes. The demand for high-quality drinking water is increasing because the number of uncontaminated water sources decreases. Furthermore, the criteria for the discharge of wastewater are becoming stricter to prevent the environment contamination and/or the infection of drinking water sources. Water becomes a potential risk to public health if it is polluted. Thus, the treatment applied to the collected water must ensure that it is free of pathogens and chemicals posing a risk to health when it is to be distributed by the water supply system. What is more, physico-chemical parameters must meet the drinking water standards required by the laws of each country [1,2]. Coagulation/flocculation is considered the most important process in surface water treatment.

The type of coagulant used has a significant impact on coagulation processes. Aluminum-based coagulants have been more widely used than ferric-based coagulants, probably because aluminum coagulants have a superior NOM (Natural Organic Matter) removal capacity, wherease ferric coagulants create water colour problems [3]. However, a large amount of alum salts has also been surspected to be harmful to people and other living organisms [4]. Consequently, an increasing attention has been placed on the identification of novel coagulants to fulfil the increasing demands for organic matter removal and a lower coagulant amount. Organic polymeric coagulants, such as polyacrylamide (PAM), have been used to improve water purification for several decades [5]. Flocs formed in this process settle in a very difficult way. A porous structure and various shapes have an effect on the settling way and velocity. The theoretical description of the sedimentation of floc suspension is only an approximate way of describing this phenomenon.

Coagulation may be carried out as a volume, contact and surface process. This is achieved in a mixing chamber and mixing time is 1-3 minutes. The second phase of coagulation is flocculation, which involves the occurrence of the phenomena leading to the formation of agglomerates and hydrolysis products of the coagulant dissociated by removing impurities. The volume coagulation is carried out in slow-stirred chambers (flocculation), but the contact coagulation is carried out in a layer of solid matter [6].

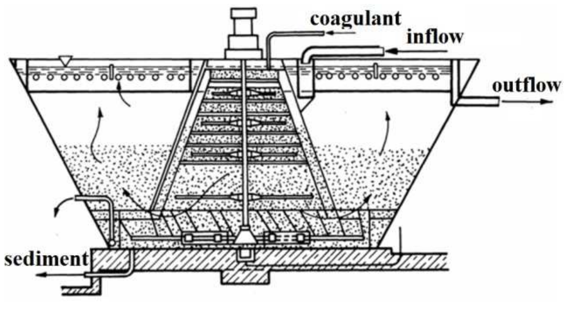


Figure 1 – . Reactor clarifier "Precipitator" [3]

A correct application of coagulation and flocculation processes and the selection of coagulants depend on the understanding of the interaction between these factors [7]. Flocs can be efficiently removed by the process of sedimentation/flotation and filtration. Contact settlers (Fig.1) with a suspended sediment are used to treat water through coagulation and are usually called clarifiers.

Classification of flocs. The shape and kind of floc has a major impact on the duration of the sedimentation process. The classification of flocs formed from the post-coagulation sediment was carried out for the purpose of the research. Due to technological reasons, the settling time of the sediment is the most important parameter. Therefore, the classification proposed by the authors mainly focuses on the separation of groups of flocs according to their characteristic features affecting the settling velocity

While examining the images, the authors decided to classify individual particles statistically. Flocs were divided into three characteristic groups. The classification takes into account the structure of a floc in terms of mass concentration (presence of macropores) and its shape. Flocs were divided into: compact particles, particles having a distinct mass concentration nucleus, around which sediment crystallization networks are formed (flocs without mass concentration), and particles having a porous sponge structure (porous conglomerates).

The classification does not take into account all characteristics. In the future, it will be used for equations describing the settling process. According to it, the most important parameters of particles were only considered. The occurrence and the size of pores in a single floc were assumed to be the criterion of division.

Based on the flocs analysis, there were three separate types of particles characterised by a different settling way (Fig.2a). Compact particles characterised a homogeneous structure. Their shape is similar to a sphere filled with the same microporous structure. Micropores are specific features of a post-coagulation material. Particles of this type fall in a straight line with a slight zigzag motion. Due to their small size and mass, their movement is repeatedly disrupted by fluid movement triggered by the movement of other particles, considerably higher in size, in their vicinity. The second group of particles (star-shaped particles) has a distinct mass concentration nucleus, around which sediment crystallisation networks are formed (flocs without mass concentration) (Fig.2b). Protruding arms cause the particle to move in a circular motion around its own axis while falling. The circular motion causes the particle, whose direction is constantly changing, to be influenced by the Magnus effect. This effect causes the particles of this type to fall in a spiral motion. Over time, star-shaped particles connect to create bigger agglomerates. In case of big agglomerates, the circular motion around their own axis gradually disappears, and particles of this type fall in a straight line. They are characterised by considerably big macropores, through which fluid may flow during their motion. This type of structure is similar to fluid flow through a porous medium [Fig.2c].

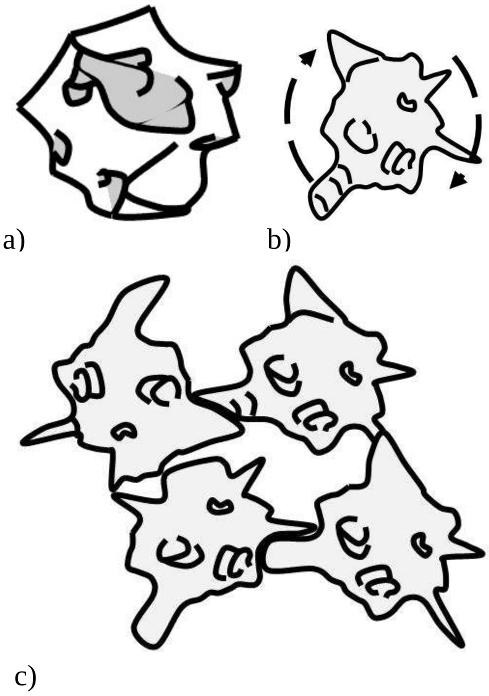


Figure 2 – Types of floc structure a) complex flocs b) star-shaped c) agglomerates

The above-mentioned classification was made on the basis of the microscope observations of the coagulation process. Due to the fact that the diversity of particles is very high, their classification to particular groups occurs on the basis of the isolation of dominating characteristic features.

Mathematical description of the sedimentation process. Due to the fact that coagulation and sedimentation proces are important in the water treatment process, it is necessary to describe these processes by means of mathematical equations [8]

This description may take place only when the definition of basis forces acting on each particle is possible. Based on the presented division of particles into three types, the formation of different mathematical models for them is necessary. The speed of particles descent is defined as the balance of forces acting on a moving particle.

It is possible to formulate equations for compact particles that describe the descent process on the basis of Stokes equations. The basic distribution of forces acting on the descending particle is shown in Fig.3. Two basic forces have an effect on the particle speed: gravity force always in the downward direction F_g and drag force opposite to the direction of motion F_v.

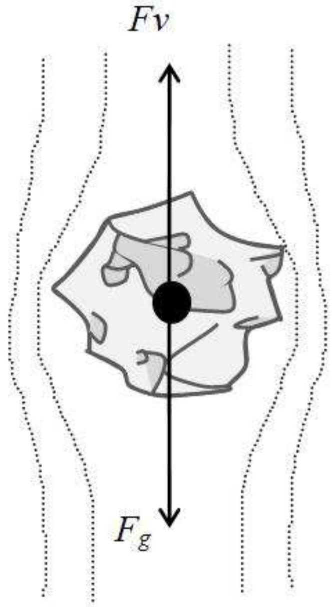


Figure 3 – Types of floc structure a) complex flocs b) star-shaped c) agglomerates

Gravity force Fg, is defined according to Archimedes' principle:

$$F_g = (\rho_s - \rho_l) g V \ , \quad (1)$$

where: g – gravitational acceleration ; V – particle volumeз; ρ_s, ρ_l – density of solid or liquid respectively.

Drag depends on the properties of the fluid and on the size, shape, and speed of the object. General formula for drag force F_v is as follows:

$$F_v = C_D A \rho_l \frac{v|v|}{2} , \quad (2)$$

where: C_D – drag coefficient; A – cross-sectional area in the horizontal plane; v_l – speed of descent.

The shape of moving particles has a direct impact on their velocity of descending. The determination of the correct value of drag coefficient D is difficult due to its complexity. In the literature many empirical dependencies are applied that allow for the determination of the drag coefficient for non-spherical particles [9,10,11]

In case of star-shaped particles, the particle in motion rotates around its own axis. It causes lateral force to occur, which changes the direction of the particle descent. Due to the fact that the shape of the particle is irregular, the velocity at which the particle rotates around its own axis is not constant. It brings the change in the lateral force value, which, consequently, causes the particle to fall in an irregular motion, which is similar to a spiral in shape. Due to the influence of lateral force, the mathematical model should take into account not only lateral force, but also the fact that the sense of drag force is always opposite to the direction of the motion. Since the particle descent does not take place in a straight line, both the sense and value of drag force will change and the descent velocity will be influenced by the horizontal component of this force (Fig.4).

$$F_v^{`} = C_D A \rho_l \frac{v|v|}{2} \cos\varphi , \qquad (3)$$

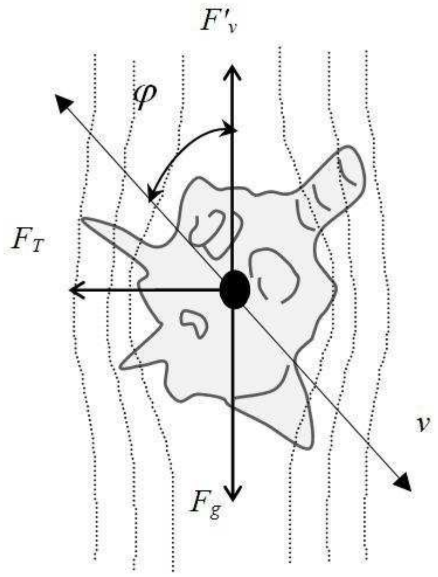


Figure 4 – Forces acting on a star-shaped particle

Rotating around its own axis, the particle induces fluid movement, which induces lateral force FT. The value of this force may be defined based on the Eцtvцs number. It describes a classic model of a stress-induced transverse rising force [12,13]:

$$F_T = -C_T \rho_C \left(\overline{v_\infty} - \overline{v_C}\right) \times rot\ \overline{v_C} , \qquad (4)$$

Coefficient C_T depends on the Reynolds number Re and a modified Eцtvцs number Eod, which is defined as:

$$Eo_d = \frac{g(\rho_s - \rho_l) d_M^2}{\sigma_l} , \qquad (5)$$

where: d_M – maximum particle diameter in a horizontal section.

Agglomerates are formed when star-shaped flocs are connected. As a result of this process, cavities are created, which are called macropores. A network of macropores forms canals, through which liquid can flow during sedimentation. It allows for the stabilisation of the falling particles, which fall in a straight line. In this case, a similar model to the one for compact particles may be used. Gravity and drag force will have a dominant impact on the velocity of the descent. Since the liquid not only flows around the agglomerate but also through macropore canals, drag force should be defined in a different method.

Liquid flow through macropore canals is well described in models for a porous medium. Drag force for porous materials is described by the Darcy-Weisbach equation [16]:

$$Q = \frac{\Delta p}{R} , \qquad (6)$$

where: R – total drag of the porous baffle; Δp – total d liquid pressure difference on both sides of the baffle.

Based on this formula, another formula for the porous material was derived, which is called the Leva equation:

$$\Delta p = \lambda \cdot \frac{h}{d_z} \cdot \frac{(1-\varepsilon)^{3-n}}{\varepsilon^3} \cdot \frac{\rho_s \cdot w^2}{2} , \qquad (7)$$

where: λ – drag coefficient; w – liquid flow velocity through a porous material; d_z – equivalent diameter of the material; ε – porosity of the material.

The value of porosity and n-coefficient should be determined experimentally. To analyse the equations that describe the velocity of falling particles, one of the most basic parameters is density of the material, from which the particle was created. The main subject of this paper is to determine this parameter experimentally. The fact that post-coagulation sediment contains all types of particles poses a difficulty in the density determination of the material. Such a sediment contains a great number of macropores and there is fluid in the closed micropores. Therefore, the density determination is no marginal matter, especially because the microporous structure cannot be destroyed. This article presents the method for the density determination of the post-coagulation sediment with the use of XMT in order to determine the volume of a sample.

B Method for determining the sediment density. This paper presents the description of the measurement method used to determine the density of the post-coagulation sediment using X-ray microtomography. The method involved the determination of the density by means of the gravimetric method. For this purpose, the post-coagulation precipitate was pressed into cylindrical samples with a diameter of 2.5 mm. The samples were weighed on laboratory scales with a measurement accuracy of 0.0004g. The volume of the sample was determined on the basis of the reconstructed three-dimensional images from XMT. The density was determined according to the following formula:

$$\rho = \frac{m}{V} \qquad (8)$$

where: ρ – floks density; m – mass; V – floks volume.

The dried post-coagulation sediment was subjected to the pressing process on the press (Fig. 5). The press is comprised of a cylindrical sleeve in which there are two squeeze rollers, between which the material is pressed. The pressing process was carried out by lifting a 1 kg hammer to a constant height of 5 cm, then dropping it. The compression was adjusted by means of the number of strokes of the hammer.

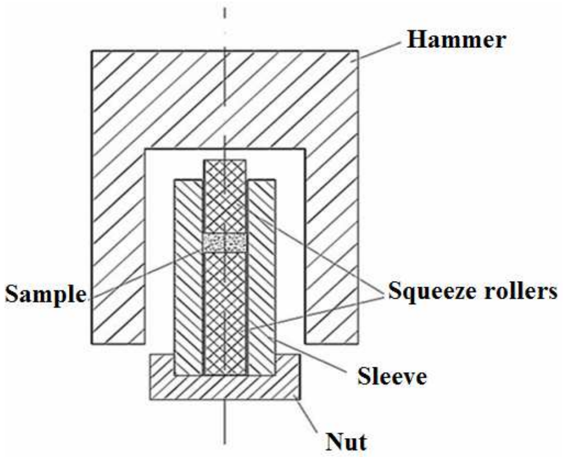
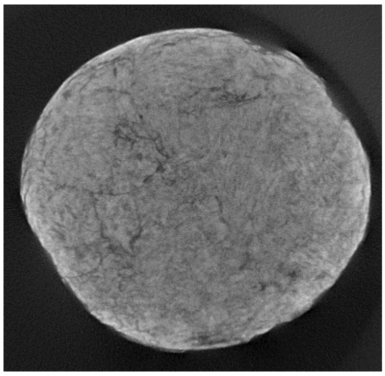


Figure 5 – Scheme of the press impact

The settling of the post-coagulation sediment is disturbed. Water-filled spaces (micro and macropores) render it difficult to determine the volume and the density of particles. Each particle of the sediment contains micropores which are a characteristic feature of every material, and macropores which should not be taken into account in the determination of the density. Due to the above-mentioned reasons, the determination of the density for the post-coagulation sediment presents difficulties [7].

During the sedimentation process, flocs form larger agglomerates, which is called flocculation. The density of the sediment is similar to the density of water, which is the result of a very high mass fraction of water in the sediment usually amounting to 95-99%. The gravity force is closely related to the density of settling particles ρs and liquid density ρl. This parameter is important for the sedimentation process. Therefore, an attempt was made to form the density pattern of post-coagulation sediment particles based on the study carried out using X-ray tomography.

a) 

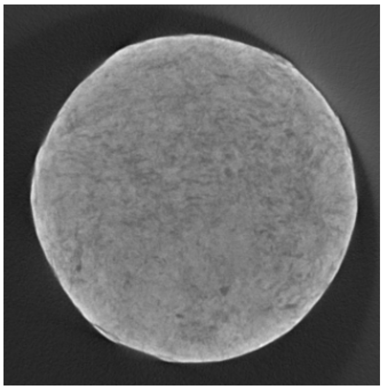
b) 

Figure 6 – Cross-section of the sample for a) 1 stroke of the press, b) 8 strokes of the press

Sediment structures formed during this process are diverse in terms of their shapes and degree of density. Taking the above into consideration, it can be observed that the modelling of this process is particularly difficult. There is a possibility of grouping flocs, taking their characteristics into account, and then forming general equations describing the movement of particles belonging to a given group. However, it is required to know the density of the material of which a floc is composed in order to properly describe the settling velocity of particles. This material has a closed microporous structure, which is a characteristic feature of the given sediment.

A series of cross-sections of the samples obtained due to the X-ray images was studied. The representative images are shown in Figure 6. The images from the scanner are shown in grayscale, where light areas represent the precipitate, and the dark area is the air in the macropores of the sample. After one stroke of the press (Fig. 6a), there were a large number of macropores in the sample, whereas after eight strokes (Fig. 6b), the number of macropores significantly decreased. However, the obtainment of a single structure will lead to the destruction of the microporous structure, which is a characteristic parameter of solids

The volume of the sample was determined by means of the sum of the volume of stored solids obtained from the cross-section images with a thickness of one pixel. In order to determine which pixel represents the sediment, the image processing was carried out. The application reads the selected folder of images. Pixels representing the sediment were isolated for subsequent scans.

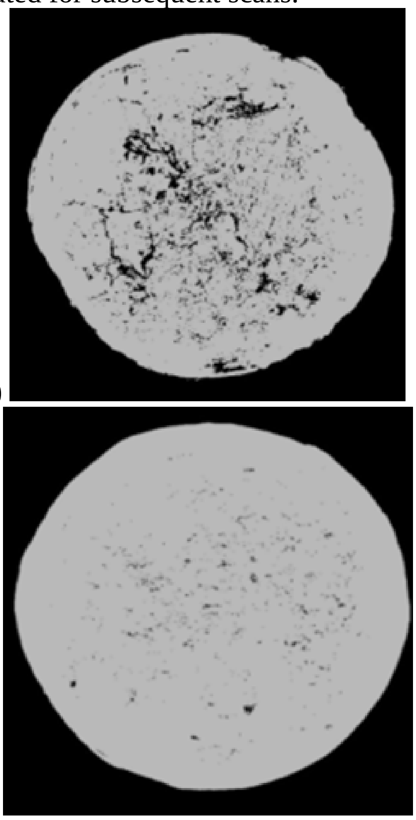
a) b)


Figure 7 – Cross-section of the sample after threshol for a) 1 stroke of the press, b) 8 strokes of the press

The first step was to determine the threshold of the gray level. In the next step, the artifacts were removed, and the number of pixels representing the sediment was determined. The volume of a single section was calculated (one pixel of thickness) on the basis of this scale. Then, the density was calculated for each sample [9]. In case of the sample subjected to only one stroke of the press, macropores are visible, which results in the incorrect determination of the volume (Fig. 7a). In case of eight strokes, the size and the number of macropores were significantly reduced, but it was not possible to eliminate them completely (Fig. 7b). The determination of the volume of a small number of macropores can be corrected, and measurement errors are not significant. However, the proper threshold in the thresholding of gray shades has to be chosen.

The volume of the sample was determined by adding the volume of the solid component obtained from the images of cross-sections with a thickness of one pixel (Fig. 8) [5]. In order to determine which pixel represents settlements, the image analysis process was carried out.

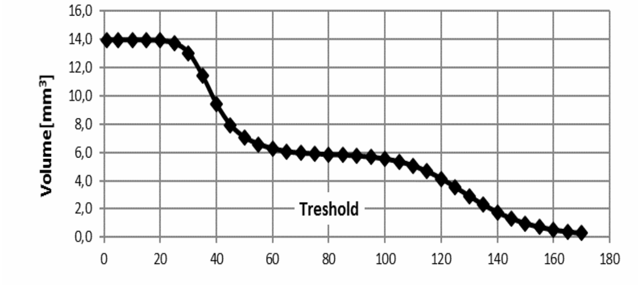


Figure 8 – Relation between the sample volume and the threshold amount

Test results. Figure 9 shows the results of densities calculated for different numbers of strokes of the hammer. Six and more strokes of the hammer provided a relatively small number of macropores in the sample without destroying its microporous structure.

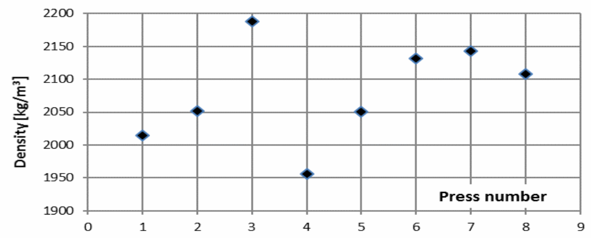


Figure 9 – Relation between the sediment density and the number of strokes of the hammer

Measurement uncertainty was estimated for the developed method of determining the density of post-coagulation sediment. The value of uncertainty was estimated for the sample after 6 strokes of the hammer. For the selected sample, 10 measurements of mass and 9 measurements of volume were performed. Then, the density for all mutual combinations was determined; as a result, 90 values were obtained. The average values for these results were calculated, and they are as follows: V_s=5.875012 mm^3, m_s=012498 g, ρ_s =2129.779 kg/m3. Based on formula (2), the value of sensitivity coefficients was computed.

$$G_m = \frac{\partial \rho}{\partial m} = -\frac{V}{m^2} = -37.61 \qquad (9)$$

$$G_V = \frac{\partial \rho}{\partial V} = \frac{1}{m} = 80010{,}97 \qquad (10)$$

The standard uncertainty of the density measurement is:

$$U_{s\rho} = \sqrt{\frac{\sum_{n=1}^{N} (\rho_i - \bar{\rho})^2}{(N-1)N}} = 77{,}3 \quad \text{kg/m}^3 \quad (11)$$

Limiting instrument errors are mm3 for the volume measurement and mg for the mass measurement respectively. On this basis, the expanded uncertainty was estimated.

$$U_\rho = \sqrt{U_{s\rho}^2 + G_m^2 \frac{D_m^2}{3} + G_V^2 \frac{D_V^2}{3}} = 77{,}302 \ \text{kg/m}^3$$

The values of the uncertainties for selected confidence intervals are listed in Table 1.

Table 1 – The values of the uncertainties for selected confidence intervals

Confidence interval [%]	68.27	90	95	99
Uncertainty [kg/m^3]	77.7	128.3	153.4	202.99

Summary. X-ray microtomography is often applied in materials science to observe material contrasts due to different densities. X-ray data have been applied to virtually every geological discipline, but new applications are being continually discovered. Bone densitometry is the most sensitive screening tool to detect osteoporosis. Another example may be X-ray imaging techniques to determine the density of food, which is a physical property used to assess the quality of food. X-ray microtomography is also a practical solution in order to determine the density of an unknown material as a homogeneous polymer sample [14,15,16]. In this paper XMT was applied to determine the density of sediments from the water treatment process. The coagulation process caused the destabilisation of the dispersion, and agglomeration of the particles that should be removed. The density of particles has an impact on the rate of descent. Drying and pressing processes may destroy the microstructure of a sediment. The sample volume was obtained through X-ray and image processing. The correct determination of the sample volume depends on a correct number of thresholds. The density determination error is below 4%, which, in case of porous materials, is satisfactory. The obtained results of the measurements of density uncertainty affirm a high accuracy of mass and volume measurements of the samples tested.

1. Bergamasco R, Konradt-Moraes L. C, Vieira M. F, Fagundes-Klen M. R, Salcedo Vieira A. M 2011 Performance of a coagulation–ultrafiltration hybrid process for water supply treatment. Chemical Engineering Journal 2 483. 2. Zemmouri H, Drouiche M, Sayeh A, Lounici H, Mameri N 2012 Coagulation Flocculation Test of Keddara's Water Dam Using Chitosan and Sulfate Aluminium Procedia Engineering 33 254–260. 3. Kowal L. A, Њwiderska-Bryї M 1997 Water Treatment PWN 115-117. 4. Orzechowski Z., Prywer J., Zarzycki R.: 2009 Fluid mechanics in protection and environmental engineering, Wydawnictwo Naukowo-Techniczne, Warsaw 5. Xu Y, Chen T, Ciu F, Shi W2016 Effect of reused alum-humic-flocs on coagulation performance and floc characteristics formed by aluminum salt coagulants in humic-acid water Chemical Engineering Journal 287 225–232. 6. – C. 38-40. 6. Tzoupanos N.D, Zouboulis A.J 2008 Coagulation-flocculation processes in water/waste water treatment: The application of new generation of chemical reagents, 6th IASME/WSEAS International Conference on Heat Transfer, Thermal Engineering And Environment (HTE'08) Rhodes, Greece, August 20-22. 7.] Saritha V, Srinivas N, Srikanth Vuppala N. V 2015Analysis and optimization of coagulation and flocculation process. Appl. Water Sci. 8. Omelia C 1998 Coagulation and sedimentation in lakes, reservoirs and water treatment plants Water Science and Technology 37/129. 10. Ganser G. H 1993 A rational approach to drag prediction of spherical and non-spherical particles Powder Technology 77 143-152. 11. Haider A, Levenspiel O 1989 Drag coefficient and terminal velocity of spherical and nonspherical particles Powder Technology 58 63-70. 12. Tomiyama A 1998 Struggle With Computational Bubble Dynamics, Third International Conference on Multiphase Flow, ICMF`98, Lyon France June 8-12. 13. Ives K. J. 1961 New concepts in filtration. Water and Water Engineering 8. 14. Kelkar S, Boushey C.J, Okos M 2015a method to determine the density of food using X-ray imaging J. Food Eng., 159 36-41. 15. Elgelhardt L. T 2010 Coagulation, Flocculation and Clarification of Drinking Water, Engelhardt 5-6. 16. Rz№sa R. M, Podgyrni E 2014 Investigation of the effects of salinity and temperature on the removal of iron from water by aeration, filtration and coagulation Pol. J. Environ. Stud. 6 2157-2151.

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