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Research paper

Analysis of the possibility of using concrete waste in the groundwater remediation

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Abstract: The annual intensive exploitation of natural resources results in the generation of substantial amounts of waste, with construction being the primary contributor. This sector is accountable for nearly half of all non-renewable resources consumed by humanity and for producing over one million construction and demolition (C&D) wastes annually. An alternative approach to managing construction and demolition wastes involves recycling and reclaiming materials on-site for environmental remediation purposes. The research aimed to explore the potential use of concrete waste as a reactive material for groundwater remediation contaminated with heavy metals (Cu, Zn) and chlorides. Concrete C&D waste was gathered from demolition sites in Warsaw, crushed to the appropriate size, and subjected to preliminary tests to assess its physical and chemical properties, including granulometric analysis, absorbability, specific surface area, and sorption capacity. Additionally, surface modification of selected concrete wastes was performed to enhance their ability to retain contaminants, alongside batch tests (kinetic and chemical equilibrium reaction) using both raw and modified materials. The kinetic studies indicated that the pseudo-second order model best fit the test results (determination coefficient R^2 value in the range of 0.72–0.99), while the chemical equilibrium studies revealed that the Langmuir, Freundlich, and Redlich–Peterson models were suitable (R^2 in the range of 0.70–0.90). The findings demonstrated that C&D waste holds promise as an adsorbent material for environmental protection and remediation purposes.

Keywords: batch test, construction and demolition waste, environmental contamination, groundwater remediation

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1. Introduction

The rapid expansion of the construction industry has led to increased material consumption, mainly due to the extensive use of natural resources. This sector accounts for roughly half of all non-renewable resources consumed by humanity and substantial amounts of waste generated during construction, refurbishment, and demolition activities [1]. Despite the implementation of new regulations and recommendations, the majority of this waste still ends up in landfills. In the USA, the European Union, and China, approximately 924 million tonnes, 534 million tonnes, and 2.3 billion tonnes of construction and demolition waste are produced, respectively. Finding an alternative approach to managing this waste presents a substantial challenge. One potential alternative method entails the on-site recycling and recovery of materials. However, the attainment of a 70% recycling and recovery rate for non-hazardous construction and demolition waste, as mandated for EU Member States by 2020, has only been achieved by a limited number of countries, with recycling rates for this waste stream hovering around 50% [2, 3]. It is widely acknowledged that finding an alternative approach to managing this waste is crucial. Consequently, another advocated alternative method for the treatment of C&D waste involves its utilization as an adsorbent material for environmental remediation e.g. in permeable reactive barriers (PRB) technology. It is widely acknowledged that finding an alternative approach to managing waste is crucial. Adsorption is considered to be one of the most effective pollution control technologies for treating heavy metals due to its cost-effectiveness, ease of operation, and high efficiency [4]. Additionally, adsorbents can be regenerated through a desorption process in some cases, as adsorption is often a reversible reaction [5].

As a result, there is a growing research interest in evaluating the adsorption potential of various waste materials, such as C&D wastes, for heavy metal treatment [6, 7]. This pollution has escalated due to the increased use of chemical fertilizers, industrial activities, wastewater treatment plants, and natural processes causing groundwater contamination [8, 9]. Additionally, stormwater runoff is a another significant source of pollution in this environment, containing heavy metals such as cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), and zinc (Zn), as well as deicing salts like NaCl [10]. Chloride (Cl–), which does not readily participate in oxidation-reduction reactions, form complexes under most groundwater conditions, or adhere to mineral surfaces, is highly mobile once it enters groundwater [11–13]. Furthermore, numerous studies have documented the release of heavy metals from natural geological deposits and soils due to road salt application [12, 14]. Given this, it is justifiable to explore approaches for the concurrent removal of heavy metals and chlorides.

Several studies have employed concrete for the removal of heavy metal ions from aqueous solutions [15–17]. Cement compounds (e.g. tricalcium silicate, dicalcium silicate, tricalcium aluminate and tetra-calcium ferrite) are generally highly heterogeneous, which helps in removing metals from aqueous solutions through precipitation and adsorption [6, 7, 18]. Thus, it is preferable to carry out this process in a stepwise manner to enable distinct characterization of the two major phases involved: the precipitation phase and the simultaneous adsorption–precipitation phase [19]. The issue with cement materials is the potential for corrosion due to contact with chloride [20]. The solution to this problem could be to use MgO. Several studies have shown that the relative fraction of MgO is responsible for the increase in chloride, as well as in heavy metal removal efficiency [21–23].

This study stands out by utilizing construction and demolition (C&D) waste from demolition sites as a cost-effective and efficient material for groundwater remediation. The waste is modified with MgO to enhance its ability to remove a mixture of contaminants through multiple mechanisms. The C&D waste was collected from demolition sites in Warsaw, crushed to the appropriate size, and then subjected to tests to assess its physical and chemical properties, including granulometric analysis, absorbability, specific surface area, and sorption capacity. The main tests focused on evaluating the kinetic and chemical equilibrium reactions using both raw and modified materials. The primary objective of this work was to validate the feasibility of using C&D materials as a low-cost and readily available medium for removing copper, zinc, and chloride from water.

2. Materials and methods

2.1. Materials

In this study, the following C&D waste was used as research material: concrete paving stones (PS), concrete paving slab (PSl), railroad sleeper (RS), concrete slab (CS) and a mixture of the above (M). The samples were taken from various places in Warsaw, Poland where concrete elements were dismantled. The decision to use a mixture of waste was dictated by the difficulty of identifying each type of material in a typical sample of construction debris. For comparison purposes, concrete paving stones were used as a relatively easy to identify material. By identifying the material, it is possible to determine its manufacturer and determine the composition of the concrete mixture. Taking into account the possibility of using these wastes as materials for groundwater remediation, they must be not only easily accessible and inexpensive but above all, neutral for the environment. Therefore, knowing the composition and source of waste is an extremely important issue in this case.

The material for testing was initially prepared by crushing to an appropriate fraction (max. 2 mm) using crushers: diaphragm jaw and impact crushers to obtain the appropriate granulometry and then sieved into 0–2 mm particles. Tests on the physical and chemical properties of the materials were performed for each of the above materials separately. On the other hand, modification and kinetic and chemical equilibrium tests were carried out on concrete paving stones (PS) and a mixture of all C&D waste (M). The prepared materials are presented in Fig. 1.

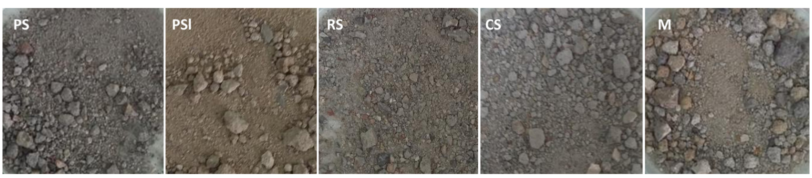

Fig. 1. The samples of C&W waste materials

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2.2. Aqueous solution

Multi-component model solutions were used in the laboratory tests carried out as part of the work. The components of the model solutions were the following heavy metals: Cu, Zn, and chlorides. The following p.a. salts were used to prepare the solutions used in batch tests, adding their appropriate amounts to distilled water:

– sodium chloride NaCl, CHEMPUR, Poland,
– copper (II) chloride 2 hydrate $CuCl_2 \times 2H_2O$, CHEMPUR, Poland,
– zinc chloride $ZnCl_2$, CHEMPUR, Poland.

2.3. Tests of physical and chemical properties

The laboratory testing of the basic physical and chemical properties of the selected materials was aimed at determining the granulometric composition by the sieve method according to BN-65 6728-01 [24] and the classification of the materials according to standards: PN-88/B-04481 [25], PN-86/B-02480 [26] and ASTM D422-6 [27], the saturated surface dry (*SSD*) according to PN-B-06714/18:1977 [28], the sorption capacity (*MBC*) and the specific surface area (S_t) according to PN-88/B-04481 [25] and PN-B-06714/18:1977 [28]. The reason for treating concrete waste according to soil standards was the potential location of the material in the soil medium.

The first stage was the determination of the particle size distribution of fine and coarse aggregates by sieving on a laboratory shaker. The results of the tests are presented in the form of graphs of granulometric curves. In the next stage of the study, a saturation test was carried out, which involved weighing out 10 g of each material (previously dried at 120°C for 24 h) and spreading them on a Petri dish so that the grains of the materials evenly covered its entire surface. Then, using an automatic pipette, portions (0.1 ml) of distilled water were applied until all pores of the materials were saturated. The saturated surface dry (*SSD*) was calculated using the following formula:

$$SSD = \left(\frac{M_{SSD} - M_{DRY}}{M_{DRY}}\right) \cdot 100\% \tag{2.1}$$

where: M_{SSD} and M_{DRY} – dry and saturated sample mass respectively [g].

The specific surface area (S_t) tests were carried out using the methylene blue adsorption method. This method is based on the assumption that the total specific surface area, which is equal to the sum of the surface projections of the individual methylene blue particles that have been adsorbed by the materials in the form of a single-particle layer, was related to the weight of the dried soil equal to 1 g dry weight. The specific surface area is expressed as the product of the sorption capacity and the *k*-factor, equal to 20.94 m^2/g. The sorption capacity and the total surface area are calculated using the following equations (Eq. 2.2, 2.3):

$$St = MBC \cdot k \tag{2.2}$$

$$MBC = \frac{100 \cdot m}{2 \cdot m_{\text{dry}}} \left(V_i + V_{i-1}\right) \tag{2.3}$$

where: k – a coefficient whose value is equal to 20.94 [m^2/g], m – mass of methylene blue contained in 1 cm^3 of solution, calculated as 3 – water [g], m_{dry} – mass of the material used for the determination, converted to a substance dried at 105–110°C [g], V_i – volume of the solution at which the sorption capacity was exceeded [cm^3], V_{i-1} – volume of the solution corresponding to the penultimate portion of methylene blue solution before the sorption capacity was exceeded [cm^3].

2.4. Modification of C&D waste materials

The process of modifying the concrete waste involved dry impregnation, also known as first-moisture impregnation, to increase the adsorption capacity of the materials being analyzed. This method entailed directly applying active ingredients, such as MgO, by filling the material's pores with a salt solution of appropriate concentration. To achieve the desired percentage of active metal in the sample, it was necessary to prepare a salt solution of suitable concentration. Before introducing the modifying solution, the carrier's sorption capacity was determined. After conducting the absorbent capacity test, solutions were prepared for materials PS and M, each with a mass of 100 g. In the case of material PS, 7.5 g of MgO was added to a 36 ml solution, while for material M, a 7.5 g balance was added to a 35 ml volume. The samples were then dried in dry air for 2 hours at 120°C and subsequently roasted in a muffle furnace at 400°C for 4 hours. Figure 2 shows a modified material.

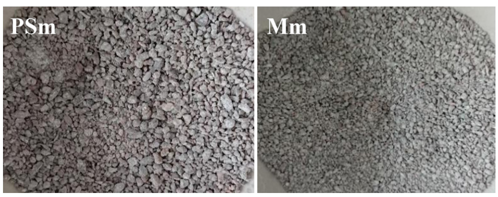

Fig. 2. The samples of modified C&D waste materials – modified concrete paving stones PSm and modified mixture of C&D waste materials Mm

2.5. Batch test – kinetic studies

In the present study, the following reaction kinetics models were used to analyze the mechanism controlling the adsorption process in selected materials: pseudo-first-order, pseudo-second-order, Elovich, and the intramolecular diffusion model in the Weber–Morris interpretation.

The most commonly used equations for determining the kinetics of adsorption reactions are the pseudo-first-order and pseudo-second-order Lagergren models. Lagergren's pseudo-first-order model is expressed by the equation [30]:

$$\frac{dq(t)}{dt} = k_1 (q_e - q_t) \tag{2.4}$$

where: q_t and q_e – are the amount of adsorbate adsorbed at any given time, t and at equilibrium, respectively, [mg/g], k_1 – the rate constant of pseudo-first model reaction [min^{-1}].

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Integrating the Eq. 2.4 for boundary conditions $q(t) = 0$, $t = 0$, $q(t) = q(t)$, $t = t$ takes the form:

$$\ln\left(\frac{q_e}{q_e - q(t)}\right) = k_1 t \tag{2.5}$$

Equation after diverted can be expressed as follows:

$$\log\left(q_e - q_{(t)}\right) = \log\left(q_e\right) - \left(\frac{k_1 \cdot t}{2.303}\right) \tag{2.6}$$

In cases when $q_e \ll k_1$ is not satisfied, equation (1) can be transformed to the pseudo-second-order rate expression, [31]:

$$\frac{\mathrm{d}q(t)}{\mathrm{d}t} = k_2\left(q_e - q_{(t)}\right)^2 \tag{2.7}$$

where: k_2 is the rate constant of pseudo-second model reaction [g/mg min], which after integration for boundary conditions $q(t) = 0$, $t = 0$, will result to:

$$\frac{t}{q_e} = \frac{1}{\left(k_2 q_e^2\right)} + \frac{t}{q_e} \tag{2.8}$$

The Elovich or Roginsky–Zeldovich kinetics model is expressed by the following equation [32]:

$$\frac{\mathrm{d}q(t)}{\mathrm{d}t} = \alpha \exp(-\beta_{q(t)}) \tag{2.9}$$

where: α – initial sorption rate [mg/g min], β – desorption constant [g/mg].

To simplify the Eq. 2.6, Chien & Clayton [33] proposed the following form, assuming that $\alpha\beta t \geq 1$ and boundary condition: $q(t = 0) = 0$ and $q(t = t) = q(t)$, $(\mathrm{d}q(t))/\mathrm{d}t = \alpha \exp(-\beta_{q(t)})$

$$q_{(t)} = \beta \ln(\alpha\beta) + \beta \ln(t) \tag{2.10}$$

Weber–Morris intraparticle diffusion model assumed, that the adsorption of dissolved ions changes more often in proportion to the contact time $t^{1/2}$ than to t. The model is expressed by a linear equation [34, 35]:

$$q_{(t)} = K_{\mathrm{int}} t^{1/2} \tag{2.11}$$

where: K_{int} – intraparticle diffusion rate constant (mg/g min$^{1/2}$), t – contact time [min$^{1/2}$].

The removal ratio R (%) of contaminants by C&W materials was calculated using the Eq. (2.12):

$$R = \frac{(C_0 - C)\,100}{C_0} \tag{2.12}$$

where: C_0 and C are the initial and final metal concentrations [mg/L].

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The kinetic tests were carried out using 1 g of selected C&W materials (PS, M, and PSm, Mm). These experiments were performed in stoppered falcon flasks by mixing 50 ml of aqueous solutions with a constant concentration of contaminants Cu and Zn – 5 mg/L and Cl – 150 mg/L with a known amount of materials. Each experiment was conducted by preparing repeated samples, double-blind (distilled water with materials), and two control samples (aqueous solutions without materials). Samples were shaken at room temperature of 21 ± 2°C and were collected at suitable time intervals of 1, 3, 6, 28, and 58 hours. The residual metal concentration in the filtrate was measured using Atomic Adsorption Spectrometer ICP – AAS (Thermo Scientific, USA) according to the PN-EN ISO 8288:2002 standard [36].

For the determination of a final chloride concentration in solution, the Mohr method (PN-ISO 9297: 1994) [37] was conducted. Moreover, at the beginning and the end of experiments, pH, temperature, and electrical conductivity were measured using a digital meter (SCHOTT Instruments GmbH, Germany).

2.6. The chemical equilibrium of reactions studies

The study of the chemical equilibrium of reactions occurring during contact of contaminants with raw and modified concrete materials formed the second stage of static testing and enabled analysis of the ability of materials to retain contaminants. To analyze the adsorption process taking place, mathematical models were applied to the analyzed materials, which differ in the physical parameters of the studied process. Interpretations of the results of the study were carried out using as many as four models of L-type or H-type adsorption isotherms: Langmuir, Freudlich, Redlich–Peterson, and Toth.

The Langmuir model states that on an energetically homogeneous surface with limited sorption capacity, adsorbate molecules can be adsorbed in the form of a so-called monolayer (a single molecule occupies one active site, i.e. no multilayer formation is possible) [38]. This model is expressed by the following equation:

$$q_e = \frac{C_{a,\max} K_L C_e}{1 + K_L C_e} \tag{2.13}$$

where: q_e – the amount of metal adsorbed per gram of the adsorbent at equilibrium (mg/g), C_e – the equilibrium concentration of adsorbate (mg/L^{-1}), $C_{a,\max}$ – maximum monolayer coverage capacity (mg/g), K_L – Langmuir isotherm constant (L/mg).

The Langmuir model relies on the dimensionless coefficient R_L to determine the effectiveness of adsorption in removing ions from solutions. This parameter can be calculated using the formula provided by Weber and Chakravorti [39]:

$$R_L = \frac{1}{1 + K_L C_e} \tag{2.14}$$

The dimensionless coefficient takes four values [40]: (1) $0 < R_L < 1$, indicating intensive and promoted adsorption, (2) $R_L > 1$, where adsorption contribution is negligible, (3) $R_L = 1$, representing linear adsorption, and (4) $R_L = 0$, indicating irreversible adsorption.

The Freundlich isotherm model is a one-parameter empirical expression that accurately describes the adsorption of solutes in solution on energetically heterogeneous surfaces, without limiting the adsorption by a monolayer [41]. Based upon these assumptions, Freundlich represented the following equation:

$$q_e = K_F C_e^{1/n} \tag{2.15}$$

where: q_e – the amount of metal adsorbed per gram of the adsorbent at equilibrium (mg/g), K_F – Freundlich isotherm constant (mg/g), n – adsorption intensity, C_e – the equilibrium concentration of adsorbate (mg/L).

The Redlich–Peterson isotherm model, developed by Redlich and Peterson in 1959, is an empirical equation with three parameters. This model combines aspects of the Langmuir and Freundlich models and suggests that the adsorption process occurs through a combination of these mechanisms. The equation can be expressed in linear form as shown by [42]. The model is given by the following equation:

$$q_e = \frac{K_R C_e}{1 + b_R C_e^{\beta}} \tag{2.16}$$

where: q_e – the amount of metal adsorbed per gram of the adsorbent at equilibrium (mg/g), K_R, b_R – the Redlich–Peterson isotherm constants (mg/g), β – a power exponent, denoted by a value ranging from 0 to 1, represents a fractional power, C_e – the equilibrium concentration of adsorbate (mg/L).

In the case where the value of the power is equal to β, the Redlich–Peterson isotherm equation goes into the Langmuir model according to the equation:

$$q_e = \frac{K_R C_e}{1 + b_R C_e} \tag{2.17}$$

If the value of the power is equal to β, the Redlich–Peterson isotherm equation reads like an expression according to Henry's law:

$$q_e = \frac{K_R C_e}{1 + b_R} \tag{2.18}$$

The Toth model's isotherm, developed by Toth in 1971, is an empirical 3-parameter equation widely used to describe heterogeneous adsorption processes for both low and high ion concentrations. This equation is expressed in convolution, as demonstrated by [43]. This model is expressed by the following equation:

$$q_e = \frac{K_T C_e}{\left(b_T C_e^{T_a}\right)^{1/T_a}} \tag{2.19}$$

where: q_e – the amount of metal adsorbed per gram of the adsorbent at equilibrium (mg/g), K_T, b_T, T_a – the Toth isotherm constants (mg/g), C_e – the equilibrium concentration of adsorbate (mg/L).

In addition, as in the case of the heavy metal ion and chloride kinetics studies, contaminant retention rates R [%] were determined during contact between materials and contaminants.

The chemical equilibrium of reactions studies were carried out using 1 g of selected C&W materials (PS, M, and mPS, mM) and placed into 50ml flasks. The experiments were conducted in duplicate for each material, including double-blind (distilled water with materials) and two control samples (aqueous solutions without materials). The materials were then inundated with the following aqueous solutions (50 mL): NaCl at concentrations of 5, 15, 25, 75, and 150 mg/l, $ZuCl_2$ and $CuCl_2$ at concentrations of 0.5, 1, 2, 5, and 10 mg/L. All tests were conducted at room temperature (21 ± 2°C). Before testing, the pH and conductivity of the samples were measured using a SCHOTT multi-parameter meter from Germany. The samples were then agitated on a GFL rotary shaker from Germany for 24 hours. After this period, the samples were filtered and the pH, conductivity, temperature, chloride (by Mohr's method), and heavy metals Cu and Zn were determined in the filtrates using an Atomic Adsorption Spectrometer ICP – AAS (Thermo Scientific, USA).

3. Results and discussion

A granulometric analysis of the chosen C&D waste materials classified the materials as medium gravel (MGr), with only the material mixture being classified as fine gravel (FGr) according to ASTM D422-63 [27] and PN-EN ISO14688 [30]. Based on this classification, the materials were identified as even-grained ($1 \le C_u \le 5$), well-grained (C_c = 1–3), and monofractured ($C_u < 6$) soils following PN-EN ISO 14688-1:2018-05. Additionally, the tested C&D materials exhibited similar absorbability. Material CS showed the lowest absorbability, while material PS exhibited the highest. In terms of sorption capacity (*MBC*) and specific surface area (S_t), material CS had the highest values, while PSl, RS, and M showed the lowest values. The results of the tests discussed are presented in Table 1.

Table 1. The physical and chemical properties of selected materials

	C_u [–]	C_c [–]	*SSD* [%]	*MBC* [g/100g]	S_t [$m^2 \cdot g^{-1}$]
PS	4.947	2.316	39.16	0.61	12.78
PSl	3.647	1.299	37.10	0.39	8.14
RS	2.282	2.308	37.10	0.39	8.14
CS	2.316	3.520	27.34	0.72	15.11
M	1.676	1.001	33.00	0.39	8.14

The results of the kinetics study were best represented by a pseudo-second-order kinetics model, particularly in relation to the retention of all impurity ions on PS. This was supported by the range of reaction kinetics constants k, which varied from 0.0007–0.0142. The avarage coefficients of determination R^2 were determined 2 to be 0.96 for PS, 0.88 for M, 0.97 for PSm, and 0.98 for Mm. Detailed kinetic test results can be found in Fig. 3 and Table 2. Several studies [45–47] have confirmed that the kinetics adsorption data of C&D waste-based adsorbent for contaminant removal are well-fitted and adhere to pseudo-second order kinetics, suggesting a possible mechanism of chemisorption. Furthermore, the pseudo-second order model implies that the adsorption rate is determined by both the rate of diffusion of the adsorbate and the rate of the chemical reaction between the adsorbate and adsorbent, involving valence forces through the sharing or exchanging of electrons, which represents the rate-limiting step.

Table 2. The physical and chemical properties of selected materials

Pseudo second-order kinetic equation	**Cu**	**Zn**	**Cl**
		PS	
k_2 [g/mg min]	0.0138	0.0107	0.0082
Determination coefficient R^2 [–]	0.92	0.97	0.99
		M	
k_2 [g/mg min]	0.0113	0.0107	0.0007
Determination coefficient R^2 [–]	0.96	0.96	0.72
		PSm	
k_2 [g/mg min]	0.0138	0.0137	0.0040
Determination coefficient R^2 [–]	0.97	0.96	0.99
		Mm	
k_2 [g/mg min]	0.0142	0.0134	0.0052
Determination coefficient R^2 [–]	0.97	0.97	0.99

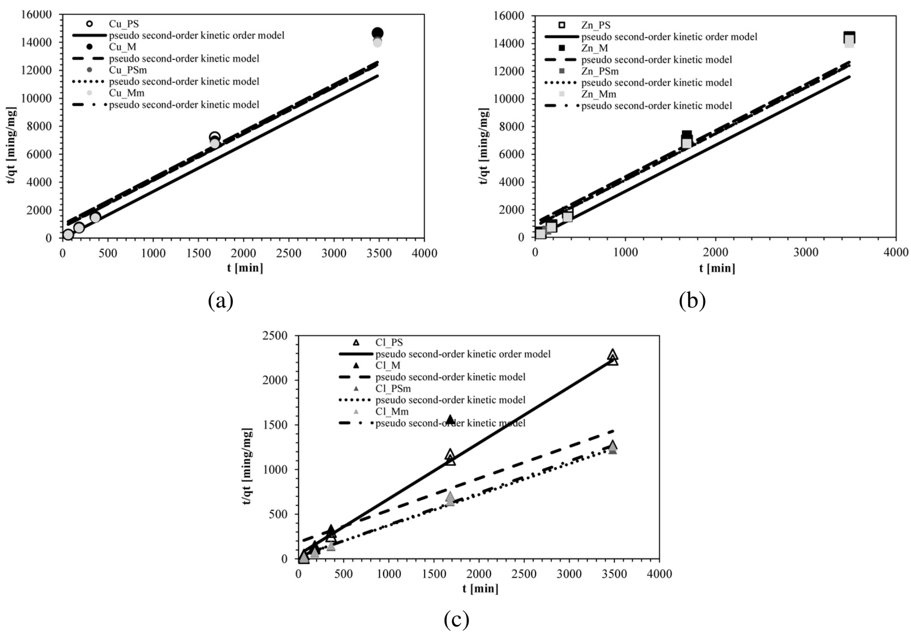

Fig. 3. Adsorption kinetic of: (a) Cu; (b) Zn; (c) Cl on PS, M, PSm and Mm

The retention factor R [%] for contaminant reduction was observed throughout the entire test for the solution components. Graphs of metal retention during contact with individual materials are shown in Fig. 4. On average, Cu showed a reduction of 93.1–99.8%. Zn showed a reduction of 73.1–99.4%, and Cl showed a reduction of 14.32–67%. The materials PSm and Mm exhibited the highest contaminant reductions. For Cu ion retention, these materials showed reductions of 99.1% and 99.7%, for Zn ion retention: 98.9% and 99.1%, and for Cl ions: 39.6% and 42.9%. Cu ion retention was most intense for PS and PSm materials at hour 3, and for M and Mm materials at hour 6. Zn ion retention was most intense at the end of the experiment for unmodified materials, while for modified materials the highest process intensity was recorded at 3 hours. The reduction of Cl ions in both PSm and Mm materials, as well as in PS material. had the highest values at the beginning (1 hour) of the experiment, while the highest reduction for M material occurred at the end of the test. The Cu and Zn removal efficiencies for unmodified materials (PS and M) improved significantly with increasing contact time (Fig. 4). Thus, the sorption rate was rapid in the initial phase, followed by a gradual decrease [49]. The slower sorption rate can be attributed to the reduction in the number of sorption sites on the reactive material. However, high removal efficiencies for modified materials were observed throughout the period of kinetic studies. This was related to other removal processes based on precipitation.

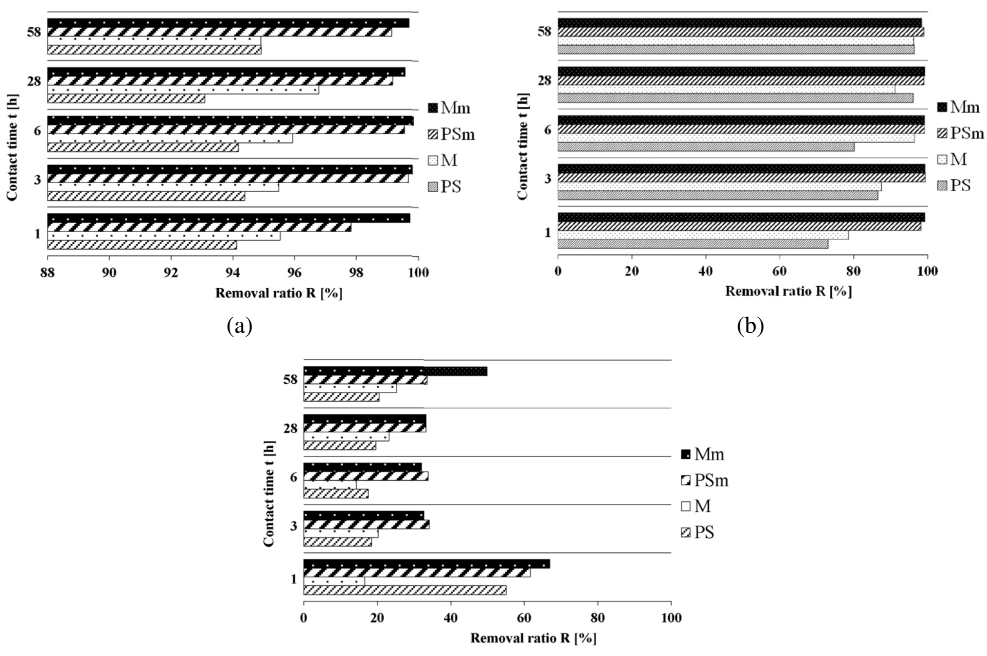

Fig. 4. The Cu (a), Zn (b) and Cl (c) removal ratio R (%) from aqueous solutions for different contact time t with PS, M, PSm, and Mm

The temperature of the solution was constant at 20°C during the experiment. The electrical conductivity of the solution with the mixture of pollutants was 0.333 mS, while in the 3rd hour. it was 0.622–0.925 mS, and in the 58th hour: 1.803–2.563 mS. The pH of the analyzed solution with the mixture of pollutants before the tests was approximately 5.44, while at the end of the experiment, the pH increased to approximately 10. Figure 5 shows the dynamics of pH and EC changes depending on the contact time of the pollutant with the material. The highest pH was recorded in the case of contact with the following materials: PSm (pH = 10.55) and Mm (pH =10.81), while the lowest in the case of PS (pH = 9.17). A high pH value in the final solutions, in addition to intensive sorption processes, may also indicate the occurrence of other contaminant removal processes, such as precipitation and co-precipitation or dissolution of minerals from the concrete materials used.

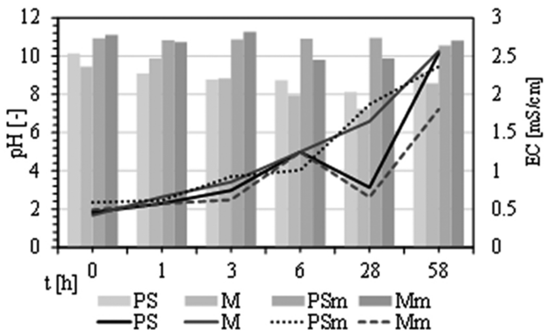

Fig. 5. The pH and EC changes during kinetic studies

The experimental data concerning the influence of the initial concentration of multiple contaminants in a test medium solution were analyzed using SOLVER (Microsoft, Excel 2019) to fit isotherm models. Graphical representations of these models are depicted in Fig. 6. A higher R^2 value closer to 1 signifies a better fit of the respective equation to the experimental data. The experimental and theoretical data did not match well with the current model likely because of the high heterogeneity of the C&D waste material, as confirmed by several studies [49, 50]. However, the experimental data are represented by non-linear curves in all model equations, with R^2 values of at least 0.70 as outlined in Fig. 6. The heavy metal determinations obtained results that were described by several isotherm models, including Langmuir, Freundlich, Henry, Redlich–Peterson, and Toth. However, it was impossible to fit the results obtained with the chemical equilibrium model for PSm material. Future studies will need to consider other models, such as anti-Langmuir or pseudo-Langmuir, in order to accurately interpret the results. The Langmuir model provided the best fit for the Zn ion in impurity mix in material M, with a coefficient of determination R^2 of 0.93. Material PS showed the best fit for the Zn ion with the Redlich–Peterson model, with an R^2 of 0.91. The Henry model showed a high coefficient of determination for Cu ions during contact with M, at 0.91. As for the Redlich–Peterson and Toth models, the highest coefficients of determination were for ions Cu in M, at 0.92 and 0.91, respectively. Figure 6 illustrates example results of sorption isotherms for the individual models. The experimental data for Mm were well fitted to the

Freundlich and Henry models for Zn, obtaining R^2 values of 0.70 and 0.77, respectively. Due to the impossibility of fitting the experimental results to adsorption isotherm models, the results of the analysis for the presence of chloride ions in the filtrate are presented as the contaminant removal ratio R [%].

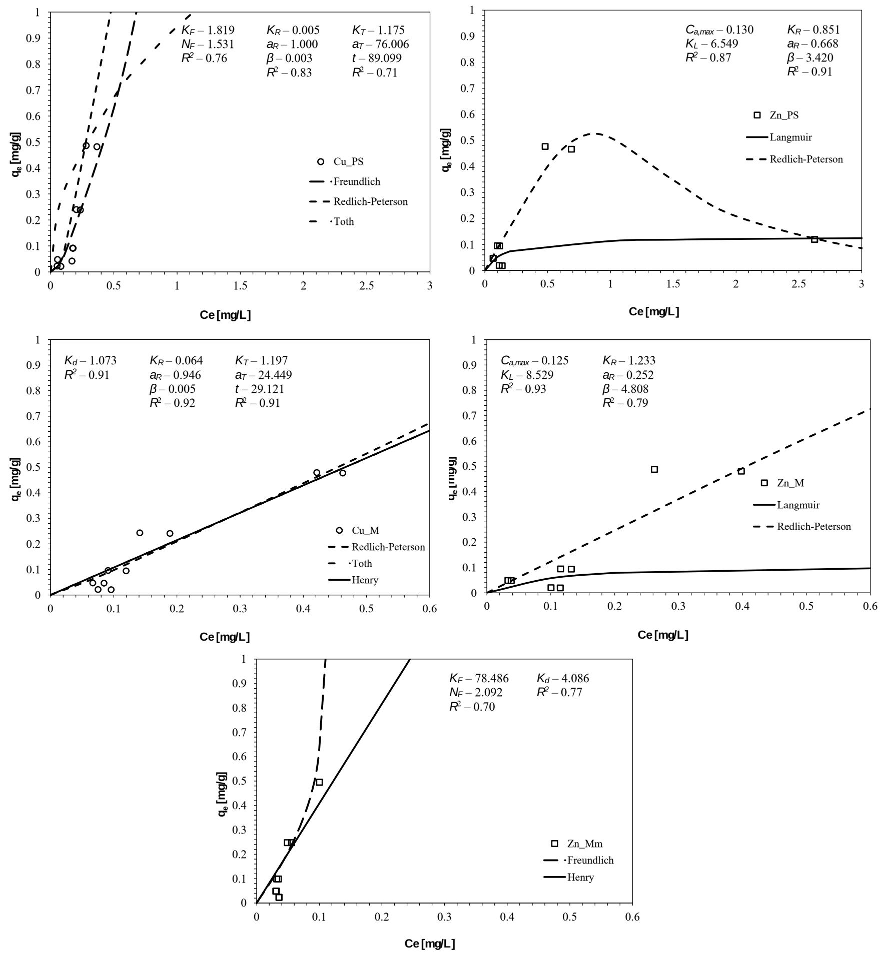

Fig. 6. The adsorption isotherm for PS, M, and Mm

The PSm and Mm materials showed the most significant average reduction in all contaminants (Fig. 7). For copper (Cu) in the contaminant aqueous solution, both PSm and Mm achieved an average reduction of 98.6%, while the reduction of zinc (Zn) was 97.2 and 97.1%, respectively. For unmodified materials, the reduction of Cu and Zn was also significant, with PS at 91.8% and 80.8% and M at 92.9% and 82.6% respectively. The average chloride removal from the solution for raw materials was low, with a rate of 12.1% for PS and 28.3% for M. However, the removal was moderate for modified materials, with a rate of 50.7% for PSm and

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46.2% for Mm. The test results indicated a higher removal percentage of chloride at lower initial concentrations and for the highest initial concentrations for raw C&D materials. Similar observations regarding heavy metal reduction from aqueous solution were made by Ali and Abd Ali [49]. It could be inferred that less-favourable sites became more active with increasing concentration [51, 52].

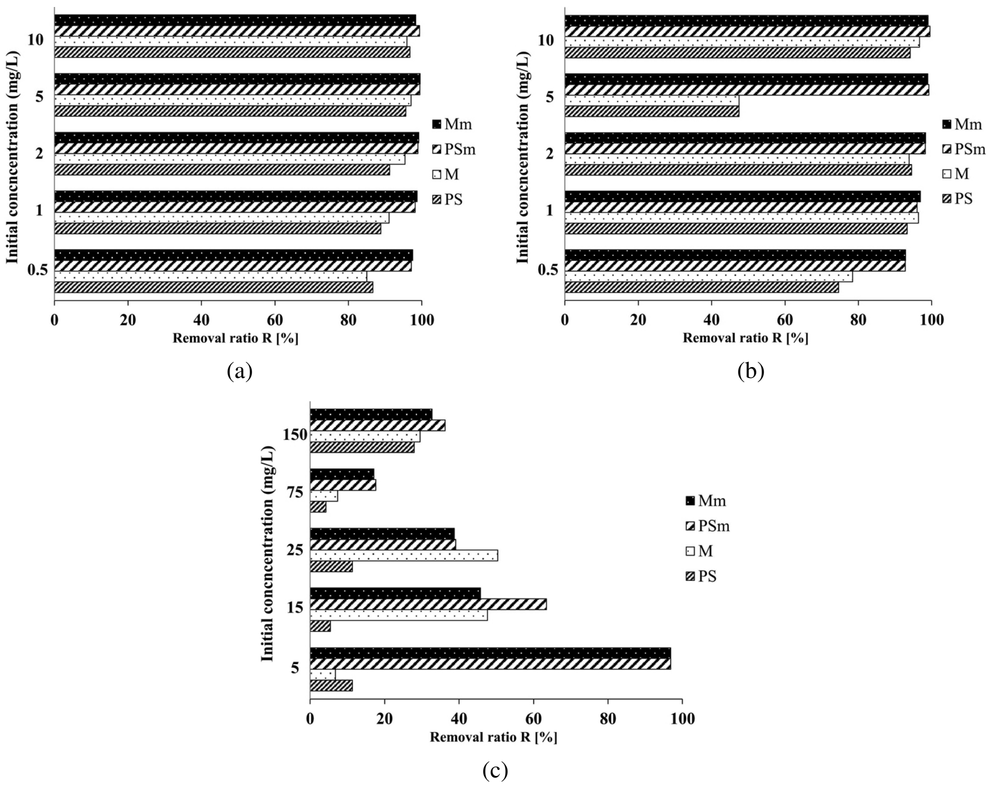

Fig. 7. The Cu (a), Zn (b), and Cl (c) removal ratio *R* (%) from aqueous solutions for different initial concentrations

The temperature of the analyzed solutions remained unchanged during the test and was 20°C. The changes in conductivity and pH are shown in Fig. 8. The conductivity for all analyses was in the range of 0.422 to 2.563 mS. The highest value of pH and The highest conductivity and the greatest increase in pH occurred when PSm material was contacted with a solution contaminated with Cl (C_0 = 150 mg/L) and Cu and Zn (C_0 = 10 mg/L). The pH of the solution at the beginning of the test was 5.98, but after 24 hours of contact with the material, the pH increased to a value of 10.54 and the conductivity was 2.364 mS. The lowest conductivity was observed for a 0.5 ml/L $CuCl_2$ solution and PS material, while the lowest pH increase was observed when a 5 mg/L $CuCl_2$ solution with a pH of 5.92 was contacted with PS material. resulting in a pH of 7.1.

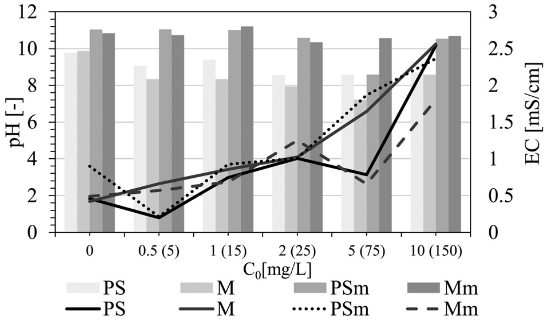

Fig. 8. The pH and EC changes during equilibrium of reactions studies

4. Conclusions

This work aimed to analyze the possibility of using concrete waste for groundwater remediation. To this end, a series of non-fluid 'batch test' kinetic and chemical equilibrium studies were carried out to understand the mechanisms of contaminant retention.

The studies carried out allowed the following conclusions to be drawn:

- The results of the reaction kinetics were described by the best-fit pseudo-second-order model.
- The impurity reduction, expressed by the removal ratio R [%], during the whole kinetic study for the solution components: Cu was on average 93.1–99.8%, for Zn 73.1–99.4% and Cl 14.32–67%; the highest impurity reductions were obtained for materials: PSm and Mm.
- The experimental results have demonstrated that the primary mechanisms involved in removing copper (Cu) and zinc (Zn) from aqueous solutions onto C&D waste materials are sorption and precipitation processes. The sorption data can be effectively represented by the Langmuir, Henry, and Redlich–Peterson models.
- The equilibrium batch investigations have confirmed that C&D waste materials are efficient sorbents and can be used as a reactive medium for remediating groundwater contaminated with heavy metals (copper and zinc) and chlorides.
- The modification has enhanced the materials' capacity to retain contaminants, thereby increasing their overall effectiveness. Under the experimental conditions of this study, the removal efficiency of modified C&D waste materials for copper, zinc, and chloride was found to be 98 %, 97%, and 50%, respectively.
- C&D materials are an efficient method for heavy metal removal.

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Analiza możliwości zastosowania odpadów betonowych w remediacji środowiska gruntowo-wodnego

Słowa kluczowe: odpady budowlane i rozbiórkowe, zanieczyszczenie środowiska, oczyszczanie środowiska, batch test

Streszczenie:

Intensywna eksploatacja zasobów naturalnych każdego roku prowadzi do powstawania ogromnych ilości odpadów. których największym wytwórcą jest budownictwo. Ten sektor gospodarki odpowiada za pozyskanie około połowy wszystkich zużywanych przez ludzkość zasobów nieodnawialnych oraz za wytwarzania ponad miliona odpadów budowlanych i rozbiórkowych (C&D, ang. construction and demolition waste) rocznie. Duże ilości odpadów powstają zarówno podczas budowy nowych inwestycji, jak i podczas renowacji lub likwidacji budynków i budowli. Odpady C&D obejmują następujące materiały: beton, drewno, asfalt, gruz, cegły, metale, gips, szkło, tworzywa sztuczne, odzyskane elementy budynków (drzwi, okna i armatura wodno-kanalizacyjna). Pomimo wprowadzenia nowych przepisów i zaleceń, odpady te w głównej mierze trafiają na składowiska odpadów. W USA, Unii Europejskiej i Chinach masa odpadów C&D wynosi odpowiednio około 924 mln ton. 534 mln ton i 2.3 mld ton. Dlatego też, ogromne wyzwanie stanowi alternatywne podejście do gospodarki tymi odpadami. Zamienną metodą przetwarzania odpadów z budowy i rozbiórki jest recykling i odzysk materiałów w miejscu ich powstania. Rozporządzenie Parlamentu Europejskiego i Rady (UE) nr 305/2011 z dnia 9 marca 2011 r. wprowadziło nowe wytyczne dotyczące zrównoważonego wykorzystania zasobów naturalnych, uwzględniające problematykę recyklingu, wykorzystania materiałów przyjaznych środowisku i pochodzących z recyklingu, jak i trwałości budynków. Nałożony na państwa członkowskie UE obowiązek osiągnięcia do 2020 roku 70% poziomu recyklingu i odzysku odpadów powstałych podczas budowy i demontażu budynków. które nie są uznawane za niebezpieczne spełniło tylko kilka państw, którym udało się osiągnąć około 50% poziomu recyklingu tego strumienia odpadów. Dlatego też, kolejną promowaną metodą alternatywną przetwarzania C&D odpadów jest wykorzystanie ich jako materiału adsorpcyjnego do oczyszczania środowiska naturalnego.W pracy zbadano możliwości wykorzystania odpadów betonowych jako reaktywnego materiału do remediacji środowiska gruntowo-wodnego zanieczyszczonego metalami ciężkimi (Cu, Zn) i chlorkami. W tym celu zebrano z miejsc rozbiórki na terenie m. st. Warszawy betonowe odpady budowlane, które skruszono do odpowiedniej frakcji i poddano badaniom wstępnym w celu określenia ich właściwości fizyko – chemicznych. tj. analizie granulometrycznej, nasiąkliwości, powierzchni właściwej i pojemności sorpcyjnej. Przeprowadzono również modyfikację powierzchni wybranych odpadów betonowych w celu zwiększenia ich zdolności do zatrzymywania zanieczyszczeń oraz badania typu batch test (kinetyczne i równowagi chemicznej reakcji) z wykorzystaniem materiałów surowych i modyfikowanych. W badaniach kinetycznych najlepszym dopasowaniem do wyników badań charakteryzował się model pseudo-drugiego rzędu (wartość współczynnika korelacji R^2 w przedziale 0.72–0.99), a w badaniach równowagi chemicznej model Langmuira, Freundlicha, Henrygo, Redlich–Petersona i Totha (R^2 w zakresie 0.70–0.93). Wyniki badań wykazały, że odpady betonowe mogą potencjalnie stać się materiałem adsorpcyjnym w zabezpieczaniu i oczyszczaniu środowiska naturalnego.

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