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An experimental study on the nitrate removal ability of aggregates used in pervious concrete

Mohammad Javad Mehrani[a], Abolghasem Alighardashi[a,*], Amir Mohammad Ramezanianpour[b]

[a]*Department of Civil, Water and Environmental Engineering, Shahid Beheshti University, Abbas Pour Blvd., P.O. Box 16765-1719, Tehran, Iran, Tel. +98 21 7393 2467; Fax: +98 21 7700 66 60; email: a_ghardashi@sbu.ac.ir (A. Alighardashi), Tel. +98 9138037221; email: mj.mehrani@gmail.com (M.J. Mehrani)*

[b]*Civil Engineering Faculty, Tehran University, Enghelab Square, P.O. Box 14155-613, Tehran, Iran, Tel. +98 21 61113602; email: ramezanian@ut.ac.ir*

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ABSTRACT

In this research, different aggregates used in pervious concrete, namely zeolite, perlite, and pumice were compared in terms of their ability to remove nitrate from water. Also, several other properties of aggregates were evaluated. Regarding water absorption, pumice and perlite aggregates had the lowest (about 10%) and the highest (40%) absorption, respectively. The results showed no reactivity of aggregates in terms of alkali-silica reaction. Permeability tests indicated that the maximum (1.64 cm/s) and the minimum (1.4 cm/s) permeability corresponded to pumice and perlite aggregates, respectively. The required time for reaching equilibrium concentration in the nitrate removal test was 30 min when perlite and pumice were used, while it was 60 min for zeolite. Moreover, when HCl and H_2SO_4 were applied as an activating agent, this time did not decrease significantly for all aggregates. For all three raw aggregates, nitrate absorption process followed Langmuir isotherm and the absorption capacity was about 70 mmol/g. Overall, pumice was selected as the best raw aggregate among other aggregates tested in this study due to its lower water absorption, having no alkali-silica reactivity, highest permeability, and highest nitrate removal ability based on the presence of metal ions.

Keywords: Nitrate removal; Pervious concrete; Pumice; Perlite; Zeolite

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1. Introduction

The concentration of nitrate (NO_3^-) in public water supplies has increased to above acceptable levels in many areas of the world. The extra usage of fertilizers and also contamination by humans and animals' waste are two major reasons for this issue [1]. The water contamination has a long-term effect on the human's health, so the international standards such as WHO prescribe allowable levels of nitrate in drinking water in the order of 50 mg/L [2]. The absorption process is considered as an acceptable way for reducing nitrate concentration due to its simplicity of design, ease of operation, and no need for final purification [3].

Pervious concrete is a special type of concrete containing cement paste and coarse aggregate, which can have a drainage function due to its porosity between 15% and 30% [4]. A lot of research has been carried out on the properties of aggregate and their impacts on the properties of pervious concrete. Rafique Bhutta et al. [5] investigated the properties of porous concrete, such as porosity, permeability, and compressive strength, made with recycled aggregate. Also, in another research conducted by Agar-Ozbek et al. [6], the properties of pervious concrete including tensile strength and adhesion resistance of cement were evaluated. Zhong and Wille [7] carried out a research about the design and

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* Corresponding author.

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specification of high-performance pervious concrete in which the compressive strength and durability were studied without disrupting the concrete hydraulic conditions. They reported that the concrete strength can be increased up to 150 MPa. In addition, the mechanical properties of lightweight aggregate including lime powder and high volume of fly ash have been evaluated by Shafigh et al. [8]. Based on their results, limestone increased the compressive strength of concrete while the fly ash had no significant effect. Liu et al. [9] investigated the effect of aggregates on properties of concrete such as water absorption, permeability, and resistance to chloride ion. Also, Zaetang et al. [10] examined pumice, diatomite, and autoclaved recycled aggregates for usage in pervious concrete. They concluded that increasing the amount of cement improves the mechanical properties of pervious concrete. In addition, concrete containing diatomite had better mechanical properties, but its thermal conductivity was less.

Several studies have been conducted on nitrate removal using aggregate. Baei et al. [11] used perlite activated by H_2SO_4 in a batch system and reported that the highest removal efficiency (91.01%) was obtained at a contact time of 120 min. Also, in another study, raw sepiolite aggregate and its activated type by HCl in the batch system had a removal efficiency of 35% and 100% at the contact time of 30 and 5 min, respectively [12]. As Golestanifar et al. [13] reported the iron-modified pumice had a removal rate of 87.6 mg/g with the contact time of 50 min [13]. Also, Alighardashi et al. [14] investigated the mechanically activated red mud aggregate and observed that it removed 75.11% nitrate in a contact time of 60 min. The magnetic nanoparticle coated on activated powder was used by Rezaei Kalantary et al. [15] for nitrate absorption process with a contact time of 60 min, and maximum monolayer capacity of 57.1 mg/g was achieved. Also, nitrate removal using some materials such as pumice powder was examined by Kim et al. [16], who reported a rate of nitrate absorption of 0.65 mg/g at 20°C for 50 mg/L concentration. The removal efficiency of granular chitosan was investigated by Sowmya and Meenakshi [17], and the absorption capacity of 90.6% was obtained at a contact time of 90 min. They also applied three isotherms, namely Langmuir, Freundlich, and Dubinin–Radushkevich to fit the equilibrium data. In addition, Hu et al. [18] kinetically studied the nitrate absorption using chitosan.

The main purpose of this study was to investigate the nitrate removal of aggregates used in pervious concrete. Perlite, pumice, and zeolite aggregates were chosen due to their extensive local applications as well as high silica content. The water absorption, reactivity, and permeability of aggregate were analyzed. In addition, the nitrate removal efficiency and absorption isotherms were studied with respect to the mechanism of nitrate reduction.

2. Materials and methods

2.1. Aggregate and cement

The aggregates used in this study were zeolite from Iran Mineral (Shahrekord, Tehran), perlite from Permis mine (Isfahan) and pumice from Ghezeljekand mine (Qorveh). The cement used in the reactivity test was a Portland type 2 manufactured by Nain Cement, Isfahan. In all experiments, the same coarse aggregates (3.8 in No. 4) and fine aggregate (No. 6, No. 8) were used. Before all tests, the aggregates were washed with distilled water several times and any additional materials were removed. Fig. 1 shows the shape of aggregates.

The chemical and physical properties of the aggregates are illustrated in Tables 1 and 2, respectively.

2.2. Chemicals

All compounds were purchased from Merck Company, Germany. Hydrogen chloride (HCl) with 37% purity and 0.75 normality, and sulfuric acid (H_2SO_4) with 95% purity and 0.15 normality were used for activation of aggregates. Also, sodium hydroxide powder (NaOH) with 100% purity was used for reactivity test and nitrate sodium ($NaNO_3$) powder with 100% purity was used for making artificial water contaminated with nitrate.

2.3. Equipment and devices

The nitrate measurement was conducted by spectrophotometry method, using a Shimadzu UV Mini 1240 model equipment.

2.4. Testing methods

The test methods for measuring aggregates' water absorption, alkali-silica reactivity, permeability, and nitrate removal ability were carried out according to Table 3.

All nitrate removal experiments were conducted at laboratory temperature (20°C ± 2°C). The rotation rate for perlite, pumice, and zeolite aggregates were 250, 160, and

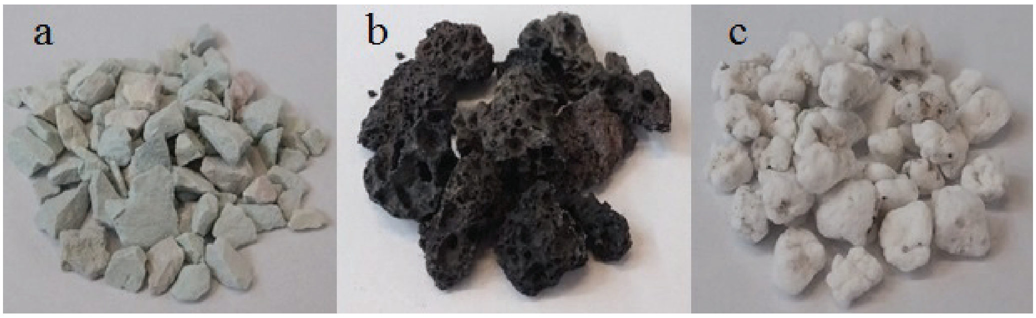

Fig. 1. The aggregates used in this research (a) zeolite, (b) pumice, and (c) perlite.

150 rpm, respectively. In addition, 2.5 g of each aggregate was added in 50 mL solution containing 70 ppm nitrate. The nitrate removal test of aggregates was conducted in compliance with the Langmuir and Freundlich isotherms using different concentrations of nitrate (30–150 ppm).

Table 1
Chemical properties of aggregates and cement

Composition (%)	Portland cement	Zeolite	Perlite	Pumice
SiO_2	20.5	72.8	73.76	72.86
Al_2O_3	5.3	14.11	14	15.8
TiO_2	–	–	0.25	–
CaO	64.5	1.47	1.5	1.56
Fe_2O_3	3.6	1.45	1.8	1.5
MgO	2.8	0.38	0.41	0.48
K_2O	–	4.46	4.35	4.1
SO_3	1.5	1.5	–	–
Na_2O	–	3.83	3.93	3.7
LOI[a]	1.8	–	–	–

[a] The rest of unknown composition.

3. Results and discussion

3.1. The water absorption of aggregate

The purpose of this test was to determine the aggregates moisture content as compared with the saturated surface dry (SSD) state. This variable is a good index to determine the aggregate's resistance to weathering and deterioration of concrete [22]. It should be noted that for samples with high absorption such as perlite, the water absorption should be considered in the mixture and the aggregate should be introduced into the mixture in the SSD state. The results of the test are shown

Table 2
Physical properties of aggregates

Property/aggregate	Perlite	Pumice	Zeolite
Specific gravity (g/cm^3)	0.8–1.2	1.6–2.4	1.8–2.8
Density (g/cm^3)	0.2	0.6–0.8	1–1.2
Hardness	5.5–7	7–8.5	8–11
Appearance	Irregular spherical	Porous	Broken
pH	7–7.02	6.3–6.8	7–7.5

Table 3
Testing activated aggregates and their procedures

Experiment	Test method	Reference
Absorption	The amount of water absorption of aggregates was determined by the weight difference of the aggregates in two states: dried in oven (m_{dry}) for a specified time (1–48 h), and in the saturated surface dry state (submerged in water for a specified time (1–48 h) (m_{ssd})): $$\text{Water absorption }(\%) = \frac{m_{ssd} - m_{dry}}{m_{ssd}} \times 100$$	[19,20]
Alkali-silica reactivity	After making ordinary concrete samples of 25 × 25 × 25 mm in size with the aggregates, cement, and water (with a specific mix design), the samples were submerged in a NaOH solution at 80°C for 16 d, and their length expansion was measured every 4 d	[21]
Permeability	The permeability coefficient (K) was obtained according to the Darcy law for this purpose, the difference in the input water and output source (h_0), and the difference level between the input water and reservoir (h_1) over a specified period of time (t) from the tube containing aggregates with a specified length (l) were measured. ((A) is the area of specimen and a is (a) area of tube): $$K = \frac{al}{At} \ln \frac{h_0}{h_1}$$	[25]
Nitrate removal by aggregates activated with HCl	After washing with distilled water, 10 g of each aggregate was placed in 400 mL of distilled water containing (HCl) in the oven for 2 h at a temperature of 87°C; then, the aggregates were washed with distilled water and dried in oven at 105°C for 2 h	[12]
Nitrate removal by aggregates activated H_2SO_4	After washing with distilled water, 10 g of each aggregate was placed in 100 mL of distilled water containing 0.14 M sulfuric acid at a temperature of 87°C for 2 h; then, the aggregates were washed with distilled water and dried in oven at 110°C for 2 h	[11]

in Fig. 2. According to the results, pumice and perlite had the minimum and maximum water absorption, respectively. In fact, the absorption of perlite was about 6 times that of pumice. The difference between the absorption of pumice and perlite was not significant.

3.2. Alkali-silica reaction

The reaction between alkali in the cement and mineral silica in reactive alkali silicate aggregates occurs in wet conditions [23]. This can lead into destruction of concrete. As such, the reactivity of aggregates in this regard has to be examined. The obtained results for this test are depicted in Fig. 3. As the results suggest, the expansion rate less than 0.1% for all samples after 16 d illustrates no serious reactivity of siliceous aggregates. In other words, all aggregates are suitable to be used in concrete with respect to the potential of alkali-silica reaction. Adding mineral additives such as natural pozzolan and fly ash to cement up to 30%, does not have any impacts on reducing resistance and can prevent reactivity between aggregate and cement [24].

3.3. Permeability

The results of permeability test performed according to the Darcy relationship using the constant hydraulic head under the aquatic conditions indicated that the permeability coefficient (*K*) was 1.64, 1.58, and 1.4 cm/s for pumice, zeolite, and perlite, respectively. According to these results, all aggregates have the same range of permeability, while pumice and perlite had the highest and lowest permeability, respectively.

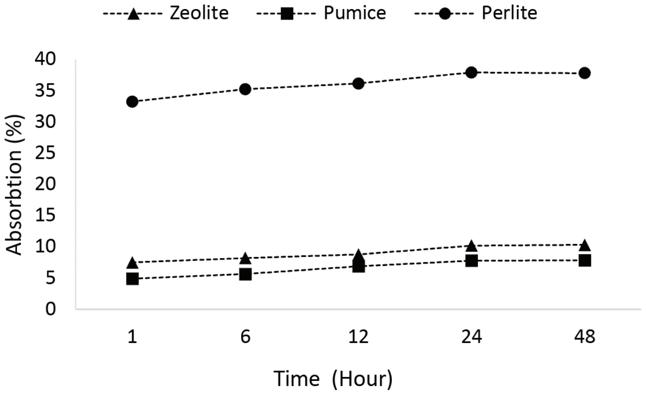

Fig. 2. Water absorption of aggregates at different times.

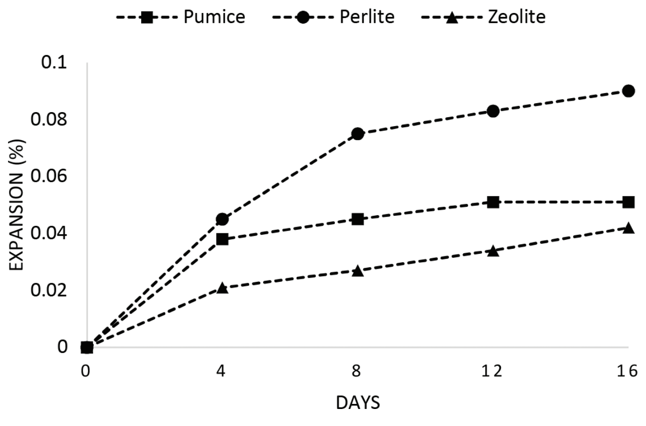

Fig. 3. Results of alkali-silica reaction test (expansion of specimens).

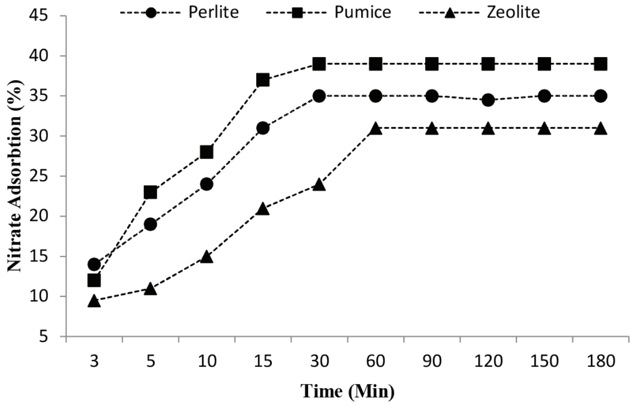

Fig. 4. Nitrate absorption by raw aggregates.

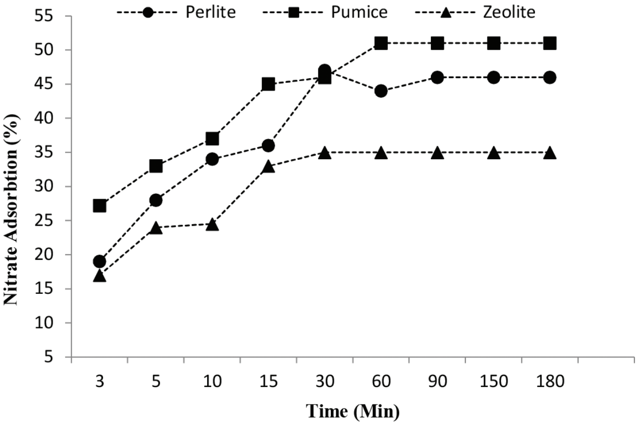

Fig. 5. Nitrate absorption by HCl activated aggregates.

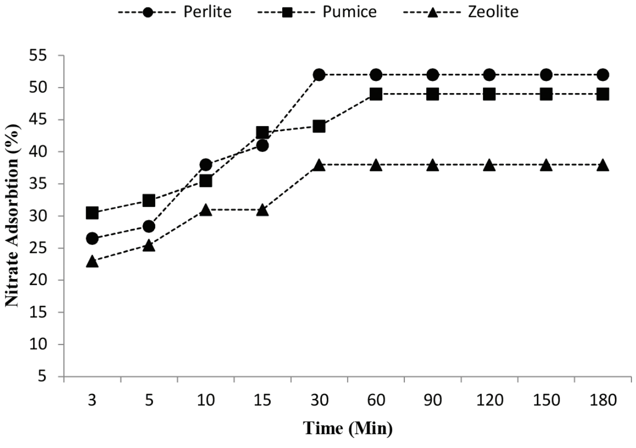

Fig. 6. Nitrate absorption by H_2SO_4 activated aggregates.

3.4. Nitrate removal using aggregate

3.4.1. Removal experiments

Batch experiments were conducted to determine the amounts of nitrate removal using aggregate as an absorbing agent. Figs. 4–6 present the absorption of nitrate at different times with raw aggregates, and aggregates activated with HCl and H_2SO_4, respectively. As shown in Fig. 4, the equilibrium concentration time for all aggregates in the raw state was about 30 min. The nitrate absorption by pumice aggregate (39%) was more than the two other aggregates (35% and 31%, respectively). Also, according to Fig. 5, the equilibrium concentration time for all aggregates in the HCl-activated mode was about 60 min. In this case, pumice had the highest nitrate absorption (50%) among all. Moreover, it can be seen in Fig. 6 that the nitrate absorption by perlite (52%) surpassed the nitrate absorption by pumice (49%) and zeolite. The equilibrium concentration time for all aggregates in H_2SO_4 activated was about 60 min. It should be noted that due to the insignificant impact of activation on aggregates against long time and the complexity of the activation procedure, the nitrate removal and other tests were continued using raw aggregates.

3.4.2. Absorption isotherms

In general, absorption isotherms are used for characterization of the industrial absorption process. In this work, the Langmuir and Freundlich isotherms were applied to describe the relationship between the amount of adsorbed NO_3 ions and its equilibrium concentration, the results of which are presented in Table 4.

3.4.2.1. Langmuir isotherm The main assumption of the Langmuir method is that the absorption uniformly occurs on the active part of the surface. In this regard, when a molecule is adsorbed on a site, it does not have any effect on other incident molecules. The Langmuir equation is as follows:

$$\frac{C_e}{q_e} = \frac{1}{bk_l} + \frac{C_e}{k_l} \tag{1}$$

where q_e and C_e are the amount of solute adsorbed per unit weight of adsorbent (mmol/g) and the equilibrium concentration of solute in the bulk solution (mmol/L), respectively. k_l and b are the Langmuir constants representing the maximum absorption capacity for the energy constant of the absorption heat.

As illustrated in Table 4, the isotherm data fitted the Langmuir equation well (R^2 = 0.99, 0.98, and 0.92 for pumice, perlite, and zeolite, respectively).

3.4.2.2. Freundlich isotherm The Freundlich equation is purely empirical based upon the absorption on the heterogeneous surface and can be written as follows [26]:

$$q_e = kc_e^{\frac{1}{n}} \tag{2}$$

By plotting $\log q_e$ vs. $\log C_e$, the constants of the Freundlich isotherm (k, $1/n$) were calculated. It is observed that the isotherm data fitted the Freundlich model well (R^2 = 0.95, 0.87, and 0.85 for pumice, perlite, and zeolite, respectively). In addition to the correlation coefficient (R^2), chi-square analysis (χ^2), and normalized standard deviation (NSD) were employed to reasonably investigate the validity of various models, defined as follows [27]:

$$\chi^2 = \Sigma \frac{(q_e^{\exp} - q_e^{\text{cal}})^2}{q_e^{\text{cal}}} \tag{3}$$

$$\text{NSD}\ (\%) = 100 \times \sqrt{\sum \frac{[(q_e^{\exp} - q_e^{\text{cal}}) / q_e^{\exp}]^2}{N-1}} \tag{4}$$

where $q_e^{\exp}$ (mmol/g), q_e^{cal} (mmol/g), and N are the experimental equilibrium absorption capacity, the absorption capacity calculated using models, and the number of experimental points, respectively.

The results of χ^2 and NSD for Langmuir and Freundlich isotherms are depicted in Table 4. It is noteworthy that the result is good when χ^2 and NSD are small numbers. The lower χ^2 and NSD values of Langmuir isotherm can be considered as the best fitting model for absorption.

The absorption isotherm of Langmuir indicates that the absorption process on the surface was homogeneous. According to Fig. 7, in which the nitrate absorption and adsorbent capacity of aggregates vs. nitrate concentration are shown, increasing nitrate concentrations decreased the nitrate removal, and absorption capacity index had an increasing trend. This can be used as an indicator of absorption efficiency and absorption capacity simultaneously [28]. Also, the nitrate concentration at approximately 60 ppm for all aggregates at the intersection of these two graphs (absorption removal and absorption capacity) can be considered as the initial concentration.

Table 4
The compliance of nitrates absorption with Freundlich and Langmuir absorption isotherms

Aggregate	Langmuir isotherm constants					Freundlich isotherm constants				
	b (L/mmol)	k_l (mmol/g)	R^2	χ^2	NSD (%)	k (mmol/g)	n	R^2	χ^2	NSD (%)
Pumice	0.0645	1.2202	0.99	0.625	0.667	0.1937	3.214	0.95	8.53	0.748
Perlite	0.0594	0.8112	0.98	0.818	0.724	0.1836	3.386	0.87	7.45	0.732
Zeolite	–0.0597	0.9011	0.92	0.961	1.39	0.1683	3.038	0.85	9.93	1.846

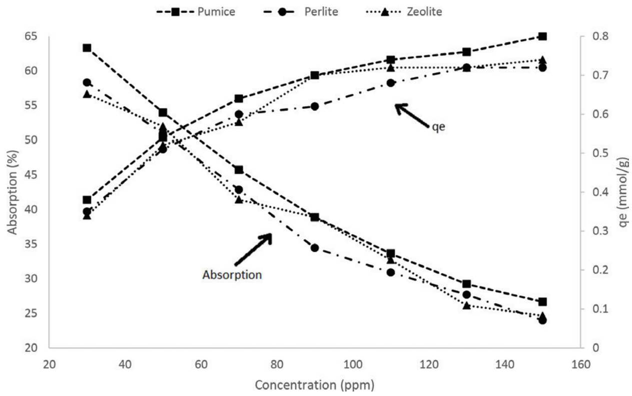

Fig. 7. Nitrate absorption and adsorbent capacity of aggregates vs. nitrate concentration.

3.5. Mechanism of nitrate removal using aggregate

Perlite, pumice, and zeolite with almost the same compositions include insoluble metal oxides [29]. Previous researches indicated that metal oxides have a positive charge, and are considered as an adsorbent with high absorption capacity for anion ions (such as nitrates) [11,12]. Also, the ligand exchange is introduced as the best exchange absorption of these aggregates regarding nitrate absorption [30]. The absorption of this anion depends on the pH, and since nitrate tends to connect to OH^- ions, neutral or lower pH solutions lead to better absorption. Nitrogen removal mechanism using aggregate's surface via chemical interaction is described by Eqs. (5)–(7):

$$MOH + H^+ \leftrightarrow MOH_2^+ \quad (5)$$

$$MOH_2^+ + NO_3^- \leftrightarrow MOH_2^+\text{- - - -}NO_3 \quad (6)$$

$$MOH_2^+ + NO_3^- \leftrightarrow MNO_3 + H_2O \quad (7)$$

where M is a metal oxide [12].

Fig. 8 shows the presence of metal ions in raw aggregates. As illustrated in the figure, in all combinations of metal ions, the trend of graphs is related to the nitrate absorption using raw aggregate. The metal ions and the amount of nitrate absorption increased in the same proportion for all three aggregates. In fact, since pumice aggregate had more metal ions, its nitrate removal was more in comparison with the two other aggregates. The relationship between absorption and total metal ions in raw aggregates is shown in Fig. 9 (this relation is not true for activated aggregate maybe due to affecting their structures by acid). Also, adding metal ions raised the removal rate. Moreover, Fig. 10 depicts how nitrate was absorbed schematically by aggregates in the presence of metal ions such as Si, Al, and Fe.

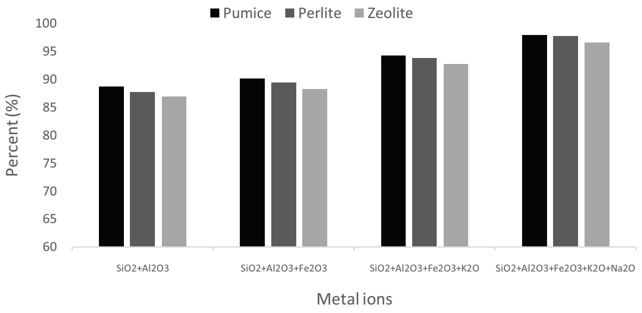

Fig. 8. Presence of metal ions in raw aggregates.

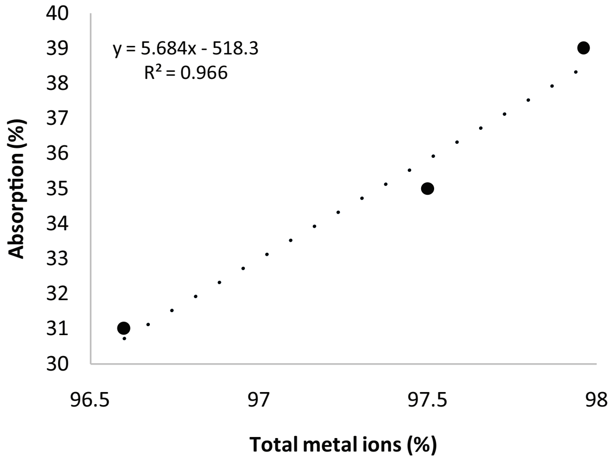

Fig. 9. Relation between absorption and total metal ions in raw aggregates.

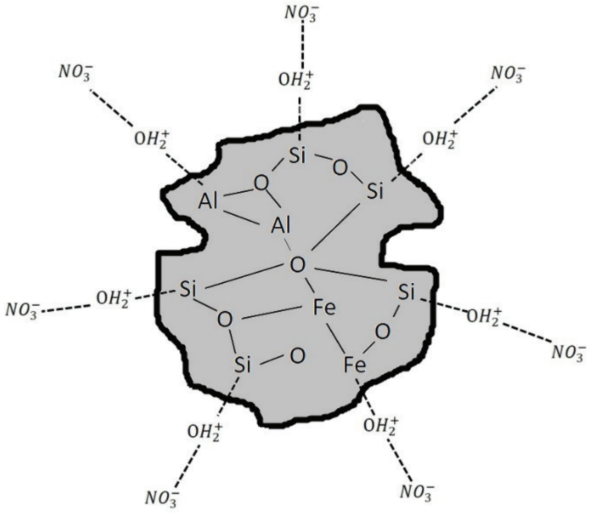

Fig. 10. Mechanism of nitrate absorption using metal ions in aggregates.

4. Conclusion

In this study, three aggregates (pumice, perlite, and zeolite) used in making pervious concrete were examined to assess their ability in reducing the concentration of nitrate from polluted water. According to the obtained results, it is noteworthy to mention that pumice was the best aggregate due to its lowest water absorption (7.84) during 48 h, lack of alkali-silica reactivity, highest permeability (1.64 cm/s), and most nitrate removal in the raw state (39%).

For all raw aggregates, nitrate absorption process followed Langmuir isotherm and the absorption capacity was about 70 mmol/g. The absorption isotherm of Langmuir indicates that the absorption process on the surface was homogeneous. Also, the nitrate concentration at approximately 60 ppm for all aggregates can be considered as the initial concentration.

All aggregates almost have the same compositions include insoluble metal ions and metal oxides. Metal oxides have a positive charge, and are considered as an adsorbent with high absorption capacity for nitrate ions. Also, the ligand exchange was introduced as the best exchange absorption of these aggregates regarding nitrate absorption.

Generally, pumice was selected as the best raw aggregate due to its lower water absorption, having no alkali-silica reactivity, highest permeability, and highest nitrate removal ability based on the presence of metal ions.

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