THE EVALUATION OF ELECTRICAL IMPEDANCE OF THREE-DIMENSIONAL FRACTAL NETWORKS EMBEDDED IN A CUBE

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Abstract

This paper presents a preliminary work to evaluate the electrical impedance of three-dimensional pore fractal networks embedded in a cube. The construction and structural parameters of pore and electrical networks are illustrated in detailed. The total impedance response (modulus and phase) of networks is primarily associated with structural parameter of network, pore structure parameter of cube, conductivity of pore and solid phases. The impedance response of cube with the evolution of fractal network is analyzed comprehensively in this work. Besides, the influence of conductivity of pore phase caused by different types of electrolytes, electrical frequencies and conductivity of solid phase on the total impedance response is systematically studied.

Keywords: Fractal; Network; Electrical Impedance; Porosity; Pore Size Distribution; Pore Tortuosity.

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1. INTRODUCTION

Fractal network structures are considered as typical branching structures abound in nature. A great number of porous systems, e.g. tree branches,^{1,2} leaf veins, blood vessels, bronchial tree, neural dendrites, cement-based materials,^{3–7} drainage network of river basins, lightning, cracks and particle aggregations, present typical fractal network features.⁸ Xu *et al.* considered that, on the one side, although the configuration and construction of fractal network embedded in natural transport systems are diverse, the statistical characteristics of natural tree-like topology are similar.⁸ However, most of the researchers only focus on the fluid flow^{9–11} and heat transfer^{12–14} of two-dimensional fractal networks due to their simplicity. The academic investigations of three-dimensional fractal networks that are actually close to the practice are limited. On the other side, the electrical properties of fractal networks have attracted broad interests from both scientists and engineers.^{15,16} The efficient electrical network systems embedded in transport systems have inspired the innovative design of artifacts¹³ such as polymer electrolyte fuel cells¹⁷ and batteries.¹⁸ Nevertheless, the comprehensive understanding of the influence of pore structure on electrical performance of fractal network or porous media is absent.^{7,19,20} For example, Tang *et al.*⁷ just regarded electrical response of pore electrolyte as a pure resistor when they attempted to correlate the impedance response to cumulative pore volume of fractal porous cement-based materials using frequency variable electrical measurement. Wei *et al.*¹⁹ proposed an electrical conductivity model for fractal porous media and indicated that the electrical conductivity was explicitly related to the porosity, electrical conductivity of pore liquid and fractal dimensions; nevertheless, this model did not take the complex form of electrical conductivity into account. Shitanda *et al.*²⁰ performed impedance simulation for analysis of a fractal structure porous carbon electrode composed of macropores and mesopores. They simply assumed that the electrolyte resistance per unit length was considered to be independent of applied frequencies. Tang *et al.*³ adopted fractal permeability model based on innovative non-contact electrical impedance measurement to evaluate the permeability of fresh cement pastes and investigated the effect of porosity and minimum and maximum pore sizes on permeability. However,

they did not take the three-dimensional properties of fractal model into account.

In this work, two three-dimensional fractal pore and electrical networks embedded in a cube are constructed. The influence of structural parameters of fractal network, pore structure parameters of cube, pore and solid phase conductivity on total impedance of cube is analyzed.

2. ELECTRICAL IMPEDANCE OF CONSTRUCTED CUBE

In the work, the constructed cube consists of pore and solid phases. Therefore, the total impedance of cube should be the summation of electrical impedance contribution of these two phases. The pore phase is taken as a typical pore fractal network. The proposed pore fractal network is analogous to the blood vessel in the body. The construction of pore fractal network is illustrated as: (1) the network is embedded into a particular cube and originates from the center of the cube. The pore fractal network evolves toward six faces of cube, namely, there are six branches of pore fractal network in the cube. (2) One of the branches of pore fractal network is constructed by the connection of cylinders and similar to the one proposed by Xu *et al.*,¹² as demonstrated in Fig. 1. Figure 1 shows the cross-section of three-dimensional

Fig. 1 The cross-section of three-dimensional pore branches at Level 4.

pore branches at Level 4. Two scale factors (length ratio γ and diameter ratio β) are defined in one of the branches of pore fractal network, to correlate the geometric relationship of cylinder at adjacent levels:

$$d_k = d_0 \beta^k, \quad (1)$$

$$l_k = l_0 \gamma^k, \quad (2)$$

where d_0 and l_0 are the diameter and length of mother pore channel of branch at Level 0, and similarly, d_k and l_k are the diameter and length of the pore channel at Level k .

The length (L_d) of the cube incorporating with pore fractal network is calculated by Eq. (3). In addition, the porosity (φ) of the cube is derived by summing up the volume of pore channels and expressed as:

$$L_d = 2 \sum_0^k l_k = 2l_0 \frac{(1 - \gamma^{k+1})}{(1 - \gamma)}, \quad (3)$$

$$\varphi = \frac{3\pi d_0^2 l_0}{2L_d^3} \left\{ \frac{3\beta^2 \gamma [1 - (3\beta^2 \gamma)^{k-1}]}{1 - 3\beta^2 \gamma} + \frac{6\beta^2 \gamma [1 - (\beta^2 \gamma)^{k-1}]}{1 - \beta^2 \gamma} + 3\beta^2 \gamma + 1 \right\}, \quad (k \geq 1). \quad (4)$$

With respect to the pore size distribution, based on the fractal scaling law recommended by Yu and Cheng,⁸ the fractal dimension (D_f) associated with pore size distribution can be correlated as:

$$N = 6 \left[4 + 3 \frac{(3^{k-1} - 1)}{2} + 6(k - 1) \right] = \left(\frac{d_0}{d_k} \right)^{D_f} = \beta^{-k D_f}, \quad (k \geq 1), \quad (5)$$

where N is the total number of pore channels with regard to the whole pore fractal network.

Additionally, it is well known that pore tortuosity plays a significant role in the electrical impedance, but the explicit expressions of tortuosity are still in debate until now.²¹ One of the directions of tortuosity of pore fractal network is defined as the ratio of total length of all pore channels in one direction (L_e) to side length of cube, which is demonstrated in Eqs. (6) and (7):

Fig. 2 The cross-section of electrical fractal network at Level 4.

$$L_e = l_0 \left\{ \frac{3\gamma^2 [1 - (3\gamma)^{k-1}]}{1 - 3\gamma} + \frac{6\gamma^2 [1 - \gamma^{k-1}]}{1 - \gamma} + 3\gamma + 1 \right\}, \quad (k \geq 1), \quad (6)$$

$$\tau = \frac{2L_e}{L_d}. \quad (7)$$

Accordingly, the pore fractal network has the corresponding electrical fractal network, as illustrated in Fig. 2. Figure 2 shows the cross-section of electrical fractal network at Level 4. In regard to impedance response of electrical fractal network, for the simplicity of analysis, it is essential to put forward two prerequisites: (1) all the pore channels of fractal network are full of conductive electrolyte and (2) the conductivity of remaining solid phases of cube is smaller than the one of pore phase.

The modulus and phase of impedance for the electrical fractal network are derived by electrical analogy. Similar to Ohm's law, the electrical impedance of a single pore channel at Levels 0 and k is derived as:

$$Z_0 = \frac{4l_0}{(\sigma_p \pi d_0^2)}, \quad (8)$$

$$Z_k = \frac{4lk}{(\sigma_p \pi d_k^2)}, \quad (9)$$

$$Z_k = \left(\frac{\gamma}{\beta^2}\right)^k Z_0, \quad (10)$$

where Z_0 and Z_k are the electrical impedances of pore channel at Levels 0 and k and σ_p is the electrical conductivity of the pore channel.

Thereinto, σ_p is derived based on multiple Debye equation²²:

$$\sigma_p = \frac{1}{(k_b T)} \sum_{\lambda=1}^2 \frac{\rho_{\lambda} q_{\lambda}^2 D_{\lambda}}{[1 - i\omega D_{\lambda} m_{\lambda} / (k_b T)]}, \quad (11)$$

where k_b is the Boltzmann constant, T the absolute temperature, ω the angular frequency and ρ_{λ} , q_{λ} , m_{λ} and D_{λ} represent the bulk number density, charge of an ion, ionic mass and self-diffusion coefficient of ionic species λ , respectively. The self-diffusion coefficient indicates the time that takes an ion to diffuse a given distance in a liquid medium.^{23,24} It is expected that the diffusion coefficient is a function of physical characteristics of both the ion and liquid medium.²⁵ Besides, it is worth pointing out that the coefficient is inversely related to the molecular weight of ion.²⁵

Based on the parallel mode of electrical fractal network, the total electrical impedance (Z_p) of pore fractal network is calculated as:

$$Z_p = \frac{Z_d}{6}, \quad (12)$$

where Z_d is the electrical impedance of a pore branch, deduced by summing up all the electrical impedances of pore channels at different levels via serial and parallel modes.

When the total impedance of the cube is considered, the contribution of solid phase should not be ignored. Therefore, the impedance of solid phase (Z_s) in the cube and total electrical impedance of the cube (Z_t) are calculated as below:

$$Z_s = Z_p \sigma_p \frac{(L_d^3 - V_p)}{(V_p \sigma_s)}, \quad (13)$$

$$\frac{1}{Z_t} = \frac{1}{Z_p} + \frac{1}{Z_s}, \quad (14)$$

where σ_s is the conductivity of the solid phase and V_p is the volume of the pore phase.

3. RESULTS AND DISCUSSION

In this section, the solving of total electrical impedance Z_t is based on Eqs. (8)–(14) and resorted to Matlab program. A detailed analysis of Z_t based on the fractal network is presented in the following, and the influencing factors of Z_t are discussed.

Fig. 3 Modulus and phase development of impedance with β .

3.1. Influence of Structural Parameters of Fractal Network on Electrical Impedance

Apart from Sec. 3.3, the conductive electrolyte filled in pore channels is supposed to be NaCl with concentration 1 mol/L and the investigated temperature about 25°. Figure 3 shows the modulus and phase development of impedance with β when γ is 0.5, k is 5, σ_s is 0.1 S/m, both l_0 and d_0 are equal to 10, 50, 100, 500 and 1000 μm . In Fig. 3, the range of β is from 0.1 to 1. The physical meaning of modulus and phase of impedance in this work are particularly illustrated as follows: the modulus of impedance reflects the hindering effect of cube to the electrical current,²⁶ whereas the phase of impedance reveals the relaxation of ion in pore channels of network.²⁶ The relaxation of ion was preliminarily explained by previous literatures: (1) Choudhary and Sengwa²⁷ considered that the ion relaxation was sometimes also referred to as Maxwell–Wagner interfacial polarization relaxation process, caused by liquid–solid interface. (2) Cations and anions having different molecular shapes, chemical structures and properties of electric-field-induced reorientation were confined in pore channels and

showed evident ion-specific effects in electrotunable friction.²⁸

With regard to Fig. 3, the modulus decreases sharply with the increase of β from 0.1 to 0.3. Nevertheless, the subsequent evolution of modulus becomes slow. This phenomenon can be explained from the percolation of impedance response in terms of pore size. Ghanbarian *et al.*²⁹ investigated the saturation dependence of transport and percolation in porous media by effective medium theory and considered that the percolation of impedance response was strongly associated with the pore size distribution. The large value of β indicates the large diameter of pore channels in the whole fractal pore network. Obviously, the large pore diameter is sure to eliminate the ion friction and the electrical resistance of pore channels,^{30–32} which is verified by the changes of values of l_0 and d_0 in Fig. 3.

On the whole, the phase presents negative value and “S” shape with β in Fig. 3. The negative value of phase implies that the lag of current falls behind that of voltage.^{33,34} The short relaxation time corresponds to large value of β and small absolute value of phase.^{35–37} Besides, it seems that the selected values (10–1000 μm) of d_0 and l_0 do not have any remarkable effect on the value of phase. It is demonstrated again that the comparably large size of mother pore channel has minor effect on the ion relaxation.^{30–32}

Figure 4 shows the modulus and phase development of impedance with γ when both d_0 and l_0 are equal to 10, 50, 100, 500 and 1000 μm . In Fig. 4, σ_s is 0.1 S/m, the range of γ is from 0.1 to 0.9, β is 0.5 and k is 5. It is expected that the modulus of impedance generally increases with the increase of γ . Large value of γ basically means long pore channel of fractal network, which will cause the large ion friction in the pore channel and large impedance modulus.^{30–32} Besides, when d_0 and l_0 are equal or more than 50 μm , the change of modulus of impedance is not evident with γ in Fig. 4. This phenomenon implies that when the pore sizes of fractal network are small enough, small pores just have little contribution to total impedance response of pore phase.³

With respect to the phase of impedance, when the value of γ increases, the phase of impedance decreases gradually. The effects of d_0 and l_0 on the phase of impedance in Fig. 4 are similar to those in Fig. 3; the dot lines are almost overlapping, irrespective of values of d_0 and l_0 . It should also be noted that phase of impedance is very small and

Fig. 4 Modulus and phase development of impedance with γ .

between 0 and 10^{-16} rad. This indicates that the electrical response of investigated cube saturated with ion water likely behaves as a pure resistor.

Figure 5 shows modulus and phase development of impedance with k when d_0 and l_0 are equal to 10, 50, 100, 500 and 1000 μm . σ_s is 0.1 S/m and β and γ are equal to 0.5. With respect to k , its values are from 1 to 10. Obviously, the increasing rate of the modulus and phase gradually becomes small with the increase of k in Fig. 5. In particular, when k is equal to or larger than 4, the modulus and phase of impedance change slowly. When the fraction of solid phase with low electrical conductivity steadily increases in the cube with increasing of k based on Eq. (3), the impedance modulus of cube is sure to increase. Besides, the influence of d_0 and l_0 on modulus in Fig. 5 cannot be ignored. When k is fixed, the large values of d_0 and l_0 lead to small modulus. Large size of mother pore channel is beneficial for the ion transportation and the gain of small modulus without doubt.

When k increases, the negative value of phase of impedance decreases as the amount of pore channel and ion friction increases at the same time in the cube. In addition, the trend of phase of impedance

Fig. 5 Modulus and phase development of impedance with k .

is similar to that in Figs. 3 and 4: the evolution of phase is not closely linked to the pore size or length of mother channel and presents characteristic percolation phenomenon.

Figure 6 shows modulus and phase development of impedance with different d_0 values (90, 95, 100, 105 and $110 \mu\text{m}$). Meanwhile, the selected values of l_0 are also set as 90, 95, 100, 105 and $110 \mu\text{m}$. Other parameters are shown as follows: σ_s is 0.1 S/m , k is 5, β and γ are equal to 0.5. From the viewpoint of electrical impedance based on Eqs. (8) and (9), the effects of d_0 and l_0 on the modulus and phase of impedance are completely reverse: large d_0 is apparently beneficial to provide sufficient space for the ion transportation and eliminate ion friction; therefore, small value of impedance modulus is obtained. It is considered that by the comparison of Figs. 5 and 6, the large d_0 plays a crucial role in the impedance modulus of the cube.^{3,38} On the contrary, when l_0 increases, the value of modulus increases, since long transportation path can result in large friction between ion and pore wall or among ions. In addition, the effect of d_0 on modulus is more profound than that of l_0 in the selected range of values (90– $110 \mu\text{m}$).

Fig. 6 Modulus and phase development of impedance with d_0 .

For the case of phase, the value of phase monotonously increases with increasing d_0 or decreasing l_0 . From the previous analysis of Figs. 3–5, the phase was primarily rooted in the pore phase of cube. Large l_0 or small d_0 brings about an obvious consequence of rising the difficulty degree of ions migrating in pore channels; the impedance phase will accordingly decrease.

3.2. Influence of Pore Structure Parameters on Electrical Impedance

3.2.1. Influence of porosity on electrical impedance

Porosity is a significant parameter to reflect the electrical transportation of heterogeneous porous media. Electrical impedance of porous media was reported to be associated with porosity and saturation dependence, according to typical Archie's law.³⁹ Figure 7 shows modulus and phase development of impedance with porosity when β is from 0.1 to 1. Other parameters are fixed as follows: σ_s is 0.1 S/m , k is 5, $d_0 = l_0 = 100 \mu\text{m}$ and γ is 0.5. The vertical axis on the left-hand side in Fig. 7 stands

Fig. 7 Modulus and phase development of impedance with porosity.

for modulus of impedance and the one on right-hand side represents phase of impedance. In the investigated cube, the electrical conductivity of the pore phase is assumed to be larger than that of the solid phase, and thus, the porosity makes great contribution to impedance response of cube, confirmed by previous literatures.^{7,40}

From Fig. 7, when the porosity of the cube is very small, the modulus and phase of impedance evolve quickly; subsequently, when porosity reaches a certain value, both modulus and phase have minor changes. In particular, the evolution of modulus presents distinct “L” shape. On the one side, when porosity is too small, the impedance modulus is primarily dependent on the intrinsic electrical conductivity of the solid phase and presents comparably large value. On the other side, in general, the large porosity of the cube apparently leads to small impedance modulus as the pore channels are assumed to be saturated with conductive electrolyte. Similar electrical percolation of porous media in terms of porosity was also found in the sand (1% porosity)³⁹ and cement pastes (18% porosity).⁷ The phase evolution in Fig. 7 is in line with the ones illustrated in Figs. 3–6: large modulus corresponds to small phase.

3.2.2. Influence of pore size distribution on electrical impedance

Song *et al.*⁴¹ developed the transmission line model based on equivalent circuits to examine the experimental impedance data of gold power electrodes. It was indicated that a wide pore size distribution

Fig. 8 Modulus and phase development of impedance with D_f .

led to porous characteristics of impedance of electrodes.⁴² Furthermore, as mentioned in Figs. 3–7, the different pore sizes take different effects on electrical impedance of fractal network. In this work, the pore size distribution is characterized by the fractal dimension D_f . Obviously, D_f is mainly related to pore size distribution and pore amount, based on Eq. (5). The two physical meanings of D_f are illustrated as: (1) when the value of d_0/d_k is fixed, the amount of pores and D_f have the same evolution trend and (2) the small value of D_f exhibits a narrow pore size distribution when the amount of pores remains unchanged.

Figure 8 shows modulus and phase development of impedance with D_f when k increases from 1 to 10. Other parameters are fixed as follows: σ_s is 0.1 S/m, $d_0 = l_0 = 100 \mu\text{m}$ and $\beta = \gamma = 0.5$. From the data trend of Fig. 8, the evolution of D_f in the selected range brings about the decrease of modulus. Similar to Figs. 3–7, the phase evolution exhibits the opposite trend with evolution of D_f , compared with the modulus evolution. Similar findings can be acquired from previous literature by Liang *et al.*⁴³ They studied the streaming current in porous media based on the capillary model and fractal theory and pointed out that when $D_f > 1.75$, the current of investigated porous media noticeably increased with the fractal dimension D_f increasing.

3.2.3. Influence of pore tortuosity on electrical impedance

It is well known that the resistance to current in pore paths is primarily related to the pore tortuosity.^{21,44} Nevertheless, the investigation of influence of pore tortuosity on electrical impedance is limited, although it is noted that the tortuous path

Fig. 9 Modulus and phase development of impedance with pore tortuosity.

is helpful to increase the electrical impedance to some degree.²¹ Figure 9 shows the modulus and phase development of impedance with pore tortuosity when γ is from 0.1 to 0.9. Other parameters are fixed as follows: σ_s is 0.1 S/m, $d_0 = l_0 = 100 \mu\text{m}$, $k = 5$ and $\beta = 0.5$. From this figure, it is observed that large τ gives rise to large absolute value of modulus and phase and vice versa. Based on Eqs. (3), (6) and (7), the tortuous conductive path is favorable to increase large resistance to current beyond all doubt. The mechanism behind is associated with the ion friction in pore channels.^{30–32} It is expected that large tortuosity of conductive path considerably makes for the large ion friction.

3.3. Influence of Types and Concentrations of Electrolytes on Electrical Impedance

In this work, the influences of types and concentrations of electrolytes on electrical impedance are discussed. According to Eq. (11), the self-diffusion coefficients of different conductive electrolytes should be carefully obtained when different electrolytes are considered.³⁹ The values of self-diffusion coefficient of different ions in water at 25° are shown in Table 1.²⁵ It is found from this table that the value of self-diffusion coefficient of H^+ is the largest, the one of Cl^- is the next and the one of Ca^{2+} is the smallest.

Figure 10 shows the modulus and phase development of impedance with different types and concentrations of electrolytes. Other parameters are fixed as: $\sigma_s = 0.1 \text{ S/m}$, $k = 5$, $d_0 = l_0 = 100 \mu\text{m}$ and $\beta = \gamma = 0.5$. The changes of both modulus and phase of impedance for different electrolytes present

Table 1 Values of Self-Diffusion Coefficient of Different Ions in Water at 25°C.²⁵

Ions	Atomic Weight (g/mol)	Diffusion Coefficient ($10^{-5} \text{ cm}^2/\text{s}$)
H^+	1.008	9.31
Na^+	22.990	1.33
K^+	39.098	1.96
Ca^{2+}	40.078	0.79
Cl^-	35.453	2.03

Fig. 10 Modulus and phase development of impedance with different types and concentrations of electrolytes.

the characteristic trends: the values of modulus and phase decrease at the same time when the concentration increases, although decreasing configurations of modulus and phase are very different. The modulus evolves quickly in the low concentration range and then tends to be stable in the high concentration range in Fig. 10. In contrast, the phase exhibits apparent linearity. Tang^{45,46} derived a theoretical equation to evaluate the ion friction and found out in a high concentration electrolyte that ions were so close to each other that the friction effect was considerably obvious. High concentration remarkably led to large ion friction from his experimental data and modeling results. From the comparison of impedance results in Fig. 10, the modulus sequence of different electrolytes is illustrated as: $\text{CaCl}_2 > \text{HCl} > \text{KCl} > \text{NaCl}$; whereas the phase sequence is as: $\text{NaCl} > \text{CaCl}_2 > \text{HCl} > \text{KCl}$. It is demonstrated that the self-diffusion coefficient of electrolytes and carried charges have great impact on total impedance of the cube. Choudhary and Sengwa²⁷ analyzed the ionic conduction and relaxation processes of nanocomposite and reported that the electrical conductivity of pore channels was

associated with charge carriers density, ionic mobility and the charge of ion. In general, the increase of conductivity of electrolyte was due to the increment in number of mobile charge carriers introduced/produced in the pore. Evidently, large self-diffusion coefficient and/or many carried charges are beneficial to small modulus and phase, as shown in Fig. 10.

3.4. Influence of Electrical Frequency on Electrical Impedance

Paluch *et al.*²⁶ and Sapoval *et al.*⁴⁷ considered that electrical impedance of porous media was influenced by electrical frequency and revealed the different Debye electrical relaxations in different frequency ranges. Chandra *et al.*⁴⁸ also reported that the electrical impedance of pore fractal network was principally associated with electrical frequency. Figure 11 shows the modulus and phase development of impedance with different electrical frequencies. In this figure, the frequency is selected from 10^{-6} to 10^6 kHz. Other parameters as fixed as: $\sigma_s = 0.1$ S/m, $k = 5$ and $\beta = \gamma = 0.5$, d_0 and l_0 are equal to $100 \mu\text{m}$. The selected electrical frequency does not have any influence on the modulus of impedance. From Fig. 11, it is found that the modulus of impedance is almost a fixed value ($0.391 \text{ k}\Omega$), regardless of frequencies. Chandra and Bagchi²² claimed that this phenomenon may be attributed to the well-known Debye–Falkenhagen effect.

With regard to phase, the sharp decreasing point at frequency 10^6 kHz may be related to the dispersion of the electrolyte at high frequency caused by fast relaxation of the ion atmosphere.²² Apart from

Fig. 11 Modulus and phase development of impedance with different electrical frequencies.

Fig. 12 Modulus and phase development of impedance with different conductivities of solid phase.

the sharp decreasing point, although the phase of impedance actually decreases from -1.8×10^{-29} to -1.8×10^{-7} rad, the curve of frequency seems to be flat between 10^{-6} and 10^5 kHz. The phenomenon also illustrates that the electrical impedance of the investigated cube likely behaves as a pure resistor within the selected frequency range, observed from Fig. 11.

3.5. Influence of Solid Conductivity on Electrical Impedance

From the observation of phase of impedance in Figs. 3–11, it can be inferred that the impedance phase of cube is so small that it can be ignored. Therefore, the conductivity of the solid phase is also assumed to be selected as 10^{-4} to 1 S/m in Fig. 12, which shows modulus and phase development of impedance with different conductivities of the solid phase. Other parameters are fixed as: $k = 5$, $d_0 = l_0 = 100 \mu\text{m}$ and $\beta = \gamma = 0.5$. The modulus and phase present the same evolution trend. Besides, from Fig. 12, when the conductivity of the solid phase is close to zero, namely, the solid phase is almost insulated, modulus and phase approach to stable values. It seems that the 0.1 S/m conductivity of the solid phase is a critical percolation value. Large value of conductivity of the solid phase indeed promotes the overall conductivity of the cube.

4. CONCLUSION

In this work, two three-dimensional fractal networks are constructed in an investigated cube. The electrical impedance (modulus and phase) of the cube is analyzed based on the changes of structural parameters, pore structure parameters, types and

concentrations of electrolytes, electrical frequency and conductivity of the solid phase of network or cube. The percolation phenomena of impedance response in terms of pore size are found. From the comparison of modeling results of structural parameters, phase of impedance reveals the relaxation of ion and is associated with ion friction. In addition, the long pore channel is believed to increase the ion friction. It is worth pointing out that apart from large pore tortuosity, small porosity and D_f lead to large absolute values of modulus and phase of impedance. Large self-diffusion coefficient and/or many carried charges are beneficial to small modulus and phase. It seems that electrical impedance of the investigated cube likely behaves as a pure resistor, regardless of electrical frequencies. Besides, large value of conductivity of the solid phase indeed promotes the overall conductivity of the cube.

Actually, the possible capacitance caused by electrical double layers between solid and electrolyte interface and inductance caused by pore tortuosity are ignored in this work. These insufficiencies of work on electrical impedance will be overcome in the future.

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REFERENCES

1. C. Li, Y. Shen, H. Ge, S. Su and Z. Yang, Analysis of spontaneous imbibition in fractal tree-like network system, *Fractals* **24**(3) (2016) 1650035.
2. X. Chen, Z. Zhang, D. Yi and Z. Hu, Numerical studies on different two-dimensional micromixers basing on a fractal-like tree network, *Microsyst. Technol.* **23**(3) (2017) 755–763.
3. S. W. Tang, Z. J. Li, H. G. Zhu, H. Y. Shao and E. Chen, Permeability interpretation for young cement paste based on impedance measurement, *Constr. Build. Mater.* **59** (2014) 120–128.
4. S. Tang, Y. Yao, C. Andrade and Z. Li, Recent durability studies on concrete structure, *Cem. Concr. Res.* **78** (2015) 143–154.
5. E. Chen and C. K. Leung, Effect of uniaxial strength and fracture parameters of concrete on its biaxial compressive strength, *J. Mater. Civil. Eng.* **26**(6) (2014) 06014001.
6. S. W. Tang, E. Chen, H. Y. Shao and Z. J. Li, A fractal approach to determine thermal conductivity in cement pastes, *Constr. Build. Mater.* **74** (2015) 73–82.
7. S. W. Tang, Z. J. Li, E. Chen and H. Y. Shao, Impedance measurement to characterize the pore structure in Portland cement paste, *Constr. Build. Mater.* **51** (2014) 106–112.
8. B. Yu and P. Cheng, A fractal permeability model for bi-dispersed porous media, *Int. J. Heat Mass Transf.* **45**(14) (2002) 2983–2993.
9. H. S. Damiri and H. K. Bardaweel, Numerical design and optimization of hydraulic resistance and wall shear stress inside pressure-driven microfluidic networks, *Lab Chip* **15**(21) (2015) 4187–4196.
10. L. Zhang, D. Peng, W. Lyu and F. Xin, Uniformity of gas and liquid two phases flowing through two microchannels in parallel, *Chem. Eng. J.* **263** (2015) 452–460.
11. Y. Su, G. Chen and E. Y. Kenig, An experimental study on the numbering-up of microchannels for liquid mixing, *Lab Chip* **15**(1) (2015) 179–187.
12. P. Xu, B. Yu, M. Yun and M. Zou, Heat conduction in fractal tree-like branched networks, *Int. J. Heat Mass Transf.* **49**(19–20) (2006) 3746–3751.
13. P. Xu and B. Yu, The scaling laws of transport properties for fractal-like tree networks, *J. Appl. Phys.* **100**(10) (2006) 104906.
14. B. Yu and B. Li, Fractal-like tree networks reducing the thermal conductivity, *Phys. Rev. E* **73**(6) (2006) 066302.
15. A. Bejan and S. Lorente, Constructal law of design and evolution: Physics, biology, technology, and society, *J. Appl. Phys.* **113**(15) (2013) 151301.
16. S. W. Tang, X. H. Cai, W. Zhou, H. Y. Shao, Z. He, Z. J. Li, W. M. Ji and E. Chen, *In-situ* and continuous monitoring of pore evolution of calcium sulfoaluminate cement at early age by electrical impedance measurement, *Constr. Build. Mater.* **117** (2016) 8–19.
17. M. N. Nasharudin, S. K. Kamarudin, U. A. Hasran and M. S. Masdar, Mass transfer and performance of membrane-less micro fuel cell: A review, *Int. J. Hydrogen Energy.* **39**(2) (2014) 1039–1055.
18. Y. Chen, F. Yao and X. Huang, Mass transfer and reaction in methanol steam reforming reactor with fractal tree-like microchannel network, *Int. J. Heat Mass Transf.* **87** (2015) 279–283.

19. W. Wei, J. Cai, X. Hu and Q. Han, An electrical conductivity model for fractal porous media, *Geophys. Res. Lett.* **42**(12) (2015) 4833–4840.
20. I. Shitanda, S. Tsujimura, H. Yanai, Y. Hoshi and M. Itagaki, Electrochemical impedance simulation of branch structure porous carbon electrode using transmission line model, *Electrochemistry* **83**(5) (2015) 335–338.
21. B. Ghanbarian, A. G. Hunt, R. P. Ewing and M. Sahimi, Tortuosity in porous media: A critical review, *Soil Sci. Soc. Am. J.* **77**(5) (2013) 1461–1477.
22. A. Chandra and B. Bagchi, Frequency dependence of ionic conductivity of electrolyte solutions, *J. Chem. Phys.* **112**(4) (2000) 1876–1886.
23. S. W. Tang, Z. J. Li, E. Chen and H. Y. Shao, Non-steady state migration of chloride ions in cement pastes at early age, *RSC Adv.* **4**(89) (2014) 48582–48589.
24. S. W. Tang, E. Chen, Z. J. Li and H. Y. Shao, Assessment of steady state diffusion of volatile organic compounds in unsaturated building materials based on fractal diffusion model, *Build. Environ.* **84** (2015) 221–227.
25. B. Hille, *Ion Channels of Excitable Membranes* (Sinauer Associates Inc., Sunderland, 2001).
26. M. Paluch, S. Pawlus, J. Klimontko, Z. Ujma, I. Gruszka and A. Molak, Electric modulus approach to the analysis of electric relaxation in highly conducting $(\text{Na}_{0.75}\text{Bi}_{0.25})(\text{Mn}_{0.25}\text{Nb}_{0.75})\text{O}_3$ ceramics, *J. Phys. D Appl. Phys.* **38**(9) (2005) 1450–1460.
27. S. Choudhary and R. J. Sengwa, Ionic conduction and relaxation processes in melt compounded poly (ethylene oxide)-lithium perchlorate trihydrate-montmorillonite nanocomposite electrolyte, *Ind. J. Eng. Mater. S* **18**(2) (2011) 147–156.
28. O. Y. Fajardo, F. Bresme, A. A. Kornyshev and M. Urbakh, Electrotunable friction with ionic liquid lubricants: How important is the molecular structure of the ions?, *J. Phys. Chem. Lett.* **6**(20) (2015) 3998–4004.
29. B. Ghanbarian, A. G. Hunt, T. E. Skinner and R. P. Ewing, Saturation dependence of transport in porous media predicted by percolation and effective medium theories, *Fractals*, **23**(01) (2015) 1540004.
30. X. Wei, M. Shao, L. Du and R. Horton, Humic acid transport in saturated porous media: Influence of flow velocity and influent concentration, *J. Environ. Sci.* **26**(12) (2014) 2554–2561.
31. A. Massoudieh, N. Lu, X. Liang, T. H. Nguyen and T. R. Ginn, Bayesian process-identification in bacteria transport in porous media, *J. Contam. Hydrol.* **153** (2013) 78–91.
32. L. L. Landkamer, R. W. Harvey, T. D. Scheibe and J. N. Ryan, Colloid transport in saturated porous media: Elimination of attachment efficiency in a new colloid transport model, *Water Resour. Res.* **49**(5) (2013) 2952–2965.
33. S. W. Tang, Z. J. Li, H. Y. Shao and E. Chen, Characterization of early-age hydration process of cement pastes based on impedance measurement, *Constr. Build. Mater.* **68** (2014) 491–500.
34. S. W. Tang, H. G. Zhu, Z. J. Li, E. Chen and H. Y. Shao, Hydration stage identification and phase transformation of calcium sulfoaluminate cement at early age, *Constr. Build. Mater.* **75** (2015) 11–18.
35. S. Muncaci, C. Mattea, S. Stapf and I. Ardelean, Frequency-dependent NMR relaxation of liquids confined inside porous media containing an increased amount of magnetic impurities, *Magn. Reson. Chem.* **51**(2) (2013) 123–128.
36. R. D. Swanson, A. Binley, K. Keating, S. France, G. Osterman, F. D. Day-Lewis and K. Singha, Anomalous solute transport in saturated porous media: Relating transport model parameters to electrical and nuclear magnetic resonance properties, *Water Resour. Res.* **51**(2) (2015) 1264–1283.
37. Y. Song, Magnetic resonance of porous media (MRPM): A perspective, *J. Magn. Reson.* **229** (2013) 12–24.
38. S. W. Tang, X. H. Cai, Z. He, W. Zhou, H. Y. Shao, Z. J. Li, T. Wu and E. Chen, The review of pore structure evaluation in cementitious materials by electrical methods, *Constr. Build. Mater.* **117** (2016) 273–284.
39. R. P. Ewing and A. G. Hunt, Dependence of the electrical conductivity on saturation in real porous media, *Vadose Zone J.* **5**(2) (2006) 731–741.
40. D. Wiedenmann, L. Keller, L. Holzer, J. Stojadinović, B. Münch, L. Suarez, B. Fumey, H. Hagedorfer, R. Brönnimann, P. Modregger, M. Gorbar, U. F. Vogt, A. Züttel, F. L. Mantia, R. Wepf and B. Grobety, Three-dimensional pore structure and ion conductivity of porous ceramic diaphragms, *AIChE J.* **59**(5) (2013) 1446–1457.
41. H.-K. Song, H.-Y. Hwang, K.-H. Lee and L. H. Dao, The effect of pore size distribution on the frequency dispersion of porous electrodes, *Electrochim. Acta* **45**(14) (2000) 2241–2257.
42. H.-K. Song, Y.-H. Jung, K.-H. Lee and L.H. Dao, Electrochemical impedance spectroscopy of porous electrodes: The effect of pore size distribution, *Electrochim. Acta* **44**(20) (1999) 3513–3519.
43. M. Liang, S. Yang and B. Yu, A fractal streaming current model for charged microscale porous media, *J. Electrostat.* **72**(6) (2014) 441–446.
44. B. Ghanbarian, A. G. Hunt, M. Sahimi, R. P. Ewing and T. E. Skinner, Percolation theory generates a physically based description of tortuosity in saturated and unsaturated porous media, *Soil Sci. Soc. Am. J.* **77**(6) (2013) 1920–1929.

45. L. Tang, Concentration dependence of diffusion and migration of chloride ions: Part 1. Theoretical considerations, *Cem. Concr. Res.* **29**(9) (1999) 1463–1468.
46. L. Tang, Concentration dependence of diffusion and migration of chloride ions: Part 2. Experimental evaluations, *Cem. Concr. Res.* **29**(9) (1999) 1469–1474.
47. B. Sapoval, J. N. Chazalviel and J. Peyrière, Electrical response of fractal and porous interfaces, *Phys. Rev. A* **38**(11) (1988) 5867–5887.
48. A. Chandra, D. Wei and G. N. Patey, The frequency dependent conductivity of electrolyte solutions, *J. Chem. Phys.* **99**(3) (1993) 2083–2094.