

# Tortuosity Model of a Three-Dimensional Stacked Cylindrical Particles Porous Medium for Using with the Thin Film Solid Oxide Fuel Cell Electrodes

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## Abstract

*This work is precisely intended to derive an analytical model for tortuosity of streamlines in a three dimensional porous material of fuel cell electrodes, especially for the thin film electrode of solid oxide fuel cells. The arrangement of the particles that form the electrodes as the cylindrical particles stack, only arrange together in the vertical direction. The model is derived assuming and averaging over some possible streamlines around the particles and for the different particle configuration arrangements, and focuses only on the flow of a streamline passing the vertical stack cylinders. The present model is only dependent on distance between particles and particle sizes. The model predictions are successfully compared to the correlation obtained from available experiments in the literature, especially at high cylinder height to diameter ratios. There is agreement on the thin film electrode characteristics of solid oxide fuel cells.*

*Keywords: analytical model, cylindrical particles stack, fuel cells, porous material, thin film*

## 1. Introduction

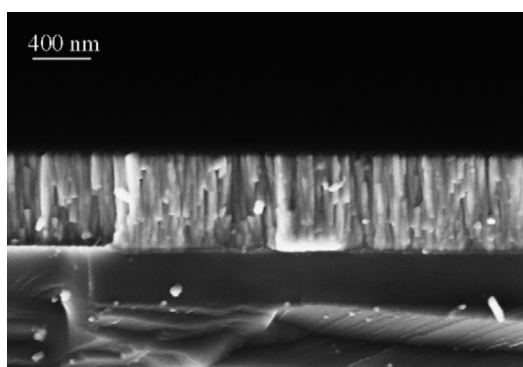
The two major properties for describing a porous medium and the associated mass transfer phenomena are the permeability coefficient (flow phenomena) and the effective diffusion (mass transfer phenomena). However, both coefficients are a function of the characteristics of the porous medium, namely the porosity and tortuosity [1, 2]. The tortuosity is an important parameter that describes pore connectivity and fluid transport and it is very complicated to determine. As a physical quantity, it can be defined in various ways. The most often encountered definition is the ratio of the average length of true flow paths to

the length of the system in the direction of the macroscopic flow. In that case, the tortuosity depends not only on the microscopic geometry of the pores, but also on the transport mechanism under consideration. However, tortuosity can also be defined without reference to a specific transport mechanism. This can be done, for example, by considering the shortest continuous path between any two points within the pore space [3]. The advantage of this definition is that the tortuosity parameter is intrinsic to the material which exclusively characterizes the porous substance itself.

The tortuosity in porous media can also be determined by experiments [4, 5] and the experimental data are thus fitted by empirical laws, also called correlations. From the experiment, the tortuosity of streamlines in porous media can also be estimated by numerical methods. Rigby and Gladden [6] applied the whole object averaging method, diffusion flux method, and random walk method. The tortuosity and the tortuosity for diffusivity of gases /fluids were expressed as series. Koponen et al [7] applied the Lattice Gas (LG) cellular automaton method to solve numerically a creeping flow of Newtonian incompressible fluid in a two – dimensional porous substance constructed by randomly packing rectangles of equal size. Wang at al [8] derived an approximate correlation between the non – Darcy coefficient tensor, tortuosity tensor and permeability tensor. Unfortunately, no explicit expression for the tortuosity tensor has been presented.

In order to achieve a practical understanding of the mechanisms for tortuosity of tortuous flow paths through porous media, an analytical model for tortuosity is desirable. Two dimensional tortuosity models are available for packed squares and disk [9, 10]. A three dimensional models was proposed by M. Yun et al. [11] suitable for packing of cubes, plates, and spheres. Recently, a geometry model for tortuosity of the packing cylindrical particles was expressed [12]. The model is averaged over only a few configurations and there is less comparison to the experimental data.

This is of high interest in the case of catalytic thin films obtained for example, by plasma sputtering [13-16] which exhibit columnar structures. That is one of the applications for producing the electrodes of thin film fuel cells. The major application of the thin film fuel cell is its use in the electrode and electrolyte of the thin film solid oxide fuel cells (or thin film SOFCs). The major limitation of using of SOFCs is their high operating temperature. As a solution of this drawback and to achieve stability and economics the use intermediate temperature SOFCs (IT-SOFCs) and low temperature SOFCs are the subject of growing interest. The techniques of thin electrolyte and electrode coating involving new materials have been chosen for intermediate and low temperature SOFCs or thin film techniques. An example of a thin film SOFC electrode is shown in Fig. 1.



**Fig. 1.** Cross-section micrographs of an anode Ni - cerium gadolinium oxide (CGO) electrode with deposition RF sputtering [17].

This work is precisely intended to derive an analytical model for tortuosity in a three dimensional porous material of the fuel cells electrode, especially for the thin film electrode of solid oxide fuel cells. The arrangement of the particles to form the electrodes as the cylinder stack is only in the vertical direction. So we focus only on the flow of a streamline passing the vertical stack cylinders. M. Yun et al.'s model [12] does not emphasize this idea, The derivation method to reach the final tortuosity equation in this present model is different from M. Yun et al.'s model [12].

Usually, the tortuosity ( $\tau$ ) is often defined by [1, 2]:

$$\tau = L_e / L \quad (1)$$

where  $L_e$  is the actual length of flow path or the total distance traveled through a packing system and  $L$  is the straight length or the shortest theoretical distance between defined starting and ending points of a packing system. So  $\tau \geq 1$ . This definition of tortuosity has been widely used. There are some other definitions of  $\tau$ :  $\tau = (L_e / L)^2$  [1],  $\tau = L_e / L$  [18] or  $L / L_e \leq 1$ . In the last case,  $\tau$  is found to be in a range 0.56 – 0.8 [1].

Several empirical correlations that suggested a relationship between tortuosity ( $\tau$ ) and porosity ( $\varepsilon$ ) have been found in the literature since the end of the nineteen century. Maxwell [19] obtained that  $\tau = 1.5 - 0.5(\varepsilon)$ , Archie [20] obtained that  $\tau = \varepsilon^{-n}$ , Weissberg [21] obtained that  $\tau = 1 - 0.5 \times \ln(\varepsilon)$ , Foscolo et al [22] obtained that  $\tau = 1 / \varepsilon$ , while Puncochar and Darhos [23] obtained that  $\tau = 1 / \sqrt{\varepsilon}$ , Boudreau [24] obtained that  $\tau = \sqrt{1 - \ln(\varepsilon^2)}$ , and also recently  $\tau = (1 + (32/9\pi)(1-\varepsilon))^2$  [25] for a model of diffusive tortuosity in marine mud. A widely used relationship, which generalizes the above mentioned forms, is proposed by Comiti and Renaud [5] and reads:

$$\tau = 1 + P \ln(1 / \varepsilon) \quad (2)$$

Equation (2) was deduced from experiments on flow through beds, made of packed spherical or cubic particles. For spherical particles  $P = 0.41$  and for cubic particles  $P = 0.63$ . Koponen et al [7], applied the Lattice Gas (LG) Cellular Automaton method to solve numerically a creeping flow of Newtonian incompressible fluid in two – dimensional porous substance constructed by randomly stacked squares. They obtained a correlation as:

$$\tau = 0.8(1-\varepsilon) + 1 \quad (3)$$

Koponen et al. have introduced the percolation threshold [26] in their LG simulations, which were carried out for porosities in the range 0.40 - 0.90. The numerical results were fitted by the following correlation:

$$\tau = 1 + 0.65 \frac{(1-\varepsilon)}{(\varepsilon-\varepsilon_c)^{0.19}} \quad (4)$$

where  $\varepsilon_c = 0.33$  is the porosity at percolation threshold.

An empirical expression the tortuosity of column particles porous medium was deduced by Mauret and Renaud [27]. The resulting equation is suitable for fibrous materials and is called the capillary model. It can be used for a high porosity porous medium with no dependence on Reynolds number. This equation can be expressed as follows:

$$\tau = 1 + 1.55 \ln(1/\varepsilon) \quad (5)$$

Our work focuses on the derivation of a tortuosity model for the flow of Newtonian incompressible fluid in a three dimensional stacked cylindrical particle porous media by using the geometrical method. The results from the model are compared with the existing experimental for column particles [27].

## 2. Geometrical model for tortuosity of streamlines in three – dimensional porous media with cylindrical particles

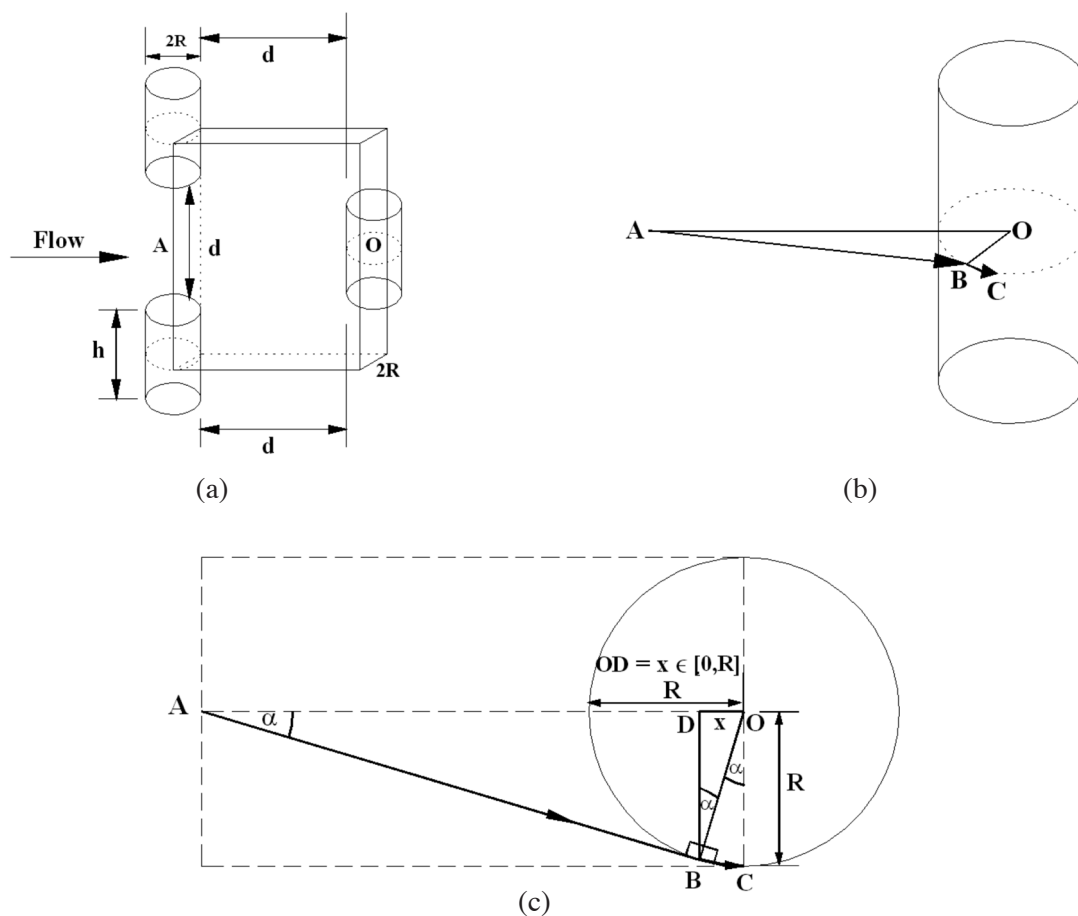
For flow in real porous media, there are many streamlines around the particles. The tortuosity of streamlines may be found by velocity field calculation in the porous medium. For simplicity we apply a geometric method by averaging over some possible streamlines around particles for tortuosity. In principle we can find the average tortuosity for the tortuous flow paths in porous media by:

$$\tau = \frac{1}{N} \sum_i \tau_i \quad (6)$$

where  $N$  is the total number of flow paths or streamlines and  $\tau_i$  is the tortuosity for the  $i^{\text{th}}$  flow path or streamline.

Figs. 2 to 5 display the possible configuration for Newtonian incompressible fluid flowing through porous media of three-dimensional cylindrical particles. Figs. 2, 3 and 4 show the particles are arranged in the form of an isosceles - triangle, while in Fig. 5 they are arranged in a square arrangement.

Fig. 2(a) depicts a three - dimensional configuration for cylindrical particles arranged in an isosceles - triangular form. Fig. 2(b) shows a typical streamline which flows around a cylindrical particle and Fig. 2(c) is a top view of the flow path of Fig. 2(b).



**Fig. 2.** (a) A three dimensional configuration for cylindrical particles arranged in an isosceles - triangle form, (b) A typical streamline flowing around cylindrical particle, (c) Top view of the flow path in Fig. 2(b).

From Fig. 2(a), the side length of the unit cell and the front length of the unit cell are equal to  $2R$  and  $d+2R$ , respectively, and the height of the unit cell is  $h+d$ , so that the total volume of the unit cell is  $V_t = (h+d).(d+2R).(2R)$ , The total pore volume in this unit cell is  $V_p = (h+d).(d+2R)(2R) - \pi R^2 h$ . Thus, for this configuration the porosity is:

$$\begin{aligned} \varepsilon = \frac{V_p}{V_t} &= 1 - \frac{\pi R^2 h}{(h+d) \cdot (d+2R) (2R)} \\ &= 1 - \frac{\pi / 4}{\left[1 + \frac{d}{h}\right] \left[1 + \frac{d}{2R}\right]} \end{aligned} \quad (7)$$

Equation (7) indicates that when  $d = 0$ ,  $\varepsilon_{min} = 1 - (\pi/4)$ . This is the minimum expected porosity when a cylinder particle layer is densely packed. The streamline AB in Fig. 2(b) is directed tangential to the particle and streamline BC flows along the surface of the particle. From Fig. 2(c), let  $OD = x \in [0, R]$  and  $OB = OC = R$ , the flow trajectory AB can be found by the relationship between two triangles ABD and ODB, then,  $l_{BC} = \sqrt{R^2 - X^2 + (d+2R-X)^2}$ . If we assume, the boundary layer is very thin ( $\ll R$ ) on the surface of cylindrical particle, then the flow path length  $l_{BC}$  approximates the BC arc length. And  $l_{BC} \approx BC$  or  $l_{BC} \approx R (\arcsin (x / R))$ . From Fig. 2(c) the actual length of flow path is defined as  $L_e^1 = (l_{AB} + l_{BC})$ . So,

$$\begin{aligned} L_e^1 &= \sqrt{(5R^2 + d^2 + 4Rd - 2xd - 4xR)} \\ &\quad + R \sin^{-1} \left[ \frac{x}{R} \right] \end{aligned} \quad (8)$$

In Fig. 2(b), the actual length of series of flow paths is:

$$\int_0^R L_e^1 dx = \int_0^R \left[ \sqrt{(5R^2 + d^2 + 4Rd - 2xd - 4xR)} + R \sin^{-1} \left[ \frac{x}{R} \right] \right] dx \quad (9)$$

According to the tortuosity definition in equation (1), we obtained the tortuosity for Fig. 2 as:

$$\tau_1 = \frac{\int_0^R L_e^1 dx}{l_{AO}} = \frac{\int_0^R L_e^1 dx}{(d + 2R)} \quad (10)$$

$$\begin{aligned} \tau_1 &= \frac{(5R^2 + d^2 + 4Rd)^{3/2}}{(2d + 4R)(d + 2R)} - \frac{(R^2 + d^2 + 2Rd)^{3/2}}{(2d + 4R)(d + 2R)} \\ &\quad + \frac{R^2}{(d + 2R)} \left[ \frac{\pi}{2} - 1 \right] \end{aligned} \quad (11)$$

In Fig. 3, the tortuosity is controlled by flow path ABCD. Therefore, the actual length of flow paths is  $l_{AB} + l_{BC} + l_{CD}$ . The corresponding straight length of flow paths is  $l_{AB} + l_{BF} + l_{FE}$ , so the tortuosity is :

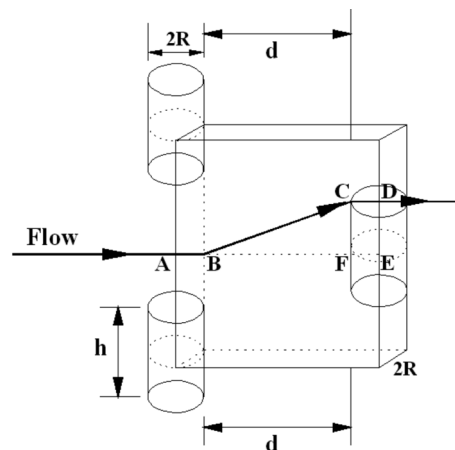
$$\tau_2 = \frac{l_{AB} + l_{BC} + l_{CD}}{l_{AB} + l_{BF} + l_{FE}} \quad (12)$$

where  $l_{AB} = l_{CD} = l_{FE} = R$ ,  $l_{BF} = d$ ,  $l_{CF} = h/2$  and  $l_{BC} = \sqrt{l_{BF}^2 + l_{CF}^2}$  or  $\sqrt{d^2 + \frac{h^2}{4}}$ , so the tortuosity is :

$$\tau_2 = \frac{\left[ \sqrt{d^2 + \frac{h^2}{4}} \right] + 2R}{(d + 2R)} \quad (13)$$

$$\tau_2 = \frac{s + \sqrt{p^2 + \frac{1}{4}}}{(s + p)} \quad (14)$$

where  $p = d/h$  and  $s = (2R/h)$



**Fig. 3.** The same three dimensional configurations for cylindrical particles arranged in an isosceles – triangular form as in Fig. 2, but for which the tortuosity is controlled by another kind of flow path.

In Fig. 4, the tortuosity is controlled by flow path ABCD but it is different from Fig. 3. Therefore, the actual length of flow paths is  $l_{AB} + l_{BC} + l_{CD}$ , and the corresponding straight length of flow paths is  $l_{AB} + l_{CD}$ , so the tortuosity is:

$$\tau_3 = \frac{l_{AB} + l_{BC} + l_{CD}}{l_{AB} + l_{CD}} \quad (15)$$

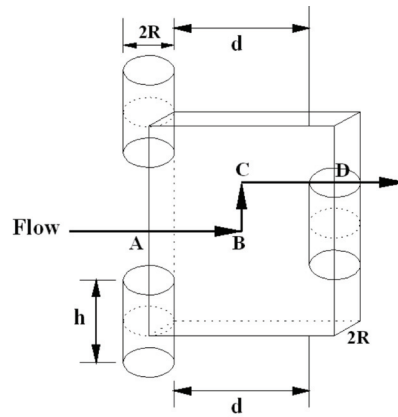
where  $l_{AB} = l_{CD} = R + (d/2)$ ,  $l_{BC} = h/2$  and  $p = d/h$  and  $s = (2R/h)$  so the tortuosity is :

$$\begin{aligned} \tau_3 &= \frac{R + d/2 + h/2 + R + d/2}{d + 2R} \\ &= \frac{d + 2R + h/2}{d + 2R} \end{aligned} \quad (16)$$

$$\tau_3 = 1 + \frac{1}{2(s+p)} \quad (17)$$

Now we can obtain the average tortuosity for flow paths/streamlines in three dimensional porous media with cylindrical particles arranged in an isosceles - triangle form by averaging over equation (11), (14) and (17).

$$\tau_3 = \frac{\tau_1 + \tau_2 + \tau_3}{3} \quad (18)$$

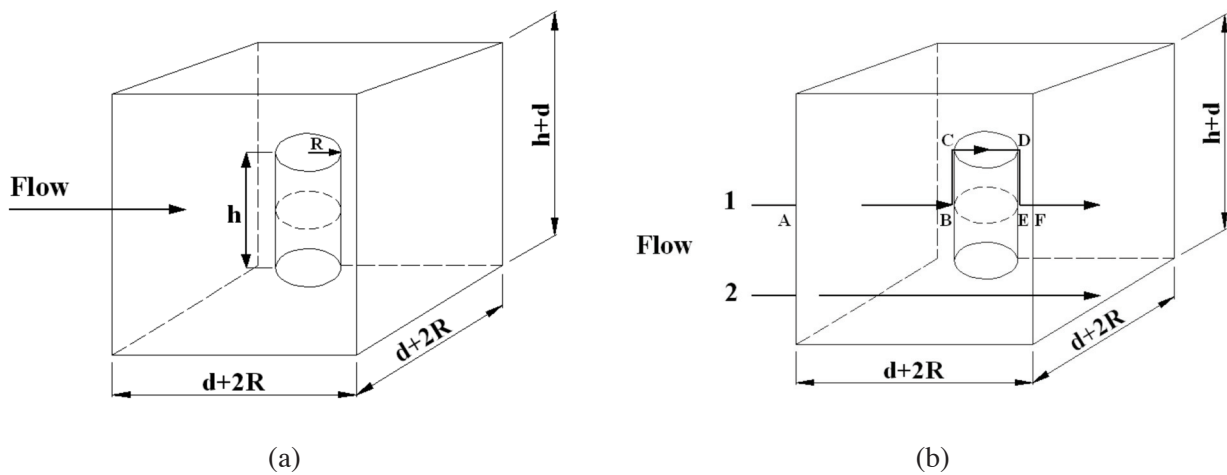


**Fig. 4.** The same three dimensional configuration for cylindrical particles arranged in an isosceles – triangular form as in Fig. 2, but for which the tortuosity is controlled by a third kind of flow path.

Another configuration for particle arrangement in this work is considered when the particles are arranged in a square form. Fig. 5(a) displays the unit cell when cylindrical particles are arranged in a square form and Fig. 5(b) shows the two representative flow paths. The side length of the unit cell as shown in Fig. 5(a) and 5(b) is  $d + 2R$ , the front length of the unit cell is  $d + 2R$ , and the height of the unit cell is  $h + d$ . So the total volume of the unit cell is  $V_l = (h + d) \cdot (d + 2R)^2$ , and the total pore volume in this unit cell is  $V_p = (h + d) \cdot (d + 2R)^2 - \pi R^2 h$ . Thus, for the Fig. 5 the porosity is:

$$\begin{aligned} \epsilon &= \frac{V_p}{V_l} = 1 - \frac{\pi R^2 h}{(h + d) \cdot (d + 2R)^2} \\ &= 1 - \frac{\pi / 4}{\left[ = 1 + \frac{d}{h} \right] \left[ = 1 + \frac{d}{2R} \right]^2} \end{aligned} \tag{19}$$

Equation (19) also leads to  $\epsilon_{min} = 1 - (\pi/4)$  when  $d = 0$  as expected.



**Fig. 5.** (a) Three dimensional configuration for cylindrical particles arranged in a square form (b) two typical streamlines flowing around cylindrical particles.

For the streamline 1 in Fig. 5(b), since the boundary layer thickness is very thin on the surface of particle then we have:

$$\tau_{4.1} = \frac{l_{AB} + l_{BC} + l_{CD} + l_{DE} + l_{EF}}{l_{AB} + l_{CD} + l_{EF}} \quad (20)$$

$$\tau_{4.1} = \frac{d/2 + h/2 + 2R + h/2 + d/2}{d/2 + 2R + d/2} = \frac{d+h+2R}{d+2R} \quad (21)$$

$$\tau_{4.1} = 1 + \frac{h}{d+2R} \quad (22)$$

For the streamline 2 in Fig. 5(b), the actual length is equal to the straight length of flow path in the unit cells, so its tortuosity is equal to 1.

$$\tau_{4.2} = 1 \quad (23)$$

It can be expected that the streamline is tortuous near the particle and is almost straight far away from the particle. It is also expected that the fraction of the straight flow path increases when increasing the porosity. Because the fraction of the tortuous flow path decreases with increasing porosity, we take the weighted average tortuosity ( $\tau_{weight}$ ) as follows:

$$\begin{aligned} \tau_{cyl2} &= \frac{(2R)^2}{(d+2R)^2} \tau_{4.1} + \left[ 1 - \frac{(2R)^2}{(d+2R)^2} \right] \tau_{4.2} \\ &= \frac{(4R)^2}{(d+2R)^2} \tau_{4.1} + \left[ 1 - \frac{(4R)^2}{(d+2R)^2} \right] \tau_{4.2} \end{aligned} \quad (24)$$

$$\tau_{cyl2} = 1 + \frac{s^2}{(p+s)^3} \quad (25)$$

where  $p = d/h$  and  $s = (2R/h)$ .

Now, we can obtain the averaged tortuosity of flow path of streamlines in three dimensional porous media with cylindrical particles arranged in different configurations by averaging over equation (18) and (25):

$$\tau_{cyl} = (\tau_{cyl1} + \tau_{cyl2}) \quad (26)$$

Equation (26) is an approximate expression for tortuosity of flow paths or streamlines in three dimensional porous media with cylindrical particles. The proposed model looks some what complicated, while it is helpful for understanding the physical mechanism for tortuosity of tortuous flow paths or streamlines in three dimensional porous media with cylindrical particles.

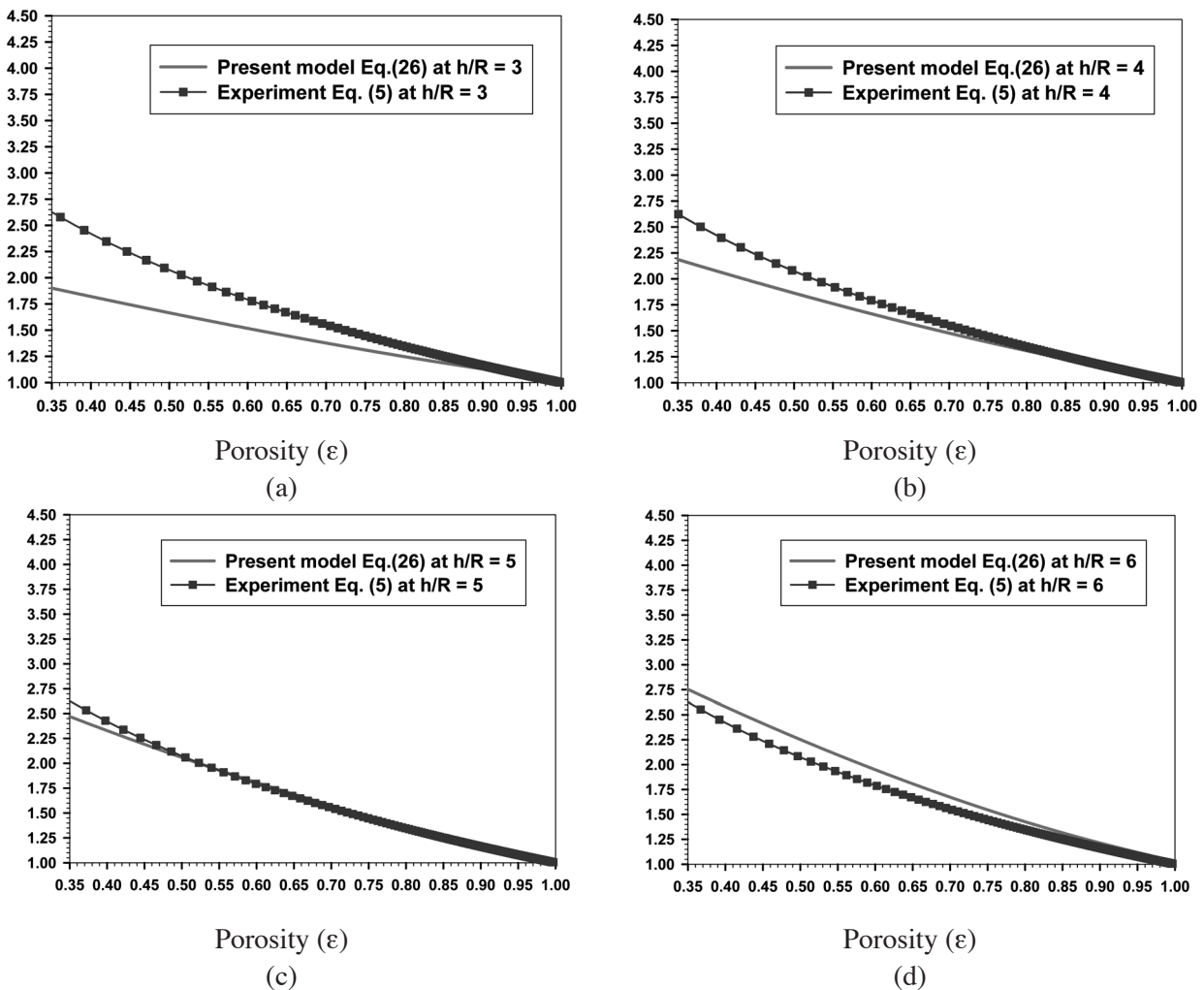
### 3. Results and discussion

Fig. 6 presents the comparison between the present model prediction (Equation 26) for cylindrical particles and the available empirical correlation (Equation 5) at different particle height to radius ratio ( $h/R$ ). It can be seen that the present model presents good agreement with the existing experimental correlation. The comparison results show that for some conditions, the experimental correlation seems to be different from the present model prediction because to the experimental correlation was fitted from much experimental data, or it is the average from the representative experiment results.



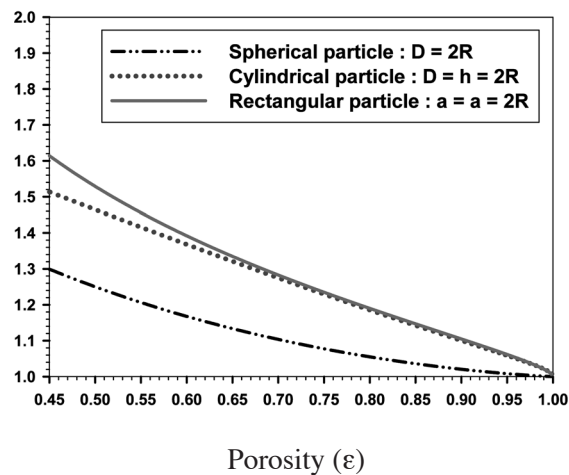
However, the comparison shows the validity of the present model for the tortuosity of flow paths or streamlines in three dimensional porous media with cylindrical particles. Through experimental correlation looks simple, it is also worth pointing out that the correlation Equation (5) allows the porosity to approach zero, which is inconsistent with practical situations with cylindrical particles because for a porous medium with cylindrical particles, which can be found by assuming  $d = 0$  (meaning : particles touch each other), the minimum porosity for this case is  $1 - \pi/4 \approx 0.21$ .

Therefore, the experimental correlation Equation (5) for cylindrical particles may present unreasonable results at low porosity. The overall agreement is better at large  $h/R$  values. At low  $h/R$  values, for low porosity, the disagreement between the present model and Eq. (5) might originate from the diverging behavior of Eq. (5) when  $\varepsilon \rightarrow 0$ .



**Fig. 6.** A comparison between the present model prediction for cylindrical particles and the available experimental correlation Equation (5) (a)  $h/R = 3$ , (b)  $h/R = 4$ , (c)  $h/R = 5$  and (d)  $h/R = 6$ .

Fig. 7 presents a comparison among the tortuosity models for spherical particle [Appendix A, 11], rectangular particle [Appendix B, 11] and the present model for cylindrical particle at the same particle sizes. The sphere stacking results in the lowest tortuosity while rectangle and cylinder stackings only differs at low porosity.



**Fig. 7.** Comparisons among the tortuosity models for spherical, rectangular, and cylindrical particles.

#### 4. Conclusions

We have proposed an analytical model for tortuosity of streamlines in a three dimensional porous material of the fuel cells electrode, especially for thin film electrode of solid oxide fuel cells. The arrangement of the particles that form the electrodes as the cylindrical particles stack are only arranged together in the vertical direction. The model is derived assuming and averaging over some possible streamlines around the particles and for the different particle configuration arrangements and focuses only on the flow of a streamline passing the vertical stack cylinders. The proposed model is a function of the particle sizes and distance between particles. The model predictions are in good agreement with the available experimental empirical laws, especially at high cylinder height to diameter ratios. There is agreement on the thin film electrode characteristics of solid oxide fuel cells. So, it might be helpful to understand the physical mechanism of tortuosity of tortuous streamlines in thin film solid oxide fuel cell electrodes.

#### 5. Appendix A :

Tortuosity model for three dimensional porous media with spherical particles [11]:

$$\tau_1 = \frac{\left( \begin{array}{l} 6\sqrt{3p^2 + 12p + 8} \\ - (3p^2 + 12p + 4) \arcsin \frac{2}{\sqrt{3}(p+2)} \\ - 6(p+2)^2 \arcsin \frac{-2}{\sqrt{3p^2 + 12p + 8}} \end{array} \right)}{8\sqrt{3}(p+2)} \quad (\text{A1})$$

$$\tau_2 = 1 + \frac{\pi - 2}{\frac{4\pi}{3(1-\varepsilon)} + \left[ \frac{4\pi}{3(1-\varepsilon)} \right]^{1/3} - 2 \left[ \frac{4\pi}{3(1-\varepsilon)} \right]^{2/3}} \quad (\text{A2})$$

$$\tau = (\tau_1 + \tau_2) / 2 \quad (\text{A3})$$

where  $p = \frac{d}{R} + \frac{2}{3} \sqrt{\frac{\sqrt{3}\pi}{1-\varepsilon}} - 2$ ,  $d$  is the distance or gap size between particles, and  $R$  is particle radius

## Appendix B :

Tortuosity model for three dimensional porous media with rectangular particles [11]:

$$\tau = \frac{11}{12} + \frac{1}{24} \left( \frac{\sqrt{1+4p^2} + \sqrt{s^2+4p^2}}{p} + \frac{1+p+ps+s^2}{(p+s)(1+p)} \right) + \frac{1+s^2-(1+s+s^2)\varepsilon}{6s} \quad (\text{B1})$$

where length, width and height of particle are  $2R$ ,  $2R$ , and  $h$  respectively, and  $d$  is distance or gap size between particle,  $s = 2R/h$  and  $p = d/h$

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