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A new approach to microstructure optimization of solid oxide fuel cell electrodes

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Abstract

Designing optimal microstructures for solid oxide fuel cell (SOFC) electrodes is complicated due to the multitude of electro-chemo-physical phenomena taking place simultaneously that directly affect working conditions of a SOFC electrode and its performance. In this study, a new design paradigm is presented to obtain a balance between electrochemical sites in the form of triple phase boundary (TPB) density and physical properties in the form of gas diffusivity in the microstructure of a SOFC electrode. The method builds on top of a previously developed methodology for digital realization of generic microstructures with different geometric properties in ionic or electronic conductor grains. The obtained realizations of SOFC electrode are then used to calculate TPB density and gas transport factor. In the next step, based on the obtained database, a neural network is trained to relate input geometrical parameters to those output properties. The results indicate that the TPB density is less sensitive to the geometry than the gas transport factor. Also, the smaller particles in the ionic and electronic conductor phase lead to a higher amount of TPB density. The presented methodology is also used to obtain the maximum feasible properties of microstructures and their related geometric characteristics for special target functions like maximum reaction sites and gas diffusivity in a realized model. The tradeoff between input and output parameters is another application of this modeling approach which demonstrates the TPB density and gas transport factor variation versus the geometric anisotropy of particles and porosity, respectively.

1. Introduction

Fuel cells offer many advantages, compared to more conventional power generation devices, including high-efficiency and low emission levels for SO_X, NO_X and CO_2 [1]. Among different types of fuel cells, solid oxide fuel cells (SOFCs) have attracted more attention primarily because of their high

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electric efficiency, high temperature exhaust heat and compactness [2]. According to many studies a composite electrode in fuel cells has a much lower over-potential compared to a single-phase electrode that is solely made of the electro-catalyst, owing to parallel pathways for electronic and ionic charge carriers [3, 4]. For example using LSM–YSZ (Lanthanum strontium manganite-Yttria stabilized Zirconia) for the cathode and Ni-YSZ for the anode, produce composite electrodes that play an important role in the development of SOFC technology as they offer a lower polarization resistance [5, 6]. The electrode's performance strongly depends on the properties of LSM, Ni and YSZ, and also more importantly on the morphological factors of the heterogeneous microstructure of these electrodes. For example, the polarization resistance (R_p) for a LSM-based cathode is directly related to the density of the three-phase boundaries (TPB's), as the primary site for electrochemical oxygen reduction [7, 8]. Similarly, the availability of a network of continuous pathways within each phase and intra-connectivity (i.e. percolation) seem to be two significant factors that affect the performance of a cell [9, 10].

These morphological parameters sometimes have inconsistent and nonlinear behaviors such that a small increase in one may result in a detrimental effect on others. For example, the tortuosity of pathways within each phase directly impacts the transport to and from the TPBs [11]. To overcome these complications, Joen et al. [12] first introduced a micro-scale model for the microstructure and performed a numerical optimization on the microstructure of the anode supported SOFC to investigated the dependence of the electrochemical reaction and the mass transport on the particle size and thickness of the functional layers [12]. A new approach was proposed by Sebdani et al. [13] to design fuel cell microstructure with the largest TPB density based on various shape of electrode particles in axial or transversal directions and their volume fractions. Since the realization of different electrode microstructures are very time-consuming and their characterization is a complex process, Riazat et al. [14], used Neural networks coupled with optimization methods to enable researchers to have a powerful search engine to find the best electrode microstructure in different scenarios. In that study a range of traditional SOFC electrodes with different porosities and geometric properties in ionic or electric phases was realized to investigate interactions among TPB density and conductivity of ions and gases in the realized models. In the present study, the mixing ratio of electronic or ionic conductor phases is added to the controllable geometric parameters of SOFC electrodes as proposed in [14]. In this way, a set of heterogeneous microstructures with different level of porosities and the presence of ion and electron conductor materials as well as the geometric anisotropies are generated using a novel Monte Carlo method which is described in detail in [15]. The active TPB density of each 3D microstructure is evaluated based on a method described in [13] using the characterization matrix of the models. Then the conductivity of voids is evaluated indirectly from the 3D realized models. To have a broader view of electrode performance, all of these input and output data are used to train a set of neural networks which can generate the domain of feasible microstructure properties and simultaneously analyze them with regard toTPB density and gas transport factor. Finally, based on the best neural networks model, a new multi-objective optimization approach is developed to explore a group of SOFC electrode models with the best constructive properties with assumed target functions.

2. Simulation methodology

The state of heterogeneity of an electrode, including the size and distribution of particles of each phase, are strongly influenced by the nucleation, growth, and initial distribution of constructive ingredients [17, 18]. In a virtual realization, heterogeneity is controlled by the nucleation and grain growth mechanisms as functions of time and morphology [13], and the state of heterogeneity can be quantified by statistical functions like the linear path function

and N-point correlation functions [17]. The Monte Carlo approach for the reconstruction of the electrode microstructure is composed of three steps: i) generation, ii) distribution, and iii) growth of the cells. In the first step, several initial seed cells are randomly placed in a unit cell of the electrode. Upon initial seed placement, the growth step starts following a cellular automation algorithm. This procedure continues till the desired volume fraction for each phase is achieved. From a different perspective, the growth step continues till all phases meet and fully occupy the grid. Penetration between the phased is avoided at all times throughout the initial distribution and growth of the cells[15].

Characterization of those realized microstructures are based on the direct investigation of the identification matrix (e.g. TPB density) or indirect evaluation of geometric properties (e.g. gas transport actor) from the 3D realized solid models as described in the following. As many studies report, e.g. see Janardhanan [18], the density of active TPBs, where the ionic conductor cluster and electric conductor are connected to each other, plays a major role in the overall microstructure of the SOFCs and significantly affects the efficiency of the SOFC. As an interface between the phases they play an essential role in the electrochemical performance and the power generation of the SOFC [18]. To evaluate this parameter in a microstructure realization a method introduced by Sebdani [13] is adopted. At first the active cluster of each phase is recognized and then the 26 neighboring voxles of each active void voxel is investigated to find the location of TPBs following an algorithm based on the calculation of overall active TPBL. Another effective parameter in the performance of SOFC electrodes is the gas diffusion, especially in high current densities. Important factors characterizing the gas diffusion in porous media are the porosity and tortuosity of the gas routes [19]. Due to its simplicity, Fick's law is commonly adopted to assess gas diffusion. For porous media, Fick's first law can be modified by introducing porous media factors as

$$D_{ij}^{eff} = \frac{\varphi}{\tau} D_{ij} \quad (1)$$

Where D_{ij} is the binary diffusivity of the gas species, D_{ij}^{eff} is the effective binary diffusivity of the gas species, and φ and τ are the porosity and tortuosity respectively. In this research, tortuosity is obtained from the effective thermal conductivity. For this purpose each section of the microstructure is converted to a 2D image so that each phase is distinguished with a different color. This image stack can be recognized as a multiphase material by a thresholding process in reconstruction software. After converting each voxel into the volume elements, using Avizo xlabTM, EFI Corporation, a temperature difference is applied between the two opposite faces of the material sample while the other faces are insulated then the thermal conductivity of the pore network, K_{eff}, is obtained by performing a thermal simulation and the tortuosity is calculated based on

$$\tau = \varphi \frac{K_{bulk}}{K_{eff}} \quad (2)$$

Where we assume K_{bulk}=1. As discussed by Zhao et al. [20] whenever the molecular distance of a gas is in the order of average pore size, the Knudsen diffusion should be directly considered. However in the current study, this effect is neglected to simplify the optimization scheme and the diffusion factor as defined regardless of the average pore size to estimate the gas transport capability in the porous electrodes by:

$$\text{Gas transport factor} = \frac{\varphi}{\tau} \quad (3)$$

Ionic and Electric Conductivity of different phases are other important electrochemical performance indicators in SOFC electrodes [21]. Since the electrode geometry is one of the main factors in determining the conductivity of ions and electrons, the modification factor of intrinsic ionic or electric conductivity in porous media can be determined by Ohm's law in the same method as the gas transport factor. In this study to simplify the optimization process, the ionic and electronic transports of material are supposed in ranges that are not rate-limiting in operational temperature and current density.

In traditional design, material and geometry are usually varied iteratively to meet design requirements. A more efficient approach to design requires simultaneous material and geometry optimization. Following this approach for the design of material, a design area is needed which consists of a set of possible microstructures existing within a region characterized by certain distribution functions obtained from the microstructure. In the other word, that restricted area or volume includes all possible effective property values predicted by sweeping input parameters defining these distribution functions, and generally a solution is a subset of this closure. Although this may be judged as an over simplistic approach, it allows us to demonstrate the concepts. Mathematically speaking, a property closure for these parameters can be obtained using an arbitrary analytical or approximate method and their boundaries represent constraints for the optimization process [16, 22]. TPB density and gas diffusivity are those critical design objectives in that design area.

In this study as described in Table 1, the realization of 3D models are carried out by choosing four levels of porosity within the range of 28-37% with eight levels of volume fractions between 25.5-36% for the first solid phase (electron conductor phase) and 31-42% for the second solid phase (Ion conductor phase). The growth rate of grains in transversal (VZ) and axial (VXY) directions (related to the electrolyte) for those phases are chosen in five levels in the range of 0.2-0.6. A 3D solid model forming one of those realized microstructures is shown in Fig. 1.

Following these input specifications and their combinations, 100 different microstructures from the feasible combination of these parameters are selected and mathematically characterized. The TPB density and gas diffusion factor of void space are evaluated

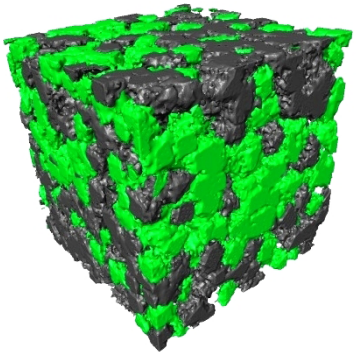


Fig. 1. A sample 3D solid model form of the realized micro-structures.

for each microstructure. Using these input and output parameters, as shown in Fig. 2, a set of neural networks are trained to predict the properties of the microstructures for a given set of input parameters.

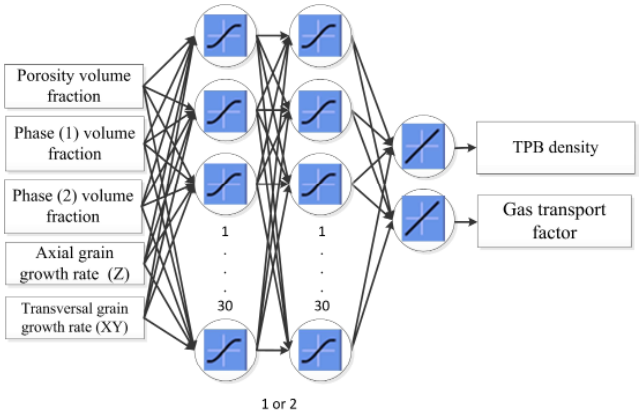


Fig. 2. Different structures of neural network tested.

In the current study, the extrapolation input and output parameters are carefully limited to account for the complex behavior of fuel cells. Because of the regressive nature of the problem, a back propagation neural network (BPNN) using the levenberg marquardt algorithm is used based on five inputs (porosity, phase 1&2 volume fractions, and axial and transversal growth rate of those solid phases) and two normalized

Table 1. Input parameters for microstructure realization

Porosity (%)	28	31	34	37			
Phase 1 Volume fraction (%)	25.5	27	28.5	30	31.5	33	34.5
Phase 2 Volume fraction (%)	31.5	33	34.5	36	37.5	39	40.5
Axial growth rate (Z)	0.2	0.3	0.4	0.5	0.6		
Transversal growth rate (XY)	0.2	0.3	0.4	0.5	0.6		

outputs (TPB density and gas diffusion factor) This process was performed in the Neural Network Toolbox provided in MATLAB™.

This best model with minimum Mean square error (MSE) consists of an input layer, one hidden layer with 20 neurons and an output layer. The value of the minimum mean square error (MSE) and the regression for the validation data obtained are 9.936e-5 and 0.9758, respectively. Using that well-trained neural network, a design space for microstructures can be obtained as shown in Fig. 3, This space can be a useful tool to analyze the results and also to perform an optimization process to find the best geometric parameters in the microstructures for different working conditions and intrinsic property of materials. In this study, each parameter is divided into ten levels in the range of the pre-realized microstructures. Based on these input parameters, the properties of those microstructures (~100000) are predicted by the neural network to form a design space for microstructures.

3. Results and Discussion

That well trained neural network is used to simulate the overall relationship between the microstructure and its properties within a limited range of input parameters. In this step, among those hypothetical microstructures, 99090 models are located in the acceptable range in the design space. The range of geometric input parameters and predicted output variables are shown in Table 2.

Based on the obtained properties, it is clear that the

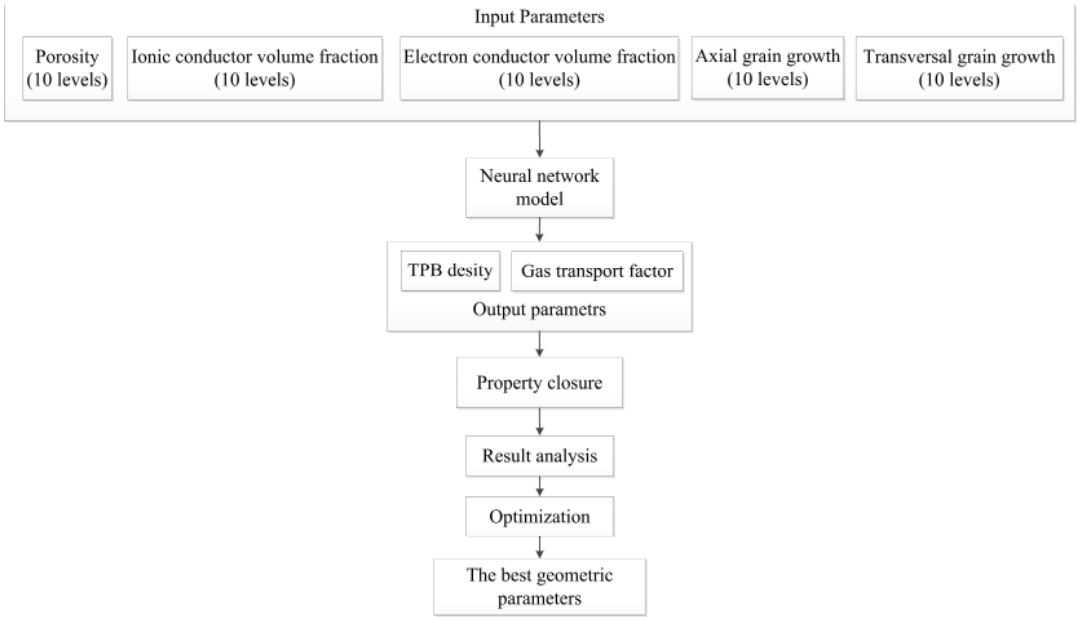


Fig. 3. Schematic diagram showing different sections of the study and their relationship.

Table 2. The range of geometric input parameters and predicted output variables.

		minimum	maximum	variation
input	Porosity volume fraction	0.2	0.4	0.2
	Phase 1 Volume fraction	0.2	0.4	0.2
	Phase 2 Volume fraction	0.2	0.4	0.2
	Axial grain growth rate (Z)	0.2	0.6	0.4
	Transversal grain growth rate (XY)	0.2	0.6	0.4
output	TPB density (L μm^{-2})	1.10E-04	0.029	0.029
	Gas transport factor	3.57E-04	0.183	0.183

gas diffusion factor is 6 times more sensitive to the geometry than the TPB density with our assumptions. Also in agreement with Sebdani et al. [13], TPB density is significantly affected by the volume fractions of phases and microstructural attributions. Although the exact relationship between the growth rates in axial or transversal directions with TPB density is rather complex, the average TPB density of realized microstructures was similarly enhanced up to 5 times, based on the anisotropy of particles in the microstructures. In addition, the variation of gas diffusion factor was obtained in the range of 0.18, approximatelythe same range of this parameter in [14]. A sample property closure obtained from the predefined input parameters is shown in Fig.4. The vertical axis is dedicated to the Pore (gas) transport factor and the horizontal axis demonstrates the TPB density (L is the length of voxel edge in nm). The position of the microstructure represents their feasible properties within the selected design space. The region of interest in the design area can be distinguished from the property closure by setting a target function. This target function is a linear (or nonlinear) combination of the microstructure properties that depends on the rate-limiting properties and their interactions in a real microstructure. For example, a higher amount of TPB density is useful when the ionic and electronic conductivity is enough to transport the ions of the electrochemical reactions [23]. Those conductivities, in addition to the geometry, depend on the intrinsic conductivity of the materials and temperature. Another example can be related to the diffusivity of reactant gas into the electrode, which can be a rate limiting phenomena in high current densities in the electrode, and a large number of TPBs and higher ionic conductivity that cannot compensate the limitation of the gas diffusions in the performance of the cell [24]. As a result, the target function should be estimated by some other in-process parameters like temperature and current density that are independent of the geometric investigations.

Based on the preferred electrochemical and physical properties of the microstructure, which result in the highest electrochemical sites or highest reactant gas transport, only a limited number of these microstructures can be chosen. For example, if the highest level of reaction sites (TPB density) is the rate limiting phenomenon in determining current density and temperature, the microstructures which are located in zone(1) are the targets and their geometric properties are suitable for highest electrochemical reactions. Also, when the reactant gas diffusivity restricts the reactions of the electrode, microstructures located in zone(2) would have the best performance in the electrode.

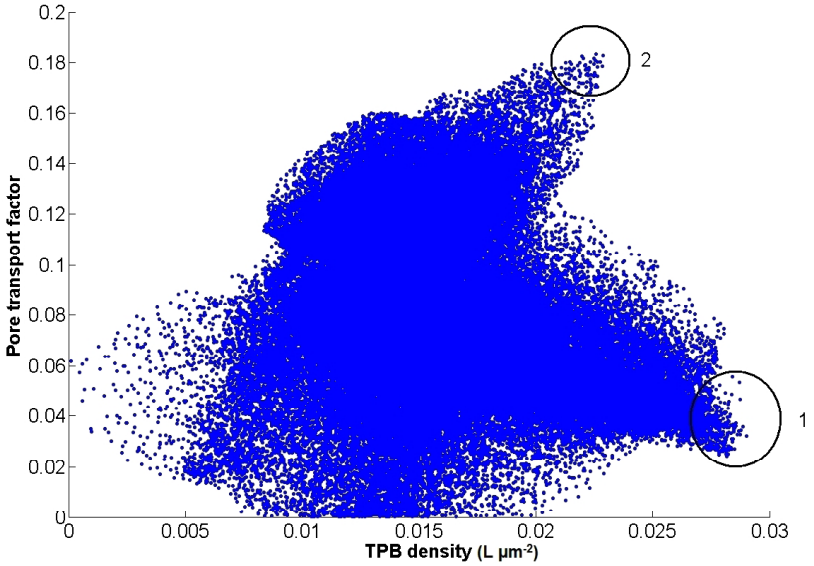


Fig. 4. Feasible properties for the realized microstructures with different geometric parameters.

From a different viewpoint, the target function can be illustrated by a restricted region in the design space which separates the desirable microstructures from the rejected ones. For example, if the target function is considered a linear function, the critical boundary is in the form of a curve that intercepts each axis based on the coefficient of each variable in the target function. The intersection between that curve and the design space is a border for decision making that divides the accepted or rejected microstructures. The enclosed area between the design space boundaries and target function curve contains the appropriate microstructures. If there is no overlap between these restricted spaces, this means that a microstructure with the preferred properties does not exist due to geometric limitation. If there is an overlapping area, an optimization method can be used to explore the corresponding inputs parameters of these optimum microstructures. In this study, a number of models located in the 1% upper band of TPB density and gas transport factor are listed in Table 3. As reported in that table, the porosity should be set near to the minimum range to achieve the maximum TPB density; also the grain growth rates are in the lowest level to form the small particles in the microstructures. On the other hand, the volume fraction of phase 1 is obtained in a higher level than phase 2.

The maximum gas transport factor is achieved when the porosity is in the highest level and the volume fractions of the solid phase are similar. In that case, the grain growth rates, especially transversal one, are obtained in a higher level. The last target function is defined as the normalized combination of TPB density and gas transport factor. The model parameters are similar to the gas transport target because of the higher sensitivity of the gas transport factor than the TPB relate to the geometric parameters.

Restrictions in micro-manufacturing methodsdid not allow the researcher to generated these realized models and characterize them, but in [13] and [14] the best geometric properties for traditional microstructure of electrodes are evaluated via simulation methods similar to this study. For example, in [13] the greatest TPB density was obtained when the volume fractions are 26%, 33% and 41% for Ni, YSZ and pores, respectively. A traditional electrode microstructure in [14], with 23% porosity, axial growth rate of 0.02, and transversal growth rate of 0.1 in solid phases was predicted which could simultaneously reach the highest level of ionic conductivity, TPB density, and gas diffusion factor. Although the general

Table 3. List of optimum properties for microstructures with different target functions

	Input parameters					Output parameters	
	Porosity	Phase1 volume fraction	Phase2 volume fraction	Axial grain growth rate (Z)	Transversal grain growth rate (XY)	TPB density (L μm^{-2})	Gas transport factor
Maximum TPB	0.263	0.326	0.2	0.2	0.2	0.028862	0.035815
	0.263	0.347	0.2	0.2	0.2	0.029005	0.032217
	0.263	0.347	0.2	0.2	0.242	0.028735	0.032723
	0.263	0.347	0.2	0.242	0.2	0.028777	0.034369
	0.305	0.389	0.284	0.2	0.2	0.028744	0.046669
	0.305	0.389	0.305	0.2	0.2	0.028727	0.053508
Maximum Gas transport	0.389	0.389	0.389	0.284	0.536	0.022032	0.181746
	0.389	0.389	0.389	0.284	0.578	0.021979	0.182417
	0.389	0.389	0.389	0.326	0.536	0.022485	0.181732
	0.389	0.389	0.389	0.326	0.578	0.022572	0.183268
	0.389	0.389	0.389	0.368	0.578	0.022864	0.18273
Maximum TPB +Maximum Gas transport (Normalized)	0.389	0.389	0.368	0.326	0.578	0.022759	0.180297
	0.389	0.389	0.389	0.326	0.578	0.022572	0.183268
	0.389	0.389	0.389	0.368	0.578	0.022864	0.18273

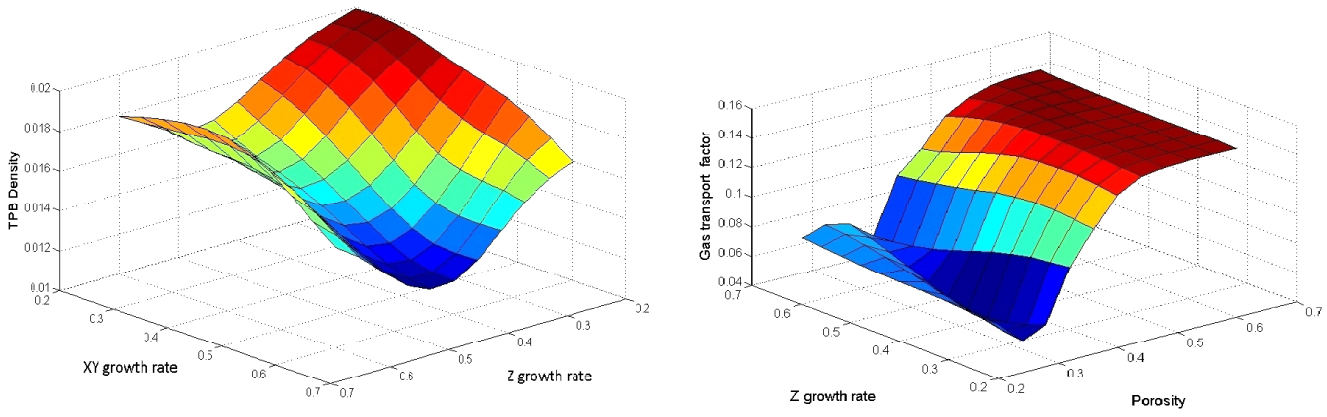
assumptions in those researches are somewhat different, similar patterns can be observed in certain areas, like the effect of porosity on the TPB density, when other phases have the same volume fractions.

In addition to the capability of finding optimum properties in the microstructures, the overall behavior of output geometric properties can be determined with regard to their constructive parameters. For example, Fig. 5-a demonstrates the variation of TPB density versus grain growth rates in axial and transversal directions when the porosity and volume fractions are set to the 0.33. As shown in that figure, the smaller grains will lead to the higher values of TPB density. Fig. 5-b demonstrates the gas transport factor versus the porosity and axial growth rate of particles in the microstructure when the volume fractions and transversal growth rate are set to the 0.33. The porosity has a clear positive effect on gas diffusivity; but being directional along the Z direction, it has a moderated impact on the gas transport capability of the microstructure especially in highly porous models.

4.Conclusions

The microstructural attribution of conventional electrodes can significantly alter the performance of a SOFC device. The main factors that affect the electrochemical performance of electrodes are TBP density, fluid diffusivity and the capability of ion and electron to transport in the electrodes to the electrolyte or current collectors. These parameters are linked with the porosity, volume fractions of each solid phase, and the 3D microstructural attributions those particles.

In this study, a number of 3D microstructure realizations were carried by varying the volume fractions, grain shapes and porosities to cover a wide range of possible isotropic and anisotropic configurations. The TPB density was calculated in those models using a developed algorithm. The stack of cross-section images from virtual microstructures was also used create 3D models to obtain the conductivity of material and gas transport factor. A combination of input and output parameters was suggested to train a neural network. A property closure was developed, containing a large number (~99090) of hypothetical microstructures, to obtain a possible microstructure for a range of input geometric parameters with a higher TPB density and gas transport factor. It was observed that the lowest rate of grain growth in particles leads to the highest level of TPB density when the porosity is near to the minimum range (26%). Also, when the volume fractions of two solid phases are similar to the higher transversal grain growth, the pore transport factor and its combination with TPB density would reach



(a) **(b)**

Fig. 5. a) The variation of TPB density versus axial and Transversal growth rates, b) and the gas transport factor versus axial growth rate and Porosity.

the maximum. Additionally, the overall behavior of output parameters can be reported versus various geometric parameters of the realized microstructures using the neural network model, the proposed method was deemed useful to design specific microstructures of material, and also to determine the limitation of obtainable performance from those experimental works. The findings of this study can be used as guidelines for future experimental investigations based on new manufacturing methods like 3D printing and additive manufacturing rapid prototyping methods.

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