Electrochemical and Solid-State Letters, **12** (2) S2-S3 (2009)
1099-0062/2008/12(2)/S2/2/$23.00 © The Electrochemical Society

Comment on "A Look at the Multiphase Mixture Model for PEM Fuel Cell Simulations" [*Electrochem. Solid-State Lett.*, 11, B132 (2008)]

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 [DOI: 10.1149/1.3041157] Manuscript received June 2, 2008. Published December 11, 2008.

The article of Gurau et al.[1] discusses the multiphase mixture (M^2) model developed by Wang and co-workers[2-6] over the last 15 years for the modeling of multiphase flow through porous media with liquid-vapor phase change. I would like to offer the following comments.

1. All published works on PEM fuel cell (PEMFC) simulations, based on either the classical multifluid approach (e.g., Ref. 19, 20, and 22 in Ref. 1 and numerous others available in more recent literature) or the M^2 model, use two-phase Darcy's law as the momentum equation for individual phase motion, which can be specifically written as

$$u_k = -\frac{k_r K}{\mu_k}(\nabla p_k - \rho_k g) \qquad [1]$$

As long as Eq. 1 is used as the momentum equation, Gurau et al. correctly proved, like numerous other researchers in the past 15 years, that the M^2 model is mathematically exactly equivalent to the classical multifluid model based on Eq. 1. That is to say, there has been neither "misleading" nor "incorrect promotion." The M^2 model has never claimed an equivalency to any models that do not invoke Eq. 1 as the momentum equation. It should also be noted that the mathematical equivalence of the M^2 model to the classical multifluid model based on Darcy's law has been reviewed and used by many electrochemists and mathematicians in works specifically related to fuel cells, e.g., Weber and Newman,[7] Birgersson et al.,[8] Mazumder and Cole,[9] You and Liu,[10] and Bridge and Wetton,[11] to name just a few. Apparently Gurau et al.[1] are neither aware of nor understand these prior publications.

2. The two-phase momentum equations presented in Ref. 1, i.e., Eq. 3a and 3b, differ from all those used in the published literature of PEMFC modeling, including even those based on the classical multifluid approach (e.g., Ref. 19, 20, and 22 in Ref. 1). Should the two-phase momentum equation be modified from Eq. 1 in the presence of phase change in a multifluid model? One can easily find the answer by consulting standard textbooks and classical works on drying of porous materials (Whitaker[12,13]), boiling in nuclear reactor debris beds (Dhir[14]), geothermal systems (Bodvarsson et al.[15]), and two-phase flow in porous media with heat transfer (Kaviany[16]), all of which involve strong phase change effects. Without exception, these textbooks and seminal works either rigorously developed or applied Eq. 1 rather than Eq. 3a and 3b in Ref. 1. For example, using the method of volume averaging, Whitaker[12,13] rigorously derived Eq. 1 as the momentum conservation equation (i.e., Eq. 231 and 232 in Ref. 8) along with a mass conservation equation (i.e., Eq. 156 in Ref. 8) which apparently features a mass source/sink term and considers phase change. Similarly, an entire chapter on phase change in the book of Kavinay[16] uses Eq. 1 as the two-phase momentum equation rather than Eq. 3a and 3b in Ref. 1.

3. A new momentum equation for two-phase flow through porous media with phase changed was presented in Ref. 1 without mathematical derivation or a single reference. Here it can be easily shown that this equation is fundamentally flawed. According to Ref. 1 and 17, the so-called new term, i.e., Group (II) in Eq. 3 of Ref. 1 or the last term in Eq. 17 of Ref. 17, is written as $\dot{m}_k u_k$ for phase k (k = gas or liquid phase). For the sake of argument, let us assume a steady system such that $\dot{m}_k = \nabla \cdot (\rho_k u_k)$ from the continuity equation for phase k. Then the new term is nothing but the inertia term, $\nabla \cdot (\rho_k u_k) u_k$, which is well known to be negligible as long as the Reynolds number based on pore diameter is less than 10; see numerous textbooks for this discussion.[16,18,19] Gurau et al. failed to recognize the simple fact that their newly suggested term is the traditional inertia term exhaustively discussed in the literature of flow through porous media. Here the Reynolds number based on pore size as characterized by $K^{1/2}$ is mathematically defined as

$$Re = \frac{u\sqrt{K}}{\nu}$$

For the catalyst layer and gas diffusion layer (GDL) of a PEMFC, $K^{1/2} \sim 10^{-6}$ m or smaller, the fluid velocity is $\sim 10^{-2}$ and 10^{-5} m/s for gas and liquid,[20-22] and the kinematic viscosity is $\sim 2 \times 10^{-5}$ and 4×10^{-7} m^2/s for gas and liquid, respectively. Thus, the Reynolds number is equal to 5×10^{-3} and 2.5×10^{-5} for gas and liquid flow, respectively, well within the validity range of Darcy's law. Yi and Nguyen[20] showed that even for interdigitated flowfield the gas velocity in GDL is of the order of 10^{-2} m/s. Obviously, the inertia term is quite negligible and the original momentum equation based on Darcy's law holds very true for PEMFC modeling. Gurau et al. did not understand why Darcy's law without inertia terms was chosen by pioneering modelers (e.g., Ref. 4 and 20-22) for the modeling of flow through porous layers of fuel cells over a decade ago.

Alternatively, one can estimate the ratio of this new term to the original Darcy's term. That is

$$\frac{\textit{Group II}}{\text{Darcy term}} = \frac{\nabla \cdot (\rho_k u_k)}{\mu_k / K} \propto \frac{u_k K / L}{\nu_k} = Re\frac{\sqrt{K}}{L} \sim 10^{-4} \text{ or } 10^{-5}$$

where L is a macroscopic length scale such as the GDL thickness, so the last term is the ratio of the pore size to a macroscopic length, which is at least $O(10^{-1})$ (for instance, for a PEMFC GDL, the pore diameter is ~ 20 μm and the minimum macroscopic length scale is the GDL thickness equal to ~ 200 μm). Again, the new term of Gurau et al. is easily shown to be negligible or irrelevant in fuel cell modeling.

4. Other comments provided in Ref. 1 are baseless, and hence no rebuttal can be provided at this time.

In summary, the two-phase Darcy's law is valid as the momentum equation to describe multiphase flow through porous layers of a fuel cell, and M^2 model is mathematically equivalent to the classical multifluid model based on two-phase Darcy's law. The new model suggested by Gurau et al. is fundamentally flawed, and all claims made by Gurau et al.[1,17] are false and inappropriate.

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