Simulation of Catalyst Layer Formation

Toshihisa Munekata[1,*], Joji Tomiyasu[2],
Takahisa Suzuki[1], Atsushi Ida[2],
Shunsuke Yamakawa[1], Ryoji Asahi[1],
Makoto Fujiuchi[2], and Yu Morimoto[1]
[1]Toyota Central R&D Labs., Inc., Nagakute, Aichi, Japan
[2]TOYOTA Motor Corp., Toyota, Aichi, Japan

INTRODUCTION

Catalyst layers (CLs) of polymer electrolyte fuel cells (PEFCs) are commonly prepared through a drying process of a colloidal ink containing platinum-loaded carbon particles, a polymer electrolyte and a solvent. While this process determines the structure of CLs and, as a result, their performance, most studies to date were only based on trial-and-error and very few theoretical studies have been done. In this study, we have developed simulation of this process to reveal controlling factors.

SIMULATION METHOD

To simulate the colloidal suspension flow during drying, a simplified solvent evaporation is modeled firstly [1]. In this model, chosen influential factors are, the gas-liquid interface dynamics, which induce pressure gradients in colloidal solution, and the polymer concentration on the particle surface, which causes viscosity change [2]. This solvent evaporation model is combined with a fluid particle dynamics method [3] and a two-phase (vapor-liquid) lattice Boltzmann method [1]. In this simulation, we include short-range inter-particle interaction affecting the initial distribution of particles, and three long-range inter-particle hydrodynamic interactions: viscosity of the polymer-containing solvent, surface tension between the gas and liquid phases, and the evaporation rate of the solvent.

RESULTS AND DISCUSSION

Figure 1 shows a simulated drying process forming a porous layer with polymer-coated carbon particles. Figure 2 shows resultant dried structures: a higher viscosity of the polymer-containing solvent gives a structure with more dispersed particles (Fig. 2(a)(b)), a higher surface tension leads to a structure with more agglomerated particles (Fig. 2(c)(d)), and a faster evaporation rate results in a more dispersed structure (Fig. 2(e)(f)). These simulations show that the three long-range inter-particle hydrodynamic forces interact with one another, and that the balance of the forces determines the pore structure. Initial particle distribution, i.e. uniform or pre-agglomerated, also strongly affects the final structure, i.e. pre-agglomeration leads to more agglomerated particle structure. Finally, the effects of the viscosity, surface tension, and evaporation rate on the dried structures are analyzed in terms of fingering phase-diagram [4].

References
[1] T. Munekata, T. Suzuki, S. Yamakawa, and R. Asahi, Phys. Rev. E (submitted).
[2] T. Okuzono, and M. Doi, Phys. Rev. E 77 (2008) 030501.
[3] H. Tanaka, and T. Araki, Phys. Rev. Lett. 85 (2000) 1338-1341.
[4] R. Lenormand, E. Touboul and C. Zarcone, J. Fluid Mech. 189 (1988) 165-187.
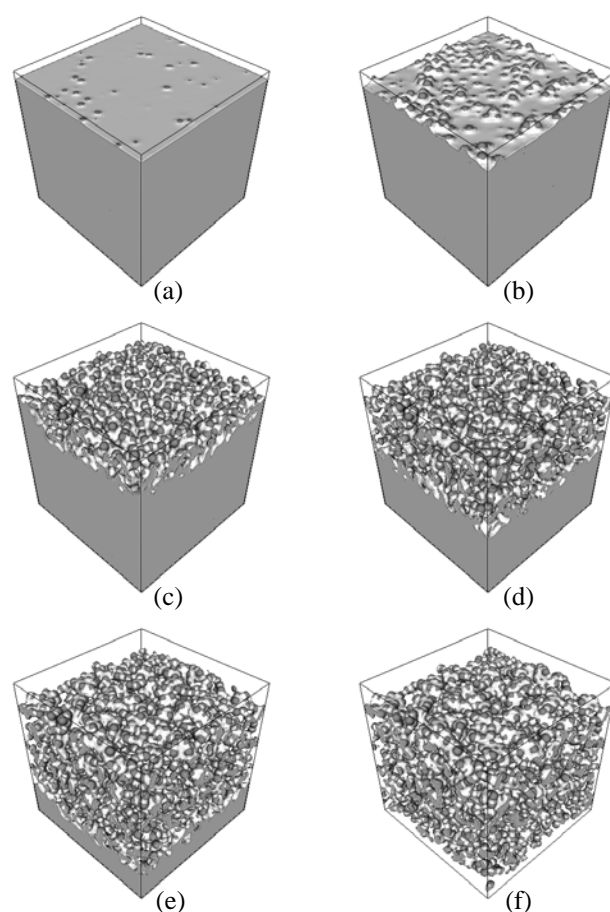
Figure 1: Simulated drying process (a)-(f). Gradually solvent evaporates, and then polymer binds particles. In the interface, surface tension agglomerates particles, and viscosity resists particle movement.

(a) Low solvent viscosity   (b) High solvent viscosity

(c) Low surface tension   (d) High surface tension

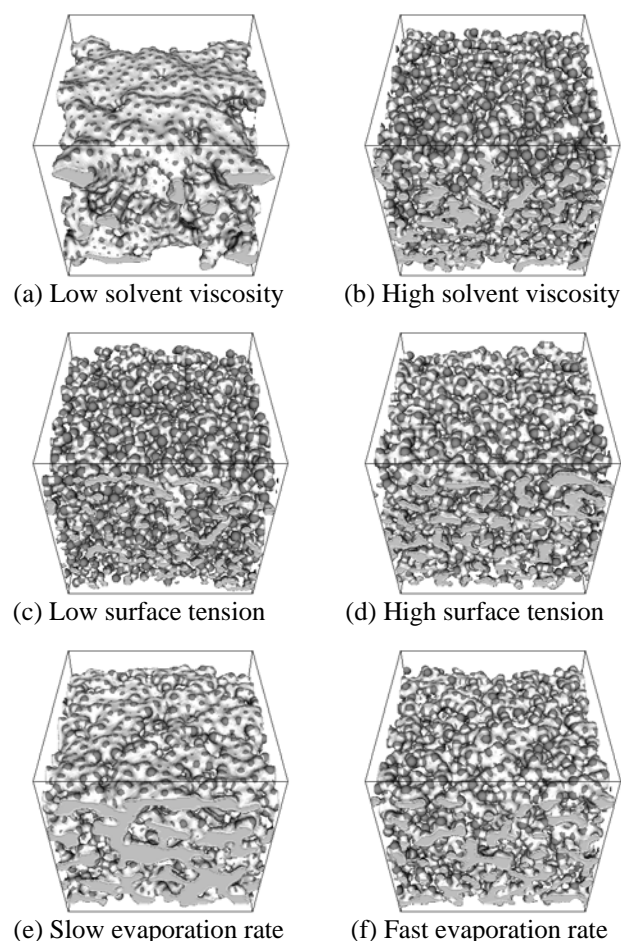(e) Slow evaporation rate   (f) Fast evaporation rate

Figure 2: Dried catalyst layer structures: effects of solvent viscosity (a)(b), surface tension (c)(d), and evaporation rate (e)(f).