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# A rapid start-up strategy for polymer electrolyte fuel cells at subzero temperatures based on control of the operating current density

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## ABSTRACT

In a previous study, we numerically described the three distinct stages of ice evolution during the cold start of a polymer electrolyte fuel cell (PEFC), namely, the freezing, undersaturated, and melting stages. Based on our numerical observations, we propose an efficient start-up strategy for achieving a rapid cell temperature rise while simultaneously mitigating the rate of ice accumulation within a cell. The key to this cold-start strategy involves raising the operating current of the PEFC in the undersaturated stage, which accelerates the cell temperature rise without any further ice accumulation. Using a three-dimensional, transient cold-start model, we numerically demonstrate that raising the cell current in the undersaturated stage is very effective and significantly improves the cold-start behavior of a PEFC. In contrast, increasing the cell current in the freezing stage has a negative impact on the PEFC cold start in that the oxygen reduction reaction causes the water production rate to increase, leading to more rapid ice growth. This ultimately results in cold-start failure and the deterioration of the porous electrode structure. This study clearly illustrates that optimization of the cold-start operation is key to improving the cold-start performance, while also pointing to the importance of the roles played by PEFC cold-start modeling and the simulations used to search for an optimum cold-start strategy.

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## Introduction

The rapid and stable start-up of polymer electrolyte fuel cells (PEFCs) in extremely cold weather is a prerequisite for the commercialization of fuel cell vehicles. In a subzero environment, water produced by the oxygen reduction reaction (ORR) on the cathode side of a PEFC would freeze into ice. This freezing inhibits oxygen transport in the electrode, reduces the active electrochemical area for the ORR, and damages the electrode structures, which leads to significant

deterioration in cell performance and durability. The traditional method for successful cold-start is simply to purge residual water within a fuel cell after a car shut down. This purging helps to avoid the possibility of damage to the PEFC components due to ice melting in the cool-down stage and also to retard the period of ice/frost nucleation during a PEFC cold start [1,2].

According to cold-start targets established by the Department of Energy, PEFCs should be able to reach 50% operating power within 30 s from  $-20^{\circ}\text{C}$  [3]. In order to accelerate the cell temperature rise from subzero temperatures while

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simultaneously suppressing the formation and growth of ice within a cell date, substantial efforts have been made to develop efficient cold-start methods and a number of cold-start strategies have been proposed [4–20]. Among them, the use of an external source of heat to warm-up a PEFC is to be avoided because doing so would increase the size and cost of the fuel cell system [4–9]. Therefore, a major focus of cold-start research has been the self-startup of PEFCs in subzero environments without increasing the system volume and/or cost [10–20].

The self-heating methods using either waste heat at cold start or in-situ chemical reaction heat have been presented in the literature [12–16]. Jiang et al. [12] suggested an effective current-ramping start-up method to achieve rapid cell warm-up from subzero temperatures. They found that using a lower ramping rate was effective in those cases in which the initial current density is lower, whereas the current ramping rate should be increased for operation at a higher initial current density. Sun et al. [13] allowed the hydrogen–oxygen catalytic reaction to occur in the anode catalyst layer (CL) to efficiently warm the cell from subzero temperatures. To ensure a safe warm-up, they recommended the use of a dilute hydrogen stream with a hydrogen fraction of less than 20%. Qian et al. [14] numerically studied the impact of the hydrogen–oxygen catalytic reaction on the cold-start behavior of a PEFC. Their model predicted successful self-startup from an initial temperature of  $-20\text{ }^{\circ}\text{C}$  with the aid of the chemical reaction. Jung et al. [15] suggested use of vanadium oxide film-coated metallic bipolar plate to enhance the cold-start capability of PEFCs. They demonstrated that the vanadium oxide thin film effectively generated additional ohmic Joule heat at the low current density and subzero temperatures. Lin et al. [16] studied the evolution of local current density distributions during cold-start using the printed circuit board technology. They showed that the lower cell voltage operations shortened the cold-start time when the cell was started at  $-10\text{ }^{\circ}\text{C}$  or higher startup temperatures but led to cell failure under the lower startup temperature ( $-15$  and  $-20\text{ }^{\circ}\text{C}$ ).

In addition to the effective thermal management of the PEFC cold start, efforts have been directed toward improving the water uptake potential and ice storage capacity of the fuel cell electrodes. Miao et al. [17] added the nanosized  $\text{SiO}_2$  into the cathode CL and showed an improved cold start performance at  $-8\text{ }^{\circ}\text{C}$ . They argued that the hydrophilic feature of  $\text{SiO}_2$  enhanced the water uptake capability of the cathode CL. Ko et al. [18] numerically showed that the use of a higher ionomer fraction in the cathode CL enhanced its ability to absorb water, successfully retarding the period of ice formation during a cold start. Later, Ko et al. [19] suggested the use of a dual-function microporous layer (MPL), made of an ionomer-based binder with a very low Pt loading, instead of the typical polytetrafluoroethylene (PTFE). The ice storage capacity of a cell was greatly improved using this dual-function MPL, because it can act as part of the cathode CL at subzero temperatures. The dual-function MPL was evaluated at a normal PEFC operating temperature of  $80\text{ }^{\circ}\text{C}$  [20]. Additional performance loss due to the use of the MPL was minimal (around 2–5 mV) even if the total amount of Pt loading in both the cathode CL and MPL was fixed to  $0.4\text{ mg/cm}^2$ .

Recently, Gwak et al. [21] presented an extended cold-start model for transient simulations over a wide range of

temperatures from subzero to the normal operating temperature, wherein ice melting and resulting membrane hydration and latent heat absorption processes were newly considered. The simulation results showed various stages of ice evolution, namely, the freezing, undersaturated, and melting stages. As the cell temperature continues to rise under the influence of the waste heat that is released during a cold start, the cell enters the undersaturated stage. During this stage, more water is stored in the vapor phase owing to the extra vapor saturation pressure. Furthermore, the water uptake potential and membrane water diffusivity are also greatly improved in this stage, which results in a stronger water back-flow from the cathode to the anode. Consequently, the rate of water loss in the cathode CL driven by these two mechanisms exceeds the rate of water production by the ORR and hence the amount of ice inside the cell remained constant without any further freezing of the produced water. This observation indicates that the current density could be increased further during this stage, which would in turn increase the rate of waste heat release and hence speed up the cell temperature rise.

The present study aims to numerically evaluate the impact of increasing the current in the undersaturated stage on the cold-start behavior of a PEFC. Particular emphasis is placed on investigating how the current-rise phase and cell thermal mass affect the rate at which the cell temperature rises, the ability of the electrolyte to absorb water, and the ice formation/evolution characteristics in the cathode catalyst layer (CL). This study clearly illustrates that the proposed current-raising strategy is an effective means of accelerating waste heat release without further ice accumulation within a cell and hence greatly facilitates the rapid start-up of a PEFC from a subzero environment.

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## Numerical model

To simulate a PEFC operating under a wide range of temperatures from subzero ( $-20\text{ }^{\circ}\text{C}$ ) to the normal operating temperature ( $80\text{ }^{\circ}\text{C}$ ), the three-dimensional, non-isothermal PEFC cold-start model developed in previous studies [18–24] was enhanced by including ice melting and the constitutive relationships for the PEFC transient simulation above  $0\text{ }^{\circ}\text{C}$ . A detailed description of the PEFC cold-start model appears in our previous paper [24]. Therefore, we present only a brief description here, with the emphasis on the new features developed in this study.

### Model assumptions

The specific assumptions made for the model are as follows:

- (1) incompressible and laminar flow in the gas channels and porous components of a PEFC, given the small pressure gradient and flow velocities;
- (2) ideal gas mixtures;
- (3) negligible gravitational effect;
- (4) isotropic and homogeneous porous layers for the GDLs and CLs, i.e., characterized by effective porosity and permeability;

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