



Numerical study on the thermal management system of a liquid metal battery module

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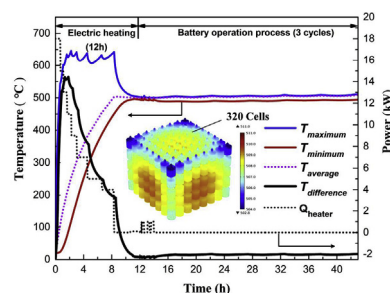


HIGHLIGHTS

- A coupling model is developed to study the thermal behavior of LMBs module.
- The heat generation and dissipation of the LMBs module are analyzed.
- A changeable-power-heating mode is proposed to achieve even heat distribution.
- The modelled LMBs module can achieve “self-heating” at 0.2 C constant current.

GRAPHICAL ABSTRACT

A changeable-power-heating mode is proposed for heating and maintaining the operating temperature inside the module.



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ABSTRACT

Liquid metal battery (LMB), with three-liquid-layer structure and high operating temperature (300–700 °C), is a newly emerging technology for large scale energy storage applications. A thermal management system is critical to achieve satisfied LMB performance and extend the life of batteries. In this work, an improved coupling model composing of a 3D heat-transfer model and a 1D electrochemical model is developed for the thermal analysis of a Li||Sb–Sn LMBs module (5.5 kWh). Key results including transient values, the contribution ratio of heat sources, temperature homogeneity and distribution, as well as the energy efficiency of the battery module, are presented. Based on the coupling model, the changeable-power-heating mode, sand filling material and vacuum insulation are further proposed to achieve the high energy efficiency and optimal performance of the LMBs module. Moreover, the LMBs module can achieve “self-heating” when operated at 0.2 C charge/discharge, under the vacuum insulation (0.01 W m⁻¹ K⁻¹ thermal conductivity, 100 mm thickness), requiring no external heating to keep the batteries at operating temperature.

1. Introduction

Liquid metal battery (LMB) is attracting increasing attentions as a low-cost, long-life, and low-maintenance energy storage technology for

the smart grid [1–11]. Structurally, LMB features a low-density liquid metal anode, a medium-density molten salt electrolyte and a high-density liquid metal cathode, which can be self-segregated into three layers due to the density difference and mutual immiscibility of

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electrodes and electrolyte when it is heated to the working temperature (300–700 °C). At present, potassium, magnesium and calcium-based LMBs (eg. K||Hg, Mg||Sb, Ca||Bi, Ca||Sb and Ca||Pb) [2,6–10,12], lithium-based LMBs (eg. Li||Bi, Li||Sb, Li||Pb, Li||Sn, Li||Zn, Li||Se and Li||Zn) [3,13–22], and sodium-based LMBs (eg. Na||Bi, Na||Hg, Na||Pb, Na||Sn and Na||Zn) [23–26], have been reported. However, these LMBs suffer from the high operation temperature (> 600 °C), low discharge voltage (< 0.8 V) and high cost of electrode materials (> 150 \$ kWh⁻¹). In 2014, Wang et al. [4] discovered Li||Sb-Pb system as an improved LMB that meets the basic requirement of the large-scale energy storage without consideration of the adverse impact of lead (> 0.87 V, 460 °C, 68 \$ kWh⁻¹). On the basis of this work, environmentally friendly liquid metal Sb-Sn cathode and Li||Sb-Sn LMBs (> 0.92 V, 500 °C, 73 \$ kWh⁻¹) were developed [3,5], showing great potential in the large scale energy storage for the grid.

So far, appreciable achievements have been made in developing new systems and high performance electrode materials in LMBs, but little research on the thermal management has been reported. Due to the high operating temperature of LMB [3], a thermal management is essential to heat the batteries to the desired temperature and maintain an even temperature in a module. The heat dissipation from the module, greatly affects the battery performance and the module efficiency, even though the LMB exhibits a wider thermal tolerance range compared with other traditional batteries [1]. However, the large cell size and numbers of cells in the module, make it difficult to accomplish temperature uniformity inside a cell or between the cells in a module. Therefore, optimal design of the thermal management system for a module is particularly crucial for practical applications of this battery system.

To design appropriate thermal management system, various methods have been used to predict the thermal performance of a battery module, which can be mainly divided into two categories: experimental investigation and numerical modeling [27–68]. Experimental tests are useful for the thermal analysis of a small scale room-temperature battery storage systems [58,59], but is not suitable for the large scale battery storage systems, since it could lead to long experimental time and high cost. Numerical models are also widely used for understanding the thermal behavior of the cell and module. Several numerical models have been reported, such as electrical-thermal models and electrochemical-thermal models. The electrical-thermal models [27,36,59–64] are based on the equation of Bernardi et al. [65] or ohmic law. Lee et al. [27] proposed a general numerical study on the thermal behavior and temperature distribution of a molten sodium-sulfur battery module using a 3D electrical-thermal model. However, the 3D electrical-thermal model ignored the actual electrochemical process and heating process from the initial heating stage of the Na-S module, which could affect the accuracy of the prediction. The electrochemical-thermal models [50,66–68] consider the charge balance, mass transport, electrochemical kinetics, energy balance, heat transfer, and heat convection. Very recently, Ghalkhani et al. [68] proposed an electrochemical-thermal model composing of a 3D thermal model and a 1D electrochemical model to estimate energy density, heat generation, and temperature distribution of lithium ion batteries. This model can be applied as a fast and accurate tool to optimize the thermal performance of a lithium ion battery pack.

However, because of the unique structural feature, reaction mechanism, and operation conditions, the numerical models for other battery systems cannot be directly used to analyze the thermal performance of LMBs. Herein, we proposed an improved coupling model covering a three-dimensional (3D) heat-transfer model and a one-dimensional (1D) electrochemical model, to analyze the thermal performance of the Li||Sb-Sn battery module (5.5 kWh). The 1D electrochemical model exhibits high accuracy with maximum error below 5%. The 3D heat-transfer model is established to study the heat generation and dissipation, the temperature variation, difference, and distribution, as well as the energy efficiency of the module. According to the simulated results, the changeable-power-heating mode is proposed to

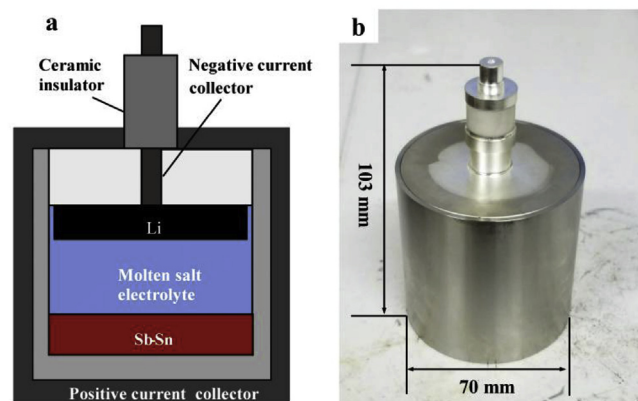


Fig. 1. (a) Cell schematic of Li||Sb-Sn liquid metal battery; (b) A 23Ah Li||Sb-Sn battery.

effectively control the temperature difference and maintain the operating temperature.

2. Experiments

Fig. 1 represents the schematic of the Li||Sb-Sn liquid metal battery, which contains lithium anode, antimony-tin alloy cathode, and Li-F–LiCl–LiBr molten salt electrolyte. The 23Ah Li||Sb-Sn LMB is assembled in an argon-atmosphere glovebox, as described in the previous works [3,5]. An electric stove is applied to heat and maintain the cell at operating temperature of 500 °C. The stove is operated with 3 kW to rise the temperature inside the stove for 5 h to make the temperature stable at 500 °C. Land battery test cabinet (3V20A8CT-Q) is used to test the cell with galvanostatic charge and discharge. The cell is fully discharged or charged at a constant current (0.1 C, 0.2 C, 0.3 C, and 0.4 C), the idle time is set to 300 s when finishing a discharge or charge process. Three-cycles data of the discharge/charge curve and energy efficiency at different constant currents are measured and recorded.

3. Model development

The coupling model is developed and computed by the finite element method (FEM) software [46,48] and Dell Precision 7710 Work-Station (Intel Xeon CPU E3-1575M-v5, 3.00 GHz, a total 64 GB random access memory (RAM)). The coupling model, including the 1D electrochemical model and the 3D heat-transfer model as shown in Fig. 2, is developed based on the 23 Ah Li||Sb-Sn LMB. The 1D model is applied for simulating the thermal and electrochemical properties of a single Li||Sb-Sn cell. The overall heat of the cell based on the 1D model, including the Joule heat, the active polarization heat, and the reaction heat, will be coupled with the 3D heat-transfer model as heat sources. The main model assumptions are described as follows:

- (1) The effect of negative and positive current collectors on the thermal performance is ignored in the 1D model due to their negligible heat contribution caused by high electrical conductivity ($1.74E+06 \text{ S m}^{-1}$) to the module;
- (2) Electro-neutrality and no concentration gradients of the lithium ion in the electrolyte of the 1D model because of the high Li^+ concentration in the molten salt electrolyte ($> 3.0E+05 \text{ mol m}^{-3}$);
- (3) The physical and electrochemical properties of the 320 cells in the module are identical;
- (4) The influence of air or other gases is neglected inside the 3D module model;
- (5) The solid geometry of electrical heaters is neglected in the 3D module model for reducing the amount of mesh and calculation, and electrical heaters are set at six internal boundary faces of the

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