Journal of Power Sources 324 (2016) 665–673

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

A thermo-mechanical stress prediction model for contemporary planar sodium sulfur (NaS) cells

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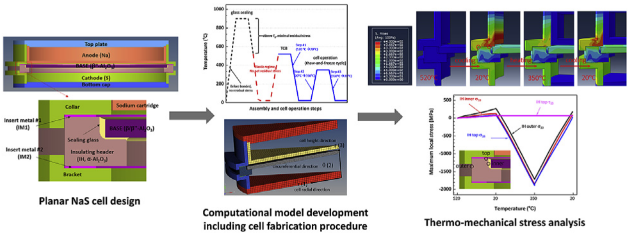
HIGHLIGHTS

- A comprehensive FEA model is introduced to predict stress in contemporary planar NaS cell.
- Model includes relevant experimental procedures for planar NaS cell assembly and operation.
- Large stresses were developed on the outer surface of insulating header and solid electrolyte.
- Cell container thickness plays an important role in the stress accumulation of planar NaS cell.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 5 April 2016
Received in revised form
29 April 2016
Accepted 27 May 2016

Keywords:
Sodium beta-alumina battery
Sodium sulfur battery
Thermo-mechanical stress
Cell failure
Finite-element analysis

ABSTRACT

We introduce a comprehensive finite-element analysis (FEA) computational model to accurately predict the thermo-mechanical stresses at heterogeneous joints and components of large-size sodium sulfur (NaS) cells during thermal cycling. Quantification of the thermo-mechanical stress is important because the accumulation of stress during cell assembly and/or operation is one of the critical issues in developing practical planar NaS cells. The computational model is developed based on relevant experimental assembly and operation conditions to predict the detailed stress field of a state-of-the-art planar NaS cell. Prior to the freeze-and-thaw thermal cycle simulation, residual stresses generated from the actual high temperature cell assembly procedures are calculated and implemented into the subsequent model. The calculation results show that large stresses are developed on the outer surface of the insulating header and the solid electrolyte, where component fracture is frequently observed in the experimental cell fabrication process. The impacts of the coefficients of thermal expansion (CTE) of glass materials and the thicknesses of cell container on the stress accumulation are also evaluated to improve the cell manufacturing procedure and to guide the material choices for enhanced thermo-mechanical stability of large-size NaS cells.

1. Introduction

Increasing demands toward reliable energy resources are continuously forcing the implementation of advanced technologies that can both economically and safely store large amounts of

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http://dx.doi.org/10.1016/j.jpowsour.2016.05.128
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energy. An energy storage technology that can effectively address the spontaneous needs of the consumer while exploiting renewable clean energy sources, is vital for the future generation. Sodium sulfur (NaS) battery has received a strong attention as one of the most promising candidates for grid scale energy storage systems (ESS) because of a number of benefits, such as its high energy density, long discharge time, long lifetime, no self-discharge, and low manufacturing costs [1–5]. The NaS cell is typically comprised of characteristic molten electrodes (i.e., Na for anode and S for cathode), β/β''-Al_2O_3 solid electrolyte (BASE), intricate sealing areas, and outer metallic cell container.

Depending on the shapes of its BASE, two different cell geometries (i.e., tubular cells and planar cells) have been suggested for practical applications of NaS cells. For grid scale energy storage applications, large-size (1,200 Wh class) tubular sodium sulfur cells have been developed by NGK Insulators, Ltd, in 2003 [6], and their products have been being successfully operated in many countries thereafter [7,8]. Currently, a couple of other groups are also in the developing stage for the stationary energy storage systems based on tubular NaS batteries aiming to the product realization of the technology [9–16]. Other than the tubular NaS cells, another possible NaS cell design relies on a planar geometry utilizing a flat plate BASE. First practical planar cells were constructed and tested in 1970's for automotive applications [17,18]. Since the geometry of a NaS cell is primarily determined by the shape of its BASE, as long as the BASE forming technology is available, other alternative cell designs can also be suggested including a flat tube-shaped cell or a cell with a curved solid electrolyte. Application of a tubular BASE with a clover leaf-shaped cross-section in sodium nickel chloride battery ($Na/NiCl_2$, also known as ZEBRA battery) is a good example of different geometries to increase the active area and to reduce the thickness of the cathode compartment [19]. However, most of the contemporary technologies to build the derivatives are more or less related to one of the two extremes (i.e., tubular or planar), therefore, development of rudimentary technology for these two extreme designs is of special importance to fabricate new cells with other shapes.

The planar NaS cell has a number of distinct advantages over tubular cells, such as easiness for stacking, inter-cell connection without external connectors, and geometric merits for mass production, thereby reducing the manufacturing cost. Furthermore, it is easier to eliminate the orientation and gravity effects, and therefore, the active area of the electrode and electrolytes can be sharply defined, and it is also easier to perform post-analysis for cell components [20]. With this, the planar design has been widely used in laboratory conditions. Accordingly, planar cell designs have been adopted for developing electrode materials/structures and for testing new cell chemistries with small circular plate-shaped BASE, of which diameter is typically in the range of 10–50 mm. For example, there are several recent efforts to reduce the operation temperatures of NaS battery systems from 300–350 °C to intermediate temperatures (95–190 °C) [21,22] or to room temperature [23–28]. In addition to these, the planar design is also useful for fundamental studies on the wetting characteristics of molten sodium on β/β''-Al_2O_3 membrane [29,30], or on testing novel electrodes for sodium metal halide chemistries [31–33].

Despite these advantages, critical issues are still remained in developing practical planar NaS cells. Since 1970's, it was well-perceived that the diameter of a BASE disk should be at least 80 mm to provide a reasonable competitive specific energy [34]. Although several pioneers delivered some insightful lessons through their efforts for the development of planar sodium beta-alumina cells for high power applications [34–36], no commercial scale success has yet been reported. Common unresolved problems include the insufficient mechanical properties of heterogeneous joints and BASE, which can result in a catastrophic cell failure from the accumulation of thermo-mechanical stresses upon cell assembly and operation (i.e., thermal cycling). The thermo-mechanical stresses are originated from the differences in the coefficients of thermal expansion (CTE) of various cell constituents (sealing glass, BASE, α-Al_2O_3, and metallic components). The degree of such stress accumulation increases as the cell size increases. Because there would be a competition between the accumulated thermo-mechanical stress and the bonding strength to avoid the cell failure, the maximum NaS cell size must be determined by the capability to resist the accumulated stresses. To develop a large commercial planar cell, therefore, two-way approach deems to be reasonable: (1) minimization of the thermo-mechanical stress accumulation through optimizing the cell design and materials choices and/or (2) maximization of bonding strength through applying robust joints and components to avoid undesired fracture. In an effort to resolve the thermo-mechanical stress issues via approach (2), recent development of commercial scale planar sodium batteries has adopted advanced joining technologies, such as electron beam welding (EBW) and laser welding for metal-metal joints, thermal compression bonding (TCB), brazing, or state-of-the-art load framing for metal-ceramic joints, and the glass sealing (GS) for ceramic-ceramic joints [37–39].

Even with the development of these state-of-the-art bonding technologies, successful experimental fabrication of large-size practical planar NaS cells presents a formidable challenge. In this study, therefore, we introduce a comprehensive *in silico* finite-element analysis (FEA) model to accurately predict the thermo-mechanical stress concentrations at the heterogeneous joints and components of planar NaS cells. This is in line with the approach (1) addressed above to minimize the thermo-mechanical stress by optimizing the cell geometry and materials. Using this prediction model, the thermo-mechanical stress field was calculated using one of our representative prototype planar NaS cells assembled with EBW, TCB, and GS bonding techniques. In the next sections, the computational methods are detailed and the calculation results are generally discussed to estimate the possible fracture location and fracture mode. In addition, we examined the impacts of the CTE of sealing glass and the thickness of cell container on the stress concentration and the cell failure by varying the CTE values of sealing glass and the thickness of metal container. Based on the computational results, the CTE value of sealing glass and the thickness of the container materials have been identified to minimize the stress concentration at the vulnerable joints for the planar cell design considered in this work.

2. Computations

2.1. Cell design and model construction

Fig. 1(a) illustrates the cross-sectional design of the representative prototype planar NaS cell. The anode and the cathode compartments of this prototype cell are filled with molten Na and S, respectively, separated by the BASE. The sealing area is composed of heterogeneous joint parts including α-Al_2O_3 insulating header (IH), insert metals (IM), glass sealing (GS), and surrounding cell container collars. For conveniences, the upper and lower insert metals are referred to as IM1 and IM2 in this work. The standard choice in dictating the size of planar NaS cell is to designate the diameter (i.e., disc size) of BASE. In our study, the disc size of BASE was set to 120 mm, as it was suggested that the minimum useful BASE disc size is 80 mm and the optimum disc size is 250 mm [1,34]. Also, note that, in our in-house cell fabrication and operation experiments at RIST (Research Institute of Industrial Science and