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Long-life, multi-tap thermal battery development

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Abstract

This paper describes an effort to develop long-life, multi-tap thermal battery technology with a minimal weight and volume. The effort has several challenging goals. Some of the development goals include an activated life of at least one hour, four voltage sections, and the ability to sustain significant pulse loads at the end of life. In order to meet these goals, advanced materials were chosen for development. The thermal battery chemistry developed consists of lithium-silicon anodes, low-melting eutectic electrolyte/separators, and cobalt disulfide cathodes. Besides evolving the electrochemistry for this battery, there are several other design challenges such as fine-tuning the heat balance so as to allow the battery to sustain the extended duration discharge. In addition, to minimize volume, the battery can is configured in a tapered shape and consequently requires a tapered Min-KTM sleeve for insulation. A new igniter design is also being used. Finally, extremely narrow voltage ranges for each of the four voltage taps have contributed to the challenges facing development engineers. This paper includes a summary of the battery design and presents test data from pre-prototype units. © 2004 Elsevier B.V. All rights reserved.

Keywords: Thermal batteries; Molten salt electrolytes; Cobalt sulfide; Lithium-silicon

1. Thermal battery development

1.1. Battery development overview

Thermal batteries have long been used in defense applications [1–4]. Sandia has continued to research and improve the technology during the last several years [5,6]. The battery described in this paper is an outgrowth of this research and represents a step forward for the technology. For this battery, due to stringent design goals it was recognized early in the development process that new electrochemical materials were needed. In particular, because of the long activated life and significant pulse loads at the end of life, a cathode material and an electrolyte that have never been used before in a Sandia-designed battery are key elements of the conceptual device. However, with unproven materials, the risk is increased because of potential contamination and/or variability issues. Other aspects of the battery are also new, such as the use of a tapered battery can, matching tapered insulation, a new igniter, and extremely narrow voltage ranges for each of the four taps.

Between 1997 and 2001, over 200 single-cell units were built and used for screening tests and material studies. In 2001-2002, 10-cell batteries (30 units) in reusable con-

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tainers were built and tested to characterize multiple-cell voltage performance. To evaluate the pre-conceptual design options, several materials and fabrication techniques required in-depth analysis. The analyses included the use of several different cathode materials, advanced separators, improved insulation, and plasma-sprayed electrodes. A materials feasibility analysis of the cobalt disulfide (CoS_2) and iron disulfide (FeS2) cathode chemistries was also conducted during the period from 1997-2002. An analysis of paper separators was also performed. These analyses provided the pros and cons of each option, as well as providing the background information to facilitate a decision as to whether the design requirements could be met.

In 2002, several design choices were made based on these analyses and evaluations. A conceptual design for the battery was developed and implemented. In the past year, Sandia has built more than 12 full-scale batteries to evaluate this design and to refine the thermal management of the battery. All of these batteries have met the design goals. Sandia began building and testing prototype I and prototype II batteries in 2002 and continues in 2003. These units represented the final cell stack design. It was at this stage that parametric studies were started that included: refining the heat balance, resolving closing force issues, and conducting reliability and aging analyses. Along with the many other activities, production and optimization analysis were also initiated. Also during the full-scale testing phase electrode

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Fig. 1. Full-scale battery stack (13 cells).

materials studies were performed. Prototype II units reflect final stack design and hardware, and some of the batteries have been tested with the exterior of the battery coated with epoxy, or "foam". Assembly of the thermal battery stack proceeds in layers and is shown is Fig. 1 (S/N 51). The finished thermal battery is shown in Fig. 2. All batteries described in this paper were built and tested at Sandia.

1.2. Thermal battery design

Sandia's background in thermal battery research and development provided significant advantages in many technology areas required for this effort. Over the years, SNL has conducted research on different cathode materials, which includes extensive research with FeS₂ and CoS₂ materials for various applications [7]. Early work on CoS₂ cathodes



Fig. 2. Thermal battery.

was conducted by Argonne National Laboratory (ANL) [8]. Energy compression devices incorporating CoS_2 are the subject of a patent presently owned by The Enser Corporation [9]. In a long-life battery application, CoS_2 has advantages because it has a higher decomposition temperature and higher conductivity. Enhanced performance and voltage stability were additional factors in making CoS_2 the material of choice for this effort. Comparison test data of two batteries made with these materials are shown in Fig. 3. Sandia also evolved a low-melting eutectic electrolyte/separator originally developed at ANL [10–13] for these requirements. Further, a tapered end on the battery case necessitated a new tapered Min-KTM insulation sleeve to provide the required thermal insulation.

The load and size requirements included: a 1 h life, four different voltage taps, a maximum weight of 1 kg (2.2 pounds) (including 230 g [\sim 0.5 lb] of foam coating), and a volume of 28 in.³ (460 cm³). The peak load requirement for the battery for all taps combined is on the order of several amperes and occurs at the end of battery life. The voltage requirements for each tap are:

- 3.3–3.7 V;
- 6.2–7.2 V;
- 9.3–10.7 V;
- 16–22 V.

The full-scale battery configuration has 13 cells in two independent sections and has a total of four taps (see Fig. 4). The following chemistry and design variables have evolved to meet the stringent design requirements.

- Cathode composition: a blend of CoS₂, electrolyte binder (EB), & Li₂O.
- Alloy anodes: Li–Si (44–56 wt.%) (no electrolyte).
- Low-melting eutectic electrolyte: LiBr-KBr-LiCl.
- Separator/EB: 70 wt.% electrolyte, 30 wt.% MgO.
- Heat powder 88/12-84 cal/g heat balance.



Fig. 3. Performance comparison of CoS₂ vs. FeS₂ cathode materials.



Fig. 4. Key design features for the battery and the electrochemical cell configuration.

- 500-650 psi battery stack closing force.
- Tapered end on battery can.
- Special tapered Min-KTM sleeve insulation.
- Eight pins for electrical connections in battery header.

2. Sub-scale battery tests

In 2000 and 2001, 10-cell batteries (30 units) in reusable containers were built and tested. Ultimately, these units validated pre-conceptual design objectives (established earlier) and provided the environment to evaluate performance of the multi-tap system as well as conduct additional analyses of insulation options and heat balance. The sub-scale testing also enabled electrical performance refinements.

At this stage of development with the minimum required masses of anode and cathode materials defined, electrode physical characteristics were specified such as cell-stack diameter and densities for anode, electrolyte, and cathode materials. Battery heat balance was also optimized, and pellet thicknesses were refined. Battery container configurations that would adequately provide heat containment using the Min-KTM insulation were also extremely challenging.

Heat balance, defined as the calorific heat content of one heat pellet divided by the total mass of a single cell, is used to characterize the heat input to a thermal battery. In general, Sandia's design guidelines suggest using relatively mild heat powder to reduce the thermal shock during ignition and the risk of warping battery components. Given the space and volume constraints of this battery, a relatively low-heat capacity heat powder, 88/12 (wt.%) was used. Also, given that the battery's activated-life requirement was one hour, a large thermal mass was critical. Based on heat-powder calorific output, desired heat balance, and the number of cells, a heat-pellet thickness was determined for the full-scale batteries.

One of the main results from sub-scale testing was that the 1 h activated life requirement was found to be achievable. Additionally, many of the electrochemical materials choices were made and several mechanical design options were resolved during the sub-scale effort prior to the full-scale tests.

3. Full-scale battery tests

Two levels of prototypes (referred to as prototype I or II) were built and tested beginning in 2002. The prototype I batteries used a cylindrical can and a generic header. The tapered shape of the can, the internal tapered thermal insulation, and a final header design used in the prototype II units were the main differentiating factors. The interior of both types of prototype batteries represented final stack design. Battery S/N 51 was a prototype I battery and relevant test data are shown in Fig. 5. The data clearly show the unit achieved its 1 h activated life and it had excellent electrochemical performance and the ability to sustain the pulse loads at the end of life.



Fig. 5. Prototype I thermal battery S/N 51.



Fig. 6. Prototype II thermal battery S/N 55.



Fig. 7. . Prototype II thermal battery S/N 59.

Battery S/N 55 (Fig. 6) was one of the first prototype II tests. Thermal management is still very much an issue at this stage of testing. As the data illustrate, the results were very promising. Battery S/N 59, also an early prototype II unit, was tested with the external insulating foam, which was an additional variable that presented new challenges. S/N 59 was also tested using a special development load

that may have contributed to higher voltages (see Fig. 7). It is clear from the comparison data of S/N 51 and 59, shown in Fig. 8, the optimized prototype I unit out-performed the still-evolving prototype II unit. The S/N 59 test was successful because it did achieve one hour of activated life, but exhibited reduced margins against the tight voltage limits. These issues are the subject of continuing development.



Fig. 8. Comparison test data of Prototype I and II batteries S/N 51 and 59.

4. Conclusions

Sandia began this work by conducting many single-cell tests that provided insights into refinements necessary to proceed to multiple-cell and multi-tap testing. After the single-cell testing, Sandia built numerous sub-scale batteries that validated the electrochemical options developed earlier in the project. In 2002, the design was scaled up to full-size batteries. At each phase of testing, results showed the design capable of providing long life in this multi-tap thermal environment.

In summary, the long-life, multi-tap battery design does achieve the 1 h activated lifetime. The voltage performance under the required load profile of several full-scale batteries was excellent. Further development will proceed as prototype II test units provide additional insights. Optimizing the heat balance is an area that tests reveal requires further investigation. However, this design shows significant potential for future applications.

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