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Current-driven flow instabilities in large-scale liquid metal batteries, and how to tame them



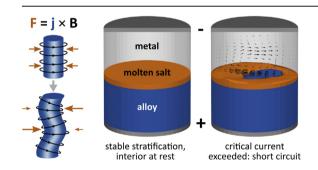
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HIGHLIGHTS

- Large liquid metal batteries can be susceptible to current driven instabilities.
- Critical parameters for the onset of the Tayler instability are derived and computed.
- Different countermeasures against the Tayler instability are demonstrated.

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ABSTRACT

The use of liquid metal batteries is considered as one promising option for electric grid stabilization. While large versions of such batteries are preferred in view of the economies of scale, they are susceptible to various magnetohydrodynamic instabilities which imply a risk of short-circuiting the battery due to the triggered fluid flow. Here we focus on the current driven Tayler instability and give critical electrical currents for its onset as well as numerical estimates for the appearing flow structures and speeds. Scaling laws for different materials, battery sizes and geometries are found. We further discuss and compare various means for preventing the instability.

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1. Introduction

With the large-scale deployment of renewable energy sources, massive and cheap electricity storage becomes indispensable since the major part of renewable electricity generation (solar, wind) is inherently fluctuating. Storage is thus essential to balance supply and demand and to stabilize the power grid.

Given that the potential for pumped storage hydropower is largely exhausted, electrolytically generated hydrogen, partly

processed to synthesized hydrocarbons, seems to be the only viable option for long-term large-scale storage on the TWh scale. However, the total efficiency of the conversion chain is relatively low due to the multitude of process steps involved. Electrochemical energy storage (EES) shows generally higher efficiencies, but needs improvements towards larger capacities at significantly lower costs [1]. If these demands can be met, EES will be an attractive candidate for short-term and mid-term stationary electricity storage.

Liquid metal batteries (LMBs) are currently discussed as a means to provide economic grid-scale energy storage [2]. Typically, an LMB consists of a liquid alkaline metal layered atop a molten salt which itself floats on a molten metal or half metal. On discharge, the alkaline metal of the anode is oxidized and cations enter the

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fused salt which is often an eutectic composed of alkali halides. At the electrolyte/cathode interface, alkaline ions leave the electrolyte, are reduced and alloy with the cathodic metal. For this setup to work, the alkaline metal needs to be lighter than the metal forming the cathode, and the molten salt's density must be in between those of both metals. A battery with a fully liquid active interior has a number of advantages: The battery is self-assembling due to stable density stratification. Liquid—liquid interface processes possess fast kinetics, thereby allowing for fast charging and discharging and high current densities (about 4—100 kA m $^{-2}$). Structureless electrodes are insusceptible to aging which allows for potentially unlimited cyclability. Scale-up on the cell level is facilitated by the simple cell construction. Drawbacks include the elevated operation temperature and the relatively low cell voltage (typically below 1 V).

LMBs, or molten salt batteries [3], were intensively investigated in the 1960s mainly as part of energy conversion systems, i.e., thermally regenerative electrochemical systems (TRES). Interestingly, already at that time the use of LMBs for off-peak energy storage received interest as well as funding [4]. Among the systems investigated early were Na—Sn [5,6] and K—Hg [7] because they can be easily separated by distillation. In the guest for higher cell voltages and current densities, among others, Na-Bi, Li-Te, and Li-Se cells were built, abstaining from thermal separability for the two latter systems. Progress in the field has been reviewed several times in the 1960s and early 1970s: Crouthamel and Recht [8] edited a special volume of Advances in Chemistry in 1967 dedicated to "regenerative EMF cells". The large body of research performed at Argonne National Laboratory was discussed in detail by Cairns et al. [9] and described in a more compressed version by Cairns and Shimotake [10]. Swinkels' [3] discussion had a slightly broader scope. A very recent account is due to Kim et al. [11] who not only review the previous achievements, but report on the ongoing research at the Massachusetts Institute of Technology (MIT) as well. The focus of the MIT activities is on the deployment of LMBs for large-scale stationary energy storage [2]. Consequently, emphasis is now on different practical and economical aspects as the utilization of abundant and cheap active materials [12].

Besides the chemistry of different material combinations, and the related corrosion problems, there are further aspects of LMBs which should not be overlooked. For traditional battery systems (apart from lead-acid and redox flow batteries) fluid dynamics does not play a significant role. This is different for LMBs due to their completely liquid interior. On the one hand, mass transfer in the electrolyte and the cathodic compartment can be facilitated by liquid motion. On the other hand, the density stratificationappearing stable at first glance-might be disturbed by flow instabilities (see Fig. 1). Here we will focus on a special magnetohydrodynamic phenomenon. After a few short remarks on interface instabilities in current carrying systems, we will proceed to the current-driven, kink-type Tayler instability, which is the main subject of the paper. It should be noticed that the significance of hydrodynamically stable interfaces for the operation of LMBs was recognized early, see, e.g., Cairns et al. [9] and Swinkels [3].

Interface instabilities are well known phenomena in Hall-Héroult, i.e., aluminum reduction cells (ARCs) [13–17]. These instabilities result from the interaction of the ionic current with a background magnetic field generated by the current supply lines. As a consequence, waves with a length comparable to the cell width develop and culminate in a sloshing motion of the aluminum. Amplitudes may become high enough for the aluminum to contact the graphite anodes, effectively short-circuiting the cell and terminating the reduction process. To keep the long-wave instabilities at bay, the cell current has to be kept below a critical value and the cryolite layer should not be thinner than about 4.5 cm. These

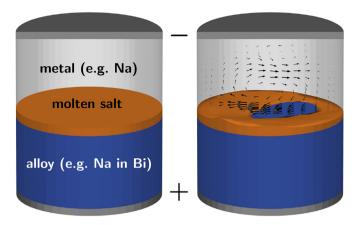


Fig. 1. Sketch of a liquid metal battery with typical inventory (left). The electrolyte works as ion conductor and separates the two liquid metals. A movement of the fluid, as it may result from magneto-hydrodynamic instabilities, could wipe the electrolyte and lead to a battery failure (right).

requirements lead to a total cell voltage of about 4.5 V. 2 V of those, i.e., around 40% of the cell voltage are spent on overcoming the ohmic resistance of the electrolyte layer, the corresponding power is converted to heat [15]. Since expenses for electricity are a major part of the overall costs of aluminum production, a reduction of the electrolyte thickness even by a few millimeters only would result in huge savings, but is impeded by the long-wave interface instabilities described above.

ARCs have two liquid layers sandwiched between the carbon anode on top and the current collector on the bottom. In contrast, LMBs are three layer systems. Presently, only little is known about current driven interface instabilities in such settings. Sneyd [18] predicts theoretically that additional short wave instabilities may arise in the case of the two neighboring liquid—liquid interfaces. To the authors' best knowledge, no experimental observations of current driven instabilities in three layer systems with closely spaced interfaces have been reported to date. Hoopes cells, i.e., three-layer cells used for the electrolytic refining of aluminum, apparently do not suffer from such instabilities. The electrolyte layer of these cells is quite massive with about 8-10 cm thickness [19,20]. Thus, a strong interaction of both interfaces is relatively unlikely. However, violent swirling motions driven by non-uniform electric and magnetic fields emerging from the cathodic current collectors were observed quite early [21]. These flows occurred, if the cathode metal thickness was chosen too small. As a result, spotty contacts of anode and cathode layers could occur.

Coming back to LMBs, the maximum thickness of the electrolyte layer is limited by the requirement that the voltage loss in the electrolyte must not exceed the available open circuit voltage (OCV). Indeed, a meaningful design requires the voltage losses to be much smaller than the OCV. For the most relevant combinations of metals and salts, this means the electrolyte's thickness should not exceed a few millimeters. Under those conditions, a careful consideration of the stability of the interfaces becomes imperative.

Exploiting the economies of scale is the usual route towards economic devices. Since current densities are large, cell scale-up (up to a few cubic meters [22]) will result in considerable total cell currents. These large currents can trigger the so-called Tayler instability (TI), as has been shown in a recent liquid metal experiment by Seilmayer et al. [23]. The TI is a kink type instability very similar to that occurring in the z-pinch of plasma physics. TI manifests itself in one or, depending on the cell's aspect ratio, more vortices rotating around an axis normal to that of the cell. Weber et al. [24] developed an integro-differential equation solver (in the sense of Meir et al. [25]) capable of reproducing the experimental

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