Convection-Diffusion Model of Lithium-Bismuth Liquid Metal Batteries

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Abstract The liquid metal battery is composed of two liquid metals with different electronegativity separated by molten salt. The three layers self-segregate based on density allowing for easy manufacturing and scaling. Lithium (Li) is one of the most widely researched anode materials, and when coupled with bismuth (Bi) cathodes, it gives a liquid metal battery that has an open circuit voltage of 0.9 V. Such a system has demonstrated impressive rate capabilities, ultra-long life cycle, and low energy cost. Here we present a two-dimensional physics-based model for Lithium-Bismuth liquid metal batteries. The model takes into account dynamical changes in the battery, including surface concentration and fluid flow. By solving the convection-diffusion equation in Bi electrodes, we are able to investigate the effect fluid flow on kinetic losses and concentration profiles in real time. The outcome of this work allows us to link electrochemistry and fluid dynamics in liquid metal batteries. Moreover, the model can also be used to guide future development of battery management systems.

Keywords Liquid metal battery · Battery model · Magnetohydrodynamics

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dynamical changes in the battery, including surface concentration and fluid flow. By solving the convection-diffusion equation in Bi electrodes, we are able to investigate the effect fluid flow on kinetic losses and concentration profiles in real time. The outcome of this work allows us to link electrochemistry and fluid dynamics in liquid metal batteries. Moreover, the model can also be used to guide future development of battery management systems.

Introduction

Today's electrical grids were designed back when energy storage did not exist. Hence, they require constant monitoring in order to maintain the balance between energy generation and consumption by end users. The integration of renewable energy sources such as wind and solar adds to the complexity of grid management due to the fact that these sources are intermittent. Energy storage can allow the seemingly impossible task of decoupling supply from demand. Moreover, it allows electrical grids to operate at a steady base-load. According to a 2013 report by the U.S Department of Energy, the United States will require 4 to 5 TWh of storage capacity in order to accommodate the rising demand for renewable energy integration [1]. However, grid-scale storage technologies must be cost-competitive without subsidies and must be able to integrate into existing infrastructure. In fact, the US Advanced Research Projects Agency-Energy (ARPA-e) has defined cost competitiveness as the ability to deliver energy at a cost of 100\$/kWh [1, 2]. One technology that meets these strict cost requirements is the liquid metal battery (LMB) [3].

LMBs are composed of two molten metals from opposite sides of the periodic table separated by a mixture of molten salts. Candidates for anode materials include alkali and alkaline metals such as Lithium (Li), [3, 4] sodium (Na), [5, 6] or calcium (Ca) [7, 8]. Candidates for cathode materials include antimony (Sb), bismuth (Bi) and zinc (Zn) [9]. Both the anode and cathode materials are selected to ensure the battery remains liquid during operation. The electrolyte mixture, typically composed of molten metal halides, hydroxides, amides or mixtures thereof, [10] is chosen in order to maximize ionic conductivity and minimize operating temperature. Since the three layers are self-segregated based on density, the assembly of these batteries does not require sophisticated manufacturing techniques, hence, lowering the overall cost of the battery. A schematic representation of typical liquid metal battery assembly is shown in Fig. 1.

Li-based LMBs are of great interest due to their high potential, low solubility in molten salts, high ionic conductivity of molten halides, [11] and ultra-long life cycle [3]. Of particular interest is the Li||Bi LMB due to its high voltage (0.9 V) and intermediate operating temperature (<400 °C). In addition, Li||Bi batteries allow deep cycling to the point where the cathode undergoes a phase transformation from liquid Bi to a completely solid Li–Bi intermetallic. Hence, it is the combination of



Fig. 1 Schematic representation of a lithium based liquid metal battery. The model focuses on transport in the positive (bismuth) electrode

these exciting features, the availability of thermodynamic data [12] and material properties [13] that attracted our attention this system.

When the LillBi LMB is connected to an external load, Li metal ions are produced via oxidation (Li⁺). These metal ions have to travel through the molten electrolyte before they are reduced at the surface of the positive electrode where they form an alloy (Li_{in-Bi}). The open circuit potential of the battery depends on the Gibbs free energy of the alloying reaction. Upon discharge, N moles of Li metal oxidize to produce a current (I). Products are then carried by convection and diffusion from the interface to the bulk of the liquid electrode. The accumulation of Li in Bi or the formation of intermetallic at the electrode-electrolyte interface causes a substantial drop in the potential of the battery due to the rise of transport over-potential. Previous experimental work by Kim et al. observed a significant decrease in capacity at high current densities due to the formation of a two-phase mixture in the positive electrode of CallBi battery [14]. The electrochemical measurements performed therein attribute the potential drop at high discharge current densities to the slow transport of Ca⁺ in the low-density Ca₁₁Bi₁₀ intermetallic forming at the electrode-electrolyte interface. Li based LMBs are subject to the formation of solid intermetallic near the electrode/electrolyte interface. Interestingly however, the diffusivity of Li⁺ ions is found to be higher in the solid LiBi, and Li₃Bi intermetallic, than liquid Bi [13]. However, since diffusion is not the sole mechanism that drives mass transport, especially in large-scale batteries, it is important to understand how the presence of fluid flow affects the overall kinetics. Additionally, knowledge of how concentration gradients are affected by fluid flow is important in order to anticipate the growth of intermetallic and prevent the risk of electrical shorts. If intermetallic grows in a manner that allows the two electrodes to come in direct contact, then they will react chemically and electrons are exchanged locally instead of being released to an external circuit, hence, the battery becomes electrically shorted.

The main focus of the present study is to develop a magnetohydrodynamicsbased model for mass transport in the positive electrode in liquid metal batteries. The present study focuses on the Li||Bi systems; however, the approach can be applied to any electrode couple given that thermodynamic data and material properties are available. Developing such models allows for experimenting with a wide range of battery parameters with minimal cost and time. Ultimately, the model can evolve into a battery management system that can be used to predict voltage, power, state of charge and state of health.

Mathematical and Numerical Model

To generate a mathematical model that describes transport in the positive electrode, we have to consider the forces acting on the electrode and understand how they generate motion. One force that can initiate mass transport is electromagnetic (Lorentz) force, which is generated from the interaction of charge or discharge current density vector \mathbf{J} with its associated magnetic field \mathbf{B} . The electromagnetic force can be determined by first solving Laplace's equation for the potential distribution in the electrode:

$$\nabla^2 V = \mathbf{0} \tag{1}$$

where ∇ is the Laplace operator and V is the electrical potential distribution. The resulting potential distribution is used to calculate the electric field (E) and current density from Eqs. (2) and (3) respectively.

$$\boldsymbol{E} = \boldsymbol{\nabla} \boldsymbol{V} \tag{2}$$

$$\boldsymbol{J} = \boldsymbol{\sigma}\boldsymbol{E} + \boldsymbol{u} \times \boldsymbol{B} \tag{3}$$

where σ is the electrical conductivity and **u** is the velocity vector. Here we consider the case where the induced current (**u** × **B**) is negligible compared to the applied Current (σE). The magnetic field is then determined from Ampere's law and the resulting electromagnetic force per unit volume is the cross product of **J** and **B** (Eqs. 4 and 5 respectively)

$$\oint \boldsymbol{B} d\boldsymbol{l} = \mu_o \oint \boldsymbol{J} d\boldsymbol{A} \tag{4}$$

$$\boldsymbol{F} = \boldsymbol{J} \times \boldsymbol{B} \tag{5}$$

where μ_o is magnetic permeability constant. In a typical liquid metal battery with cylindrical geometry, the current density vector **J** is symmetric about the central axis with components in the radial (**J**_r) and axial component (**J**_z). This will generate an azimuthal magnetic field **B**₀ and in the absence of thermal convection will tend to drive an axisymmetric flow that sinks in the center and rises near the sidewalls [15]. Note that switching from charge to discharge does not change the direction of the electromagnetic force.

Another force that can initiate mass transport is thermal buoyancy. Buoyant forces arise whenever temperature gradients are generated in a body of fluid: a recirculating flow is created in which gravitational energy is converted into kinetic energy. However, these forces are also opposed by viscosity, and in the case where viscous forces are strong enough, motion can be completely suppressed. The onset at which buoyancy overcomes viscosity is predicted by a dimensionless quantity that represents their relative ratio as

$$Ra = \frac{g\beta\Delta TL^3}{v\alpha} \tag{6}$$

where β is the coefficient of thermal expansion, g is the gravitational acceleration, ΔT is the temperature difference, L is the thickness of the electrode, v is the kinematic viscosity and α is the thermal diffusivity. This dimensionless quantity is called the Rayleigh number and the transition from viscous to buoyancy-dominated regime is called the Rayleigh-Bénard instability [15]. For geometries relevant to liquid metal batteries with one free surface, the Rayleigh-Bénard instability occurs at a critical value of Ra_c ~ 1100, though the exact value depends on geometric details of the fluid vessel [15].

Typical liquid metal batteries require heating a positive current collector from the bottom. Consequently, temperature gradients are generated along the depth of each fluid layer. Moreover, with the use of imperfect thermal insulation, we expect the bottom fluid (cathode) to be hotter near the center and colder near the edges. Therefore, we expect thermal buoyancy to drive a flow in which fluid elements rise near the central axis and sink near the edges [16]. Combining all terms, momentum transfer is governed by the Navier-Stokes equation, which is the application of Newton's second law to viscous fluid systems,

$$\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla)\mathbf{u} = v\nabla^2 \mathbf{u} - \frac{1}{\rho}\nabla p + \frac{1}{\rho}(J \times B) - \alpha \Delta T g \hat{z}$$
(7)

where $\frac{\partial}{\partial t}$ is the time derivative, p is pressure, ρ is the density of the electrode, and \hat{z} is a unit vector in the vertical direction. Note that buoyancy driven flow would be moving in an opposite direction to that driven by electromagnetic forces. Hence, one would naturally ask: will buoyant or electromagnetic forces dominate the flow? Current experimental and theoretical work is undertaken by our group to answer this question. However, for the purpose of our model, we shall assume that electromagnetic forces are negligible and do not affect the overall flow structure. This

assumption is supported by the theoretical work of Ejik et al. dealing with flow transitions in vacuum arc re-melting, [17] as well as the mathematical models developed by Davidson [15]. In fact, both references state that in the ideal case where temperature gradients are fixed with increasing current density, there exists a wide range of current densities at which the electromagnetic force seems to have no effect on the flow structure.

So far our discussion has focused on building a physical intuition of the forces that can initiate flow. Let us now consider how we can link chemical concentration with fluid flow. Changes in chemical concentration are described by the convection-diffusion equation as

$$\frac{\partial c}{\partial t} = \nabla \cdot (\boldsymbol{D} \nabla \boldsymbol{c}) - \nabla \cdot (\boldsymbol{u} \boldsymbol{c})$$
(8)

where c is the concentration profile, D is the material diffusivity, and u is the velocity vector. One approach is to sequentially solve Eqs. (7) and (8). Clearly, this is not an easy task since solving the momentum equation by itself is computationally challenging. An alternative is to use a stream function that approximates the dynamics in the core and neglects boundary layer effects. A time independent stream function (Ψ) that represents symmetric convection rolls is written as

$$\Psi \sim \sin \frac{2\pi x}{d} \sin \frac{\pi y}{L} \tag{9}$$

where x and y are horizontal and axial spatial coordinates, respectively, and d is the diameter of the positive electrode. It is important to note that a stream function will always satisfy the incompressibility condition $(\nabla \cdot \boldsymbol{u} = \boldsymbol{0})$. This will further simplify Eq. (8), and guarantees the conservation of mass within the electrode. In each simulation run, the velocity profile $(\boldsymbol{u} = U(\nabla \times \Psi))$ is considered an input in the convection-diffusion solver, where U is the characteristic flow speed. The overall flow field is shown in Fig. 2. The operating current using Faraday's law sets the flux across the interface:

$$\mathbf{C} = \frac{\mathbf{J} \, \mathrm{dt}}{n_e F} \tag{10}$$



Fig. 2 Flow field specified by Eq. 9 and intended to model thermal convection inside a bismuth electrode

where C is the molar concentration (mol/cm^2) , n_e is the number of electrons exchanged in the reaction, and F is Faraday's constant (96,485 C mol⁻¹). The numerical solver uses first-order finite differences for spatial derivatives and a second-order Adams-Bashforth scheme for time derivatives. The employed boundary conditions are constant flux across the cathode-electrolyte interface, which represents a battery discharged at a constant current, and no-flux across other walls.

Previously published electromotive force data [12, 13] are used to estimate the open circuit voltage at different molar fraction (X_{Li}) . The molar fraction and the corresponding open circuit voltage are calculated using Eqs. 11 and 12 respectively

$$X_{Li} = \frac{C_{Li}}{C_{Li} + C_{Bi}} \tag{11}$$

$$V(\mathbf{X}_{Li}) = \begin{cases} \frac{0.67}{\mathbf{X}_{Li}} & for \, \mathbf{X}_{Li} < 0.3\\ 0.75 & for \, 0.3 \le \mathbf{X}_{Li} < 0.74\\ 0.05 & for \, \mathbf{X}_{Li} > 0.74 \end{cases}$$
(12)

where C_{Li} and C_{Bi} are the molar concentration of Li and Bi respectively, and $V(X_{Li})$ is open circuit voltage. The operating temperature is chosen to be 400 °C, and the electrolyte layer is composed of a eutectic mixture of LiCl and KCl. The change in diffusivity with concentration is accounted for in the model, and the overall diffusivity D in cm²/s is found in the work of Newhouse et al. [13] as

$$D = e^{\frac{(-36.57X_{Li} - 4.27)}{(X_{Li}^2 + 3.07X_{Li} + 0.5)}}$$
(13)

Results and Discussion

We first choose an electrode thickness such that the Rayleigh number in Eq. (6) is greater than the estimated critical value of 1100 even when temperature gradients are as low as 2 °C. For this case, we choose a thickness of 1.5 cm in a 10 cm diameter container with a 6 cm foam negative current collector similar to those observed in previously published work [4]. The phase diagram of Li–Bi predicts the onset of the two-phase region at $X_{Li} < 30\%$. Hence, knowing that a total of 5.6 mol of Bismuth are used in the positive electrode, and since there are 26.8 Ahr/mole, the total capacity for this all-liquid battery model is approximately 45 Ahr.

Since current travels through the path of least resistance, we anticipate that current density lines will spread little through the depth of the electrolyte. Therefore, we choose to set our constant flux boundary condition such that Li ions enter the Bi electrode from an area equivalent to that of the negative electrode. To investigate the effect of convection on the overall concentration profile of Li ions, we solved Eq. (8) with constant flux corresponding to a battery discharged at a

current density of 200 mA/cm² for 10^4 s (total discharge capacity 43 Ahr). Initially, the cathode was pure Bi; at time t = 0, Li⁺ began entering the cathode via convection and diffusion. Concentration profiles for characteristic flow speeds of U = 0, 0.5 and 1 mm/s are shown in Fig. 3. The range of flow speeds is chosen to match previously observed speeds in numerical simulations [18, 19] and experimental models [16]. In the case where transport is purely driven by diffusion (U = 0), the highest concentration is observed near the electrode-electrolyte interface. When a convective flow is allowed, the maximum concentration still appears near the interface, however, the gradient is now along the direction of the flow. Due to the axisymmetric structure of the flow, the region of the central axis appears to represent a mixing barrier. Furthermore, the centers of the counter rotating vortices seem to hold lower concentrations of lithium compared to the edges. This shows that motion of Li⁺ ions is driven by convection along the edges of the vortices and by diffusion towards their centers.

To follow the development of the concentration profile, we fixed convection speed to 1 mm/s and plotted snapshots of Li^+ concentration at different times. Figure 4 demonstrates that ions move much more slowly towards the centers of the vortices compared to their edges. If growth of intermetallic were allowed, then one would anticipate the direction of propagation of solid fronts to be in the same



Fig. 3 Concentration of Li⁺ in a Bi cathode after 10^4 s with J = 200 mA/cm² and different characteristic flow speeds: 0 mm/s (top), 0.5 mm/s (middle) and 1 mm/s (bottom). Thermal convection transports Li along the top surface and down the edges of the cathode, whereas diffusion spreads Li smoothly from the negative current collector



Fig. 4 Time evolution of Li⁺ concentration in a Bi cathode at time $t = 10^2$ s (top), $t = 10^3$ (middle) and $t = 10^4$ s (bottom). In all cases, the characteristic flow speed is 1 mm/s. Li⁺ is transported primarily around the vortices, via convection

direction as the concentration gradient. Consequently, intermetallic nuclei are expected to form at the interface and grow along the edges of the vortices when thermal convection is present, and uniformly towards the bottom of the electrode in the absence of convection [4].

So far we have only considered the impact of the flow on the concentration of ions. To study its impact on battery performance, we shall calculate the over-potential associated with mass transport at different flow speeds. To do this, we will use the average concentration at the interface (C^+) to calculate the surface molar fraction (X^+) of Li⁺ in Bi using Eq. 11 and the surface potential V(X⁺) using Eq. 12. The voltage drop is then calculated by subtracting the surface potential from that of the bulk as

$$\eta_{mt} = V(X^+) - V(X_{Li}) \tag{14}$$

where η_{mt} is the voltage drop due to the accumulation of products at the electrode-electrolyte interface. The above expression takes into account local changes in the chemical activity of Li. Another possible way to calculate the over potential is to use the Nernst equation and assume that activity does not change significantly. However, since EMF measurements are available, it is more accurate to use the expression in Eq. (12). This approach was also used by Newhouse to calculate transport over-potentials in both LillBi and MgllSb systems [13].

In Fig. 5, we calculated mass transport over-potential as a function of time for different characteristic flow speeds. The rapid drop in over-potential with time is expected since ions initially accumulate at the interface and then get swept away by either diffusion or convection. Introducing fluid flow is found to lower the overall voltage losses from the accumulation of Li near the interface of the Bi electrode. However, the overall changes in voltage losses are low, especially as the discharge time increases. This is attributed to the high diffusivity of Li in Bi. To compare η_{mt} with the voltage drop due to the resistance of the electrolyte, an ionic conductivity of 1.27 S/cm, and an inter-electrode distance of 0.7 cm are used to calculate an overall ohmic drop of 0.12 V at a discharge current density of 200 mA/cm² [20]. Here, we have neglected any volume changes that might affect the distance between the two electrodes. The final step to estimate the discharge voltage of the battery is to subtract the ohmic drop and transport over-voltage from the open circuit voltage in Eq. 12. The resulting discharge voltage profile is shown in Fig. 6. The initial discharge voltage is only slightly higher for the electrode with a 2 mm/s flow compared to that where transport is purely driven by diffusion.



Fig. 5 Transport over-potential varying over time, at different characteristic flow speeds. $J = 200 \text{ mA/cm}^2$ in all cases, and the inset shows rapid over-potential drop during the first few seconds after discharge begins. Higher characteristic flow speeds reduce transport over-potential



Fig. 6 Estimated discharge voltage for Li||LiCl,KCl||Bi with transport in the cathode driven by diffusion (dashed line) and 2 mm/s flow (solid blue)

Conclusion

A two-dimensional convection-diffusion model was developed to link fluid flow to battery electrochemistry. Using a simplified stream function to represent thermal convection, we studied the development of concentration profile at different flow speeds and calculated the overall kinetic losses from mass transport. Increasing the characteristic flow speed caused a slight decrease in transport over-potential. However, due to the high diffusivity of Li in Bi, the overall voltage drop due to mass transport was found to be insignificant. Therefore, our findings suggest that at parameters typical for liquid metal batteries, LillBi systems are kinetically limited by ohmic losses in the electrolyte and not mass transport in the cathode. Future research will focus on comparing experimental discharge voltage profiles with model predictions as well as exploring the impact of flow structure and speed on the growth of intermetallic.

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