ResonantAcoustic® Mixing for Lithium-ion Battery Manufacture

This paper describes work to investigate the application of ResonantAcoustic® Mixing (RAM) for blending materials used in the formation of Li-ion battery electrodes. The experiments demonstrated that the mixing process is rapid, e.g. requiring less than one minute mixing time, compared to six hours typically necessary to mix this product. Post-mix analysis showed that the mixture was very uniform, fully meeting the product specification. Moreover, this complex mix formulation demonstrated that the RAM technology was capable of powder-powder mixing, powder coating, and powder-resin blending without the need for any system modification.

Material Formation Process

In Li-ion battery production the mixing process plays a critical role in determining the quality of the electrode materials, and hence, the battery electrochemical performance. Figure 1 shows a simplified example process for synthesizing positive electrode material.
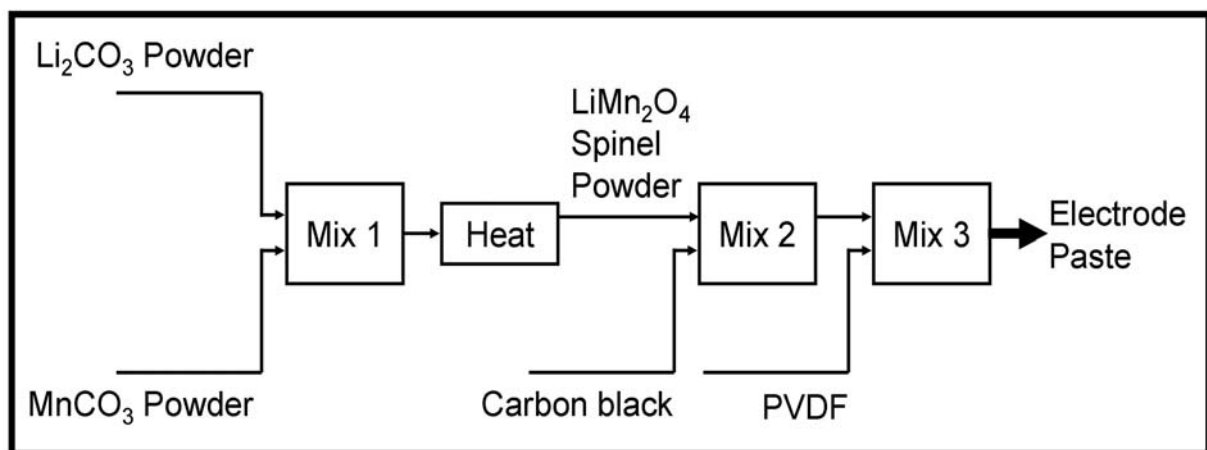

Figure 1. Simplified process for forming positive electrode material.

Powder compounds of lithium carbonate and manganese carbonate are blended and subsequently heated to approximately $750°C$ to form the $LiMn_2O_4$ spinel. The source materials have significantly different bulk densities that can often be difficult to mix, and which are susceptible to particle separation, or material segregation in conventional mixing processes that can typically take up to six hours. Uneven mixing of these materials results in localized regions of lithium-rich, or manganese-rich powders in the spinel, leading to poor cell performances.

After formulation, the $LiMn_2O_4$ powder is coated with carbon black in a subsequent dry-mixing operation before being combined with the PVDF resin binder in a third mixing operation. This material is then blended into the paste that is subsequently applied to the aluminum foil to form the positive battery electrode.

Formation of the negative electrode is a simplified operation involving only the blending of graphite powder with the PVDF resin, and coating onto the copper foil.

Mixing Test

The Resodyn Acoustic Mixers LabRAM mixer, shown in Figure 2, was used to perform the three mixing operations described above.

Figure 2. LabRAM bench-top ResonantAcoustic® Mixer.

The requisite molar masses of the two compounds were added in powder from into a sealed glass vessel and mixed on the LabRAM ResonantAcoustic® Mixer. **Mixing time was 1 minute.** (*When conventional mixing technology is employed the mixing times for this process are a few hours.*)

The combined powder was subsequently fired in a ceramic crucible at 750°C for several hours. After cooling, samples of the powder were analyzed by X-Ray diffraction in order to verify the phase of the resulting material. The 2-Theta plot shown in Figure 3 confirms all of the base materials were consumed in the reaction and that the target $LiMn_2O_4$ was formed.

A similar test using $NiMnCO_3$ as a starting material resulted in the formation of a $LiNiMnO_2$ compound that is optimized for plug-in hybrid electric vehicle (PHEV) applications. ResonantAcoustic® Mixing time was again **one minute**, and the homogenous blending confirmed by the XRD 2-Theta plot shown in Figure 4.
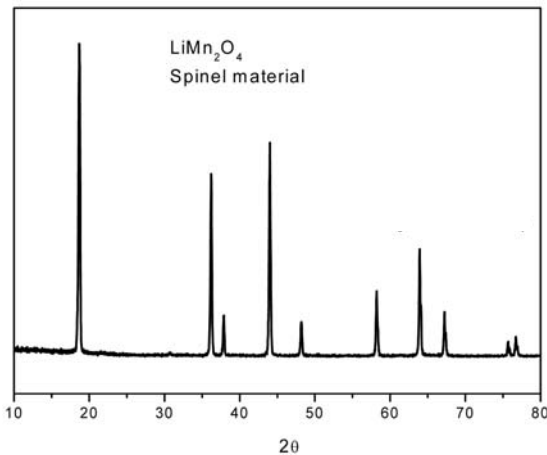

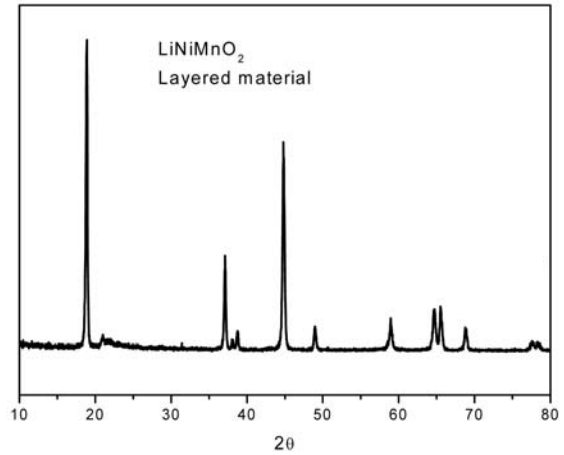Figure 3. XRD 2-Theta analysis showing formation of $LiMn_2O_4$ with no un-reacted starting materials.

Figure 4. XRD 2-Theta analysis showing formation of $LiNiMnO_2$ High Energy Density material for PHEV batteries.

The spinel powder and approximately 10% of carbon black were loaded into a vessel and mixed on the LabRAM mixer for **one minute**. Approximately 10% of PVDF binder dissolved in a NMP solvent,

Resodyn Acoustic Mixers, Inc.   130 N. Main Street, Suite 630 Butte, MT 59701
Ph: (406) 497 5333   FX: (406) 497 5206   Info@ResodynMixers.com   www. ResodynMixers.com

2

having a viscosity of approximately 10,000 cP, was added to the powder mix, and mixed again for a further **one minute**. (*When conventional mixing technology is employed the mixing times for this process are few minutes to hours, depending of the quantity of materials involved.*)

In a similar manner, graphite powder was mixed with the same PVDF binder for **one minute** to form the negative electrode paste. (*When conventional mixing technology is employed the mixing times for this process are few minutes to hours, depending of the quantity of materials involved.*)

The spinel and graphite electrode pastes were spread with a thickness of approximately 20 microns on strips of aluminum and copper foil respectively, and dried. The resulting electrodes are shown in Figure 5.
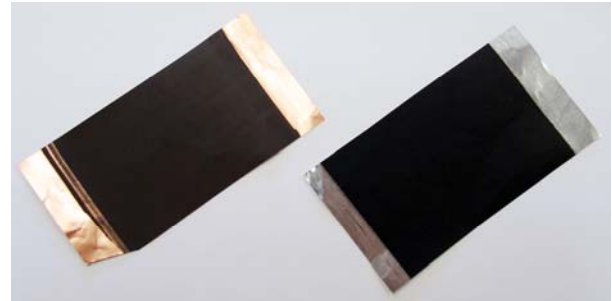
Electrochemical characterization of the electrodes produced the voltage-capacity curves shown in Figures 6 and 7 for the anode and cathode, respectively. The negative electrode (Figure. 6) generated a specific capacity of over 300 mAh/g, while the positive electrode (Figure. 7) delivered a specific capacity of over 80 mAh/g. Both

Figure 5. Copper and aluminum foils coated with electrode paste made by ResonantAcoustic® Mixing.

electrodes exhibited very stable cycling over time with high efficiency, which *attests to the high quality of both the materials and electrodes processed using the LabRAM mixer*. Under conventional mixing methods requiring hours of materials and electrodes processing, such performances typically attest to the completeness of reactions and the uniformity of electrodes. It is noteworthy that these characteristics were reached in this process evaluation in a very short time.

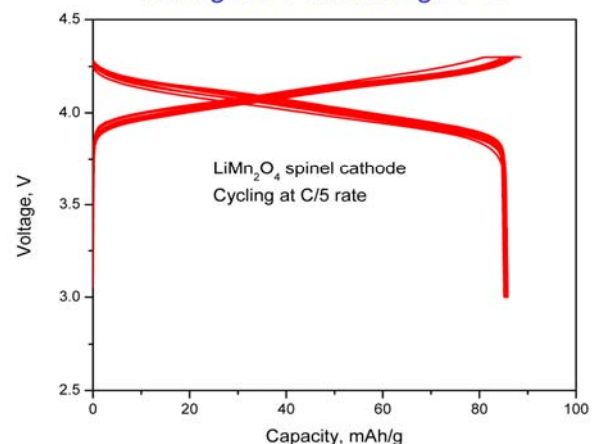Figure 6. Charge-discharge characteristics of the graphite anode cycled at C/10.

Figure 7. Charge-discharge characteristics of the LiMn$_2$O$_4$ cathode cycled at C/5.

Scalability of ResonantAcoustic® Mixing

The preceding tests were performed at the laboratory scale, but directly relate to a manufacturing process. ResonantAcoustic® Mixers are produced in three sizes: ½ pint, 5-gallon and 55-gallon maximum capacities. A unique feature of the RAM technology is that mixing can be completed on both large and small amounts of material with the similar mixing durations. For example, Figure 8 depicts mixing tests that were performed on a high viscous material at two batch sizes, 1 pint and 30 gallons; each mixed for two minutes. From these results and for other examples (see RAM Applications Note: *Scalability of Resonant-Acoustic® Mixing* for details) it is concluded that mixing time is typically independent of the amount of material being mixed.
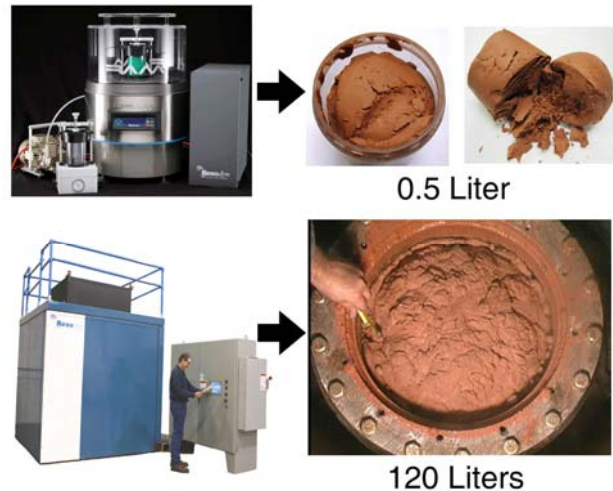

0.5 Liter

120 Liters

Figure 8. Example showing time to mix for two different size mixes for a 10,000,000 cP mix of aluminum oxide powder and light corn syrup (77% solids loading). Two minute mix time at 80 *g* of acceleration for each operation.

SUMMARY

Powder mixing, powder coating and paste blending processes required for the manufacture of Li-ion battery electrode materials have been performed on a ResonantAcoustic® Mixer and characterized by leading battery technologists. Mixing times were one minute for each operation, and analysis confirmed that completely uniform blending of the powders and the pastes had occurred. This was achieved on a single mixing device, with no hardware changes being made between mixing the various materials.

This work confirmed the efficiency and flexibility of ResonantAcoustic® Mixing as a technology that is well suited to solid, powder and liquid-phase processes. This attribute enables a single production mixer to perform multiple operations in battery manufacture, thereby maximizing mixer utilization. Mixer set-up steps are eliminated, and mixer operation is very simple; minimizing the need for maintenance, training and skilled operations staff. Finally, the dramatic reduction in ResonantAcoustic® Mixing time, compared to traditional approaches, e.g., from hours to minutes, offers Li-ion battery manufacturers a means to significantly reduce manufacturing costs.

Contact

Resodyn Acoustic Mixers, Inc.   130 N. Main Street, Suite 630 Butte, MT 59701
Ph: (406) 497 5333   FX: (406) 497 5206   Info@ResodynMixers.com   www. ResodynMixers.com

4