



Mixing of electrode slurries – shaken, not stirred

Dr. Christopher G. Clark Jr & David Manke

We are at the beginning of a green-energy renaissance, where battery technologies not only have the potential to supplant fossil-fuel powered vehicles but are expected to account for over half of new vehicles by 2030. This is an especially ambitious goal, given that most of the necessary manufacturing infrastructure needs to be built and the sector is even now reliant on inefficient technology from the 1900s for mixing electrode formulation components before coating conductive foils to form electrodes. Today, industrial planetary mixers for electrode slurry production can contain 3000 L, but nevertheless require upwards of 3h of mixing time and another 1-3 h of cleaning between batches with thousands of machines needed to meet global demand. Manual powder handling by scores of workers in hazmat suits is currently the norm, raising the required factory footprint and cost of manufacture. The Batt-TDS™, a next-generation mixing platform for high-viscosity slurries, changes the paradigm with dust-free powder induction into a continuous stream of liquid and high-productivity slurry mixing (up to more than 5000 L/hour) with as much as a ten-fold reduction in the mixing equipment footprint.

The architecture of lithium-ion batteries employs a bicontinuous network that supports electron and lithium-ion transport in separate channels.

Mixing provides two functions in the preparation of slurries

- Dispersal of conductive materials like carbon black, a nanomaterial with extremely high surface area.
- Distribution of the conductive material around the major-component active material, 50-200x larger in size. In the electrode, the conductive material serves as a conduit for electrons, whereas lithium mass transport to and from the active materials takes place via the porous structure between the active material and conductive network by the electrolyte. Cathode active materials like lithium iron phosphate, NMC and NCA, are not especially conductive. Poor dispersion and distribution translate to fewer pathways for electron conduction and thus increased resistance, potentially requiring more conductive material to solve what is otherwise a process problem, requiring a reduction in the amount of active material that can be used practically. The consequence of poor dispersion is a faster deterioration of cell performance from many charge / discharge cycles due to side reactions in the electrochemical cell, which raise the internal cell resistance over its lifetime. Practically, this would mean needing to carry a larger, heavier battery to compensate for the eventual loss in storage capacity or else the driving range is compromised in the final years of service life.

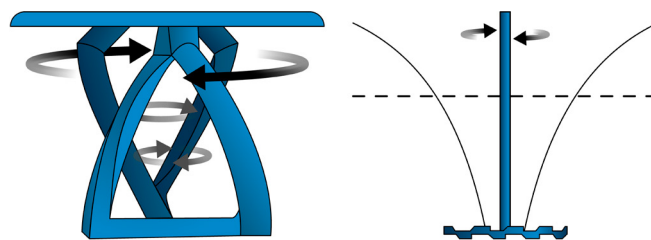


Figure 1. Illustrations of a planetary mixer and high-speed disperser/dissolver (HSD), conventional mixing machines for electrode slurries.

planetary mixer (Figure 1) requires high viscosity (millions of centipoise) to generate shear stresses needed for dispersion, consistent with Newton's Law (Equation 1). With the shear rate being determined by the rotational speed of the mixing blades, the higher the viscosity, the higher is the shear stress possible, but at very high viscosities, the motors needed to achieve motion require high torque and power. With only a fraction of its power is available for dispersion at any given time, most of the power is wasted in an inefficient process. All or most of the solvent needs to be withheld to maintain these shear stresses, contributing to wear and metal contamination from hard cathode powders. The binder is normally predissolved at elevated temperature with a high-speed disperser/dissolver (HSD, Figure 1), an inefficient powder wetting technology, cooled, filtered and added slowly to the already dry-mixed active and conductive materials. The final batch is then mixed with another set of HSD mixers, often in the same vessel with the planetary mixer as the viscosity is reduced. For a process with so many machines and process steps, it is perhaps surprising that the variables for control of slurry quality are mainly mixing times and rates.

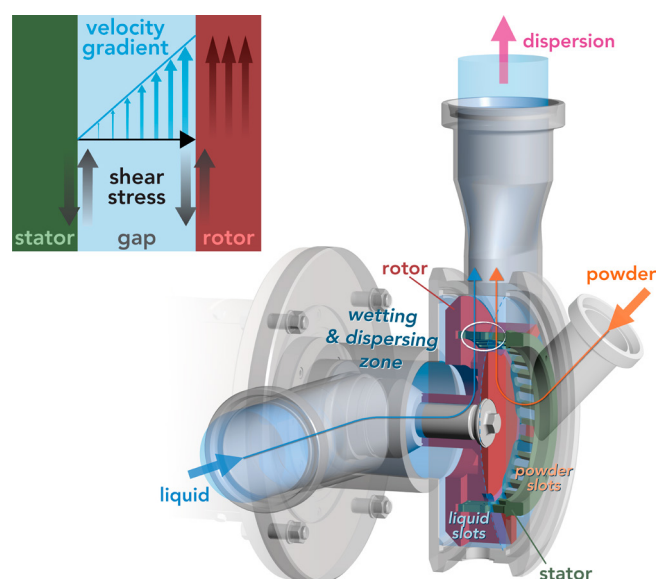


Figure 2. Illustrations of a Conti-TDS™ dispersing machine and its working principle for creating shear stresses within the rotor-stator gap.

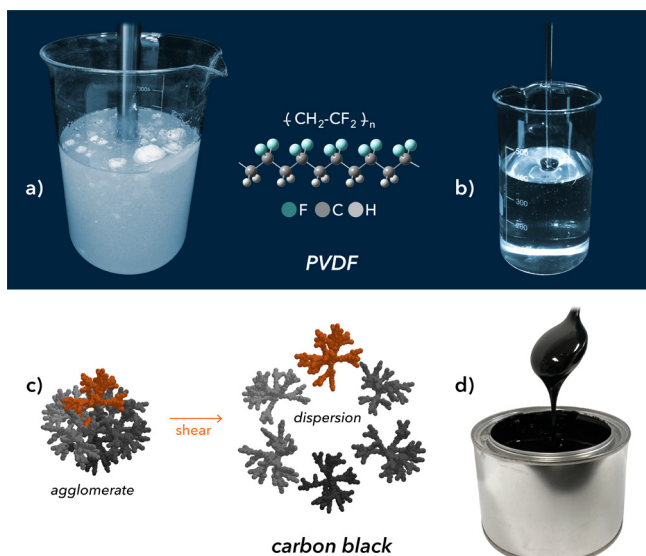


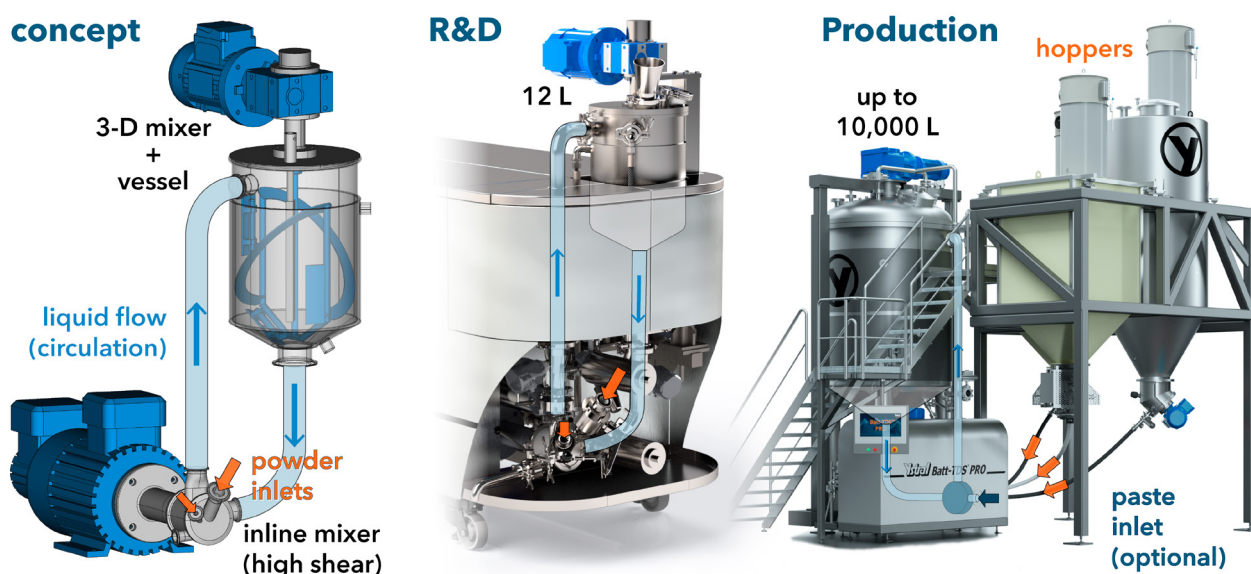
Figure 3. a) Images of an attempt to dissolve Solef 5130 (Solvay) PVDF powder (8.2 wt%) in NMP for 60 min vs b) 3 min after powder induction with a Conti-TDS (<1 min) at room temperature. c) Illustration of deagglomeration of carbon black and d) a C65 (Imerys) carbon black paste produced by dispersing carbon black in an NMP solution of PVDF (1:1 C65:Solef 5130, 11 wt% in NMP).

The Conti-TDS (Figure 2) inline mixer is a time- and energy-saving alternative to an HSD. It works by operating at a significantly higher shear rate (Equation 1), instead of requiring a high viscosity, to achieve high shear stress, which is inversely proportional to the sub-mm gap between a spinning rotor and stator (Equation 2). Powerful velocity gradients, equivalent to a shear rate, are created in this dispersing zone. Together with a dynamically generated vacuum, which propels apart the powders, these phenomena expand access to the surface area for wetting of powders into liquids. The increase in the specific power can be 10,000-fold higher and the exposure to shear stress

concentrated to a few milliseconds of dispersing time in the active mixing zone, instead of hours of conventional motor-driven mixing. By installing the rotor-stator outside the vessel, nearly all of the power can be focused on wetting and dispersion, allowing an increase in batch size to 10,000 L, significantly compacting the factory's footprint for a given productivity. They have been used successfully over the past 2 decades across numerous industries to simultaneously induct, wet, disperse and microscopically mix at powder induction rates as high as 500 kg/min. Applied to battery materials, they can disperse and dissolve binder materials such as PVDF (Figure 3b) and CMC used in cathodes and anodes in near real-time and even at room temperature, e.g. 3 min after powder induction, independent of the batch size, compared to using an HSD (Figure 3a), without requiring heating or filtration. Furthermore, they can be used to disperse carbon black (Figures 3c, 3d) orders of magnitude faster than conventional HSDs at large scale.

Rotor-stator systems have been typically limited to viscosities lower than those relevant to electrode slurries. The Batt-TDS (Figure 4) with a large stator diameter is designed to accommodate them and control the exposure time at high shear by decoupling the shear rate from the controlled powder and liquid flow rates. With the ability to generate shear stresses on demand, independent of viscosity, gentle but efficient separation of agglomerates enables powder dispersion in near real-time across a wide range of viscosities, but lower than those used by the planetary mixer. A key advantage of such a simple concept is that the shear rates can be tailored according to the process requirements of an individual powder, allowing for a shear-stress- and energy-optimized process in a sequence (Figure 5), and without requiring different types of equipment for each powder type. The system is furthermore equipped with a 3-D mixer in a large process

Figure 4. Batt-TDS™ mixing concept for electrode slurry, containing both an inline mixer for powder induction and dispersion in a stream of liquid as well as a low shear-rate 3-D mixer in the vessel for batch homogenization.



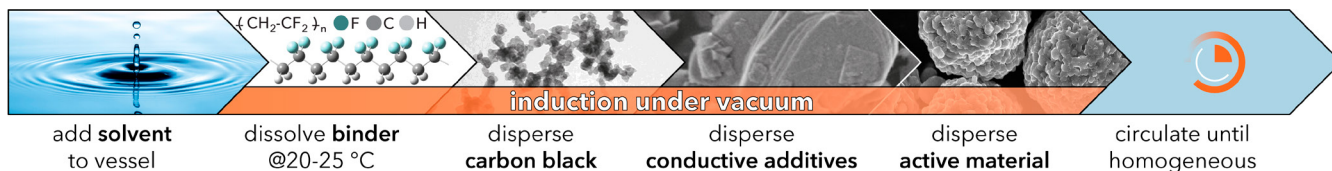


Figure 5. Process sequence for slurry preparation with a Batt-TDS.

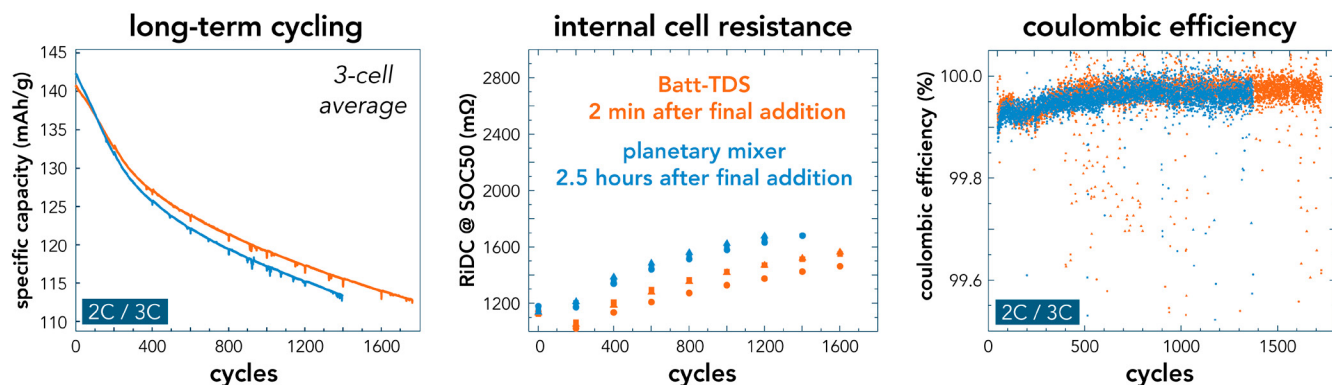


Figure 6. Performance comparison of NMC 622 /graphite cells with cathodes made with a Batt-TDS for 1 min after powder induction vs a planetary mixer for 2.5 h: a) long-term cell cycling, b) internal resistance and Coulombic efficiency with 2C/3C charge/discharge rates, respectively.

vessel, which accomplishes fast homogenization at low shear rate and power consumption during circulation through the inline mixer.

On an R&D scale, NMC 622 cathodes [92/3/3/2 wt% NMC 622 (BASF) / Solef 5130 PVDF (Solvay) / C65 carbon black (Imerys) / KS6L graphite (Imerys) / at 72 wt% in NMP (BASF)] were coated from slurries separately produced in 12-15 L batches by a Batt-TDS navi and a planetary mixer (Inoue TX-15 Trimix) with mixing times of 1 min (after powder induction) and 2.5 h, respectively. Cells made via Batt-TDS cathode slurry and an identical anode however showed a 27% increase in the long-term cycling performance over the planetary mixed materials, apparently due to improved cell resistance, with sufficient Coulombic efficiency to be used in an EV (Figure 6). Furthermore, no metal contamination from iron was found (ICP-OES, limit of detection: 8-ppm Fe) in the NMC slurry from the liquid-lubricated Batt-TDS process.

Being able to mix R&D batches in a few minutes is a tool that can produce more robust battery products from faster iteration in development cycles. Another benefit of using Batt-TDS Technology is predictable scaling behavior from 12-L batches from the R&D-scale Batt-TDS navi up to 10,000 L batches with a Batt-TDS PRO. Beyond the quality benefits, the main reason for the current timing of Batt-TDS introduction is that for the first time, factories are planned to be sufficiently large to reap significant rewards on space, and CapEx (even compared to Chinese-made planetary mixers) and operational savings of making bigger batches from far less machines (Figure 7). In addition, with large factories needing to be climate controlled for cathode processes, being able to produce such a large amount of slurry from a small space translates into large energy savings even for the building required to house the equipment.

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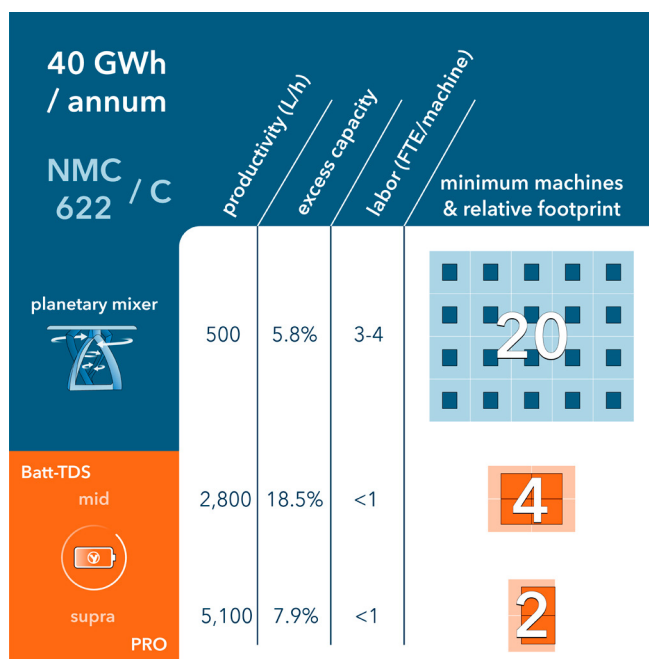


Figure 7. Comparison of estimated productivity, labor and space requirements of planetary mixers vs Batt-TDS systems for electrode slurry production at the scale for factories currently in planning, illustrated for NMC 622/graphite batteries.

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