

INTISARI

Peraturan Presiden Nomor 55 Tahun 2019 mendorong penggunaan kendaraan bermotor listrik berbasis baterai (KBLBB) di Indonesia. Komponen terpenting dari KBLBB adalah baterai litium yang memiliki umur pakai hanya 8 hingga 10 tahun dan kemudian menjadi limbah yang wajib didaur ulang. Proses daur ulang (*recycling*) menjadi krusial karena mendukung keberlanjutan material, perlindungan lingkungan, dan penerapan ekonomi sirkular. Namun, pada proses daur ulang baterai litium terdapat tantangan utama pada tahap akhir yaitu presipitasi litium dalam bentuk litium karbonat (Li_2CO_3) karena rendahnya konsentrasi litium dan tingginya kandungan pengotor seperti ion Na^+ dan SO_4^{2-} , yang berdampak pada penurunan kemurnian dan tingkat *recovery* produk. Selain itu, spesifikasi distribusi ukuran produk Li_2CO_3 yang sangat ketat turut menjadi tantangan tersendiri. Oleh karena itu, penelitian ini bertujuan mengoptimalkan kondisi presipitasi Li_2CO_3 dari daur ulang baterai litium dengan target *recovery* $\geq 70\%$, distribusi ukuran $d_{10} \geq 1 \mu\text{m}$, $3 \mu\text{m} \leq d_{50} \leq 8 \mu\text{m}$, $d_{90} \leq 15 \mu\text{m}$, dan kemurnian $\geq 99,5\%$, seperti spesifikasi pasar. Dua pendekatan digunakan, yaitu presipitasi konvensional dan berbantu ultrasound untuk mengevaluasi pengaruh konsentrasi CO_3^{2-} , SO_4^{2-} , dan suhu terhadap kemurnian dan *recovery*, serta pengaruh sistem pengadukan (konvensional dan ultrasound) terhadap distribusi ukuran produk. Analisis pemodelan termodinamika dan kinetika turut dilakukan guna memahami fenomena yang terjadi secara fundamental.

Hasil penelitian menunjukkan bahwa ion CO_3^{2-} meningkatkan *recovery* produk Li_2CO_3 , sedangkan ion SO_4^{2-} secara konsisten menurunkan kemurnian dan *recovery*. Pada rasio $\text{CO}_3^{2-}/\text{Li}^+ \leq 2,0$, kemurnian yang dihasilkan tetap memenuhi standar. Namun, pada rasio 2,5 terbentuk pengotor Na_2CO_3 yang menurunkan kemurnian menjadi 92,17%, meskipun *recovery* naik menjadi 81,64%. Konsentrasi ion SO_4^{2-} yang tinggi juga menurunkan kualitas produk, tetapi pada rasio $\text{SO}_4^{2-}/\text{Li}^+ = 0,5$ diperoleh kemurnian 99,89% dan *recovery* 80,54%. Pada rasio $\text{SO}_4^{2-}/\text{Li}^+ > 0,5$, diperlukan *pretreatment* untuk menurunkan konsentrasi ion SO_4^{2-} agar kemurnian Li_2CO_3 tetap di atas 99,5%. Selain itu, peningkatan suhu juga meningkatkan *recovery* Li_2CO_3 akibat penurunan kelarutannya pada suhu tinggi. Namun, pada konsentrasi ion SO_4^{2-} tinggi, suhu tinggi dapat memicu terbentuknya pengotor Na_2SO_4 . Suhu presipitasi yang tinggi juga berdampak negatif terhadap ukuran partikel Li_2CO_3 yang cenderung besar dan teraglomerasi sehingga tidak memenuhi standar. Pada sistem pengadukan konvensional 250 rpm, ukuran d_{10} , d_{50} , dan d_{90} masing-masing sebesar 59,33 μm ; 79,55 μm ; dan 108,52 μm . Meskipun peningkatan kecepatan pengadukan hingga 650 rpm dapat memperkecil ukuran, namun hasil akhir distribusi ukuran partikel tetap diluar standar. Sebaliknya, presipitasi berbantu ultrasound pada daya 260–320 W menghasilkan partikel sesuai spesifikasi ($d_{10} \sim 3 \mu\text{m}$, $d_{50} 5\text{--}6 \mu\text{m}$, $d_{90} < 15 \mu\text{m}$). Selain itu, prekursor Li_2CO_3 hasil presipitasi ultrasound juga terbukti meningkatkan kualitas struktur katoda NMC yang disintesis.

Penelitian ini juga mengevaluasi pemodelan termodinamika dan kinetika untuk memahami fundamental proses presipitasi, terutama dalam kontrol kemurnian, *recovery*, dan distribusi ukuran produk. Model termodinamika berbasis persamaan Gibbs *excess* dengan modifikasi Full Debye-Hückel dan *Ionic Association Term* (FDH-IAT) secara akurat memprediksi kelarutan Li_2CO_3 dalam sistem non-ideal. Model kinetika berbasis *Population Balance Model* (PBM) berhasil menggambarkan dinamika nukleasi dan pertumbuhan partikel pada sistem presipitasi berbantu ultrasound. Model termodinamika dan kinetika yang dikembangkan tersebut menghasilkan parameter yang sensitif dan general, sehingga mendukung simulasi dan *scaling-up* proses industri daur ulang baterai NMC terutama dalam meningkatkan kontrol dan prediksi kemurnian, *recovery*, dan distribusi ukuran produk Li_2CO_3 .

Kata kunci: daur ulang, presipitasi Li_2CO_3 , *recovery*, kemurnian, distribusi ukuran

ABSTRACT

Presidential Decree No. 55/2019 promotes the adoption of battery electric vehicles (BEVs) in Indonesia. The most critical component of BEVs is the lithium-ion battery, which has a limited lifespan of 8 to 10 years and eventually becomes waste that must be recycled. Recycling spent batteries is essential to support material sustainability, environmental protection, and the implementation of a circular economy. However, recycling of lithium-ion batteries faces a major challenge in the final stage, which involves the precipitation of lithium as lithium carbonate (Li_2CO_3). This challenge arises from low lithium concentration and presence of impurities such as Na^+ and SO_4^{2-} ions, which reduce both the purity and recovery rate of Li_2CO_3 . In addition, the strict specifications for Li_2CO_3 particle size distribution pose an additional challenge. This study aims to optimize the precipitation conditions for Li_2CO_3 recovery from recycled lithium batteries, targeting a recovery rate of $\geq 70\%$, a particle size distribution of $d_{10} \geq 1 \mu\text{m}$, $3 \mu\text{m} \leq d_{50} \leq 8 \mu\text{m}$, $d_{90} \leq 15 \mu\text{m}$, and a purity of $\geq 99.5\%$, in accordance with battery-grade standards. Two approaches, conventional precipitation and ultrasound-assisted precipitation, were applied to the study of Li_2CO_3 precipitation. The effects of CO_3^{2-} and SO_4^{2-} concentrations and temperature on purity and recovery rate were evaluated, along with the effects of different agitation methods (conventional and ultrasound) on particle size distribution. Thermodynamic and kinetic modeling was also conducted to understand the underlying mechanisms of the precipitation process.

The results show that CO_3^{2-} ions increase the recovery of Li_2CO_3 , while SO_4^{2-} ions consistently reduce both purity and recovery. At a $\text{CO}_3^{2-}/\text{Li}^+$ molar ratio of ≤ 2.0 , the resulting purity meets the required standard. However, at a ratio of 2.5, Na_2CO_3 impurities form, reducing the purity to 92.17% despite an increased recovery of 81.64%. A high concentration of SO_4^{2-} also lowers product quality, but at a $\text{SO}_4^{2-}/\text{Li}^+$ ratio of 0.5, a purity of 99.89% and a recovery of 80.54% were achieved. When the $\text{SO}_4^{2-}/\text{Li}^+$ ratio exceeds 0.5, pretreatment is required to reduce the SO_4^{2-} concentration in order to maintain Li_2CO_3 purity above 99.5%. Temperature also affects recovery, with higher temperatures increasing Li_2CO_3 recovery due to reduced solubility. However, in the presence of high SO_4^{2-} concentrations, elevated temperatures can lead to the formation of Na_2SO_4 impurities. In addition, high precipitation temperatures negatively affect particle size, resulting in large and agglomerated Li_2CO_3 particles that do not meet specifications. Under conventional agitation at 250 rpm, the resulting particle sizes were d_{10} 59.33 μm , d_{50} 79.55 μm , and d_{90} 108.52 μm . Although increasing the agitation speed reduced particle size, the final distribution still did not meet the standard. In contrast, ultrasound-assisted precipitation at 260–320 W produced particles that met specifications ($d_{10} \sim 3 \mu\text{m}$, d_{50} 5–6 μm , and $d_{90} < 15 \mu\text{m}$). Furthermore, Li_2CO_3 precursors obtained through ultrasound-assisted precipitation were shown to enhance the structural quality of synthesized NMC-111 cathodes.

The study also included thermodynamic and kinetic modeling to better understand the Li_2CO_3 precipitation process, particularly to control purity, recovery, and particle size distribution. The thermodynamic model, based on the excess Gibbs free energy equation with Full Debye-Hückel and Ionic Association Term (FDH-IAT) modifications, accurately predicted Li_2CO_3 solubility in non-ideal systems. The kinetic model, using the Population Balance Model (PBM), successfully described nucleation and particle growth dynamics in the ultrasound-assisted system. These models provided sensitive and general parameters that support simulation and industrial scale-up of NMC battery recycling processes, especially for enhancing control and prediction of Li_2CO_3 purity, recovery, and size distribution.

Keywords: recycling, Li_2CO_3 precipitation, recovery, purity, particle size distribution