Validating the direct aqueous attrition-leaching carbonation process by coupled thermodynamic and kinetic modelling

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The direct aqueous attrition-leaching process is an under development mineralization process proposed by the authors (Julcour et al., 2014, Chem. Eng. J., 262, 716-726). It was conceived as a robust additive-free means to overcome the formation of passivation layers at the surface of leached particles, which is a severe impediment to direct aqueous mineralization. It merges the leaching conditions necessary to process Mg, Ca or Fe bearing materials with the attrition efficiency of a stirred media mill, a process that is scalable to handle large industrial throughputs commensurate with mitigation of CO₂ emissions. Up to this day, the attrition-leaching process has been validated under a CO₂ partial pressure of 20 bars and a temperature of 180 °C to produce stable carbonate solid phases. Starting with minus 100 µm particles, the process has demonstrated a carbonation efficiency in excess of 80% for a variety of natural silicate ores and mining wastes. With the initial batch process being currently modified to operate continuously, deciding upon the most suitable operating conditions is critical, in terms of energy and rate efficiency.

The complexity and diversity of the reactive system eliminates the possibility of deciding firsthand upon the most energy efficient operating conditions. Fortunately, by maintaining the surface of reacting particles in an unpassivated state by continuous mechanical removal of surface leach layers, attrition-leaching process reactions can be modelled from thermodynamic equilibria combined with fresh surface dissolution kinetics throughout the duration of the process. The proposed modelling approach merges the properties of the ore and the process operating conditions, thereby providing a design tool for the direct aqueous attrition-leaching carbonation process.

Using a set of controlled laboratory experiments, the paper confirms the ability of geochemical modelling to predict the attrition-leaching process performance for silicate ores, an abundant feedstock for mitigating CO₂ emissions. Within the 5 to 30 bar CO₂ partial pressure range and the 90 to 200 °C temperature range, geochemical modelling predicts radically different mineralization performance due to the appearance of phyllosilicate phases such as talc, whose precipitation is very sensitive to both CO₂ partial pressure and temperature. To a lesser extent, minor elements present in the ores, especially Al also influence the carbonation yield and the nature of the phyllosilicates.

Geochemical modeling proved to be of value for selecting the right grinding medium used by the attrition-leaching process. Indeed, under the operating CO₂ partial pressure and temperature, many grinding media can become reactive and prove inadequate. As an example, geochemical modelling predicted that steel grinding beads would convert to siderite (FeCO₃) under a wide range of operating conditions. As this was verified experimentally, it led the authors to revisit this initial choice of grinding medium for the attrition-leaching process and select SiC beads as a suitable compromise in terms of attrition efficiency and reaction innocuity. Predictions are also made about
the possibility for using the attrition-leaching process in autogenous mode, which is desirable from an operational standpoint.

With leaching being a surface phenomenon, particle size distribution (PSD) was fed to the geochemical model through a series of discrete size classes following the Rosin-Rammler distribution of the feed material. Using dissolution rate data from the literature and subjecting individual size classes to dissolution of the shrinking particle type, model predictions satisfactorily matched the measured carbon dioxide consumption by the carbonation reaction for different mineral carbonation feedstocks, namely olivine, harzburgite and nickel slags. The agreement between predicted and measured carbonation rates is evidence that the process is indeed driven by comminution through surface particle attrition rather than particle breakage, as intended. Overall, the approach provided insights and guidance about the effect of particle size on mineralization rate. It was used to reflect upon the possibility of applying autogenous attrition to the attrition-leaching process, where feedstock particles are used both as the grinding and the reactive media.

Finally, coupling geochemical prediction of thermodynamic equilibria and PSD-driven dissolution kinetics, the paper ends with a complete set of geochemistry-based specifications for the attrition-leaching mineral carbonation process, which is currently being used to commission a 100 L/hour continuous demonstrator. The reliability with which geochemical modeling can predict the outcome of complex aqueous mineralization systems confirms that geochemical modelling is a great asset for designing the attrition-leaching mineralization process. By extension, it is thought to be of great value for designing any aqueous mineralization process.

Geochemistry-based modelling framework of the attrition-leaching process design