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Carbon Dioxide Removal via pH-Controlled Mineralization of Coal Fly Ash: Extraction–Carbonation Optimization, Impurity Control, and U.S.-Scale Potential

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Since the industrial revolution, CO_2 has been accumulating in the atmosphere and industrial wastes have been released into the environment, polluting ecosystems. Coal power plants in particular have been producing millions of tons of coal fly ash (CFA), most of it accumulated in pounds or landfills, which can lead to environmental hazards.

This research investigates the indirect carbonation potential of three U.S. CFA feedstocks: Scherer (Georgia), Miller (Alabama), and Cooper (Kentucky), spanning subbituminous and bituminous coal types and varying physicochemical properties. This indirect carbonation process involves four steps: calcium extraction at low pH, leachate neutralization, impurities filtration, and carbonation. A systematic leaching study across mineral acids, organic acids, amino acid, and ammonium salts reveals that final leachate pH, rather than reagent identity or concentration, governs calcium extraction efficiency (17-99%) across all conditions tested. CFA mineralogy emerged as the critical factor controlling impurity mobilization during neutralization. Indeed, the addition of base to the leachate of the weathered CFA from bituminous coal triggers iron and aluminum hydroxide precipitation and silica polymerization across all leachate at pH ≤ 6.5, while for leachates from fresh CFA from subbituminous coal it leads to a clean solution at pH ≥ 3.5. Constant-pH carbonation at pH 7.5, ambient temperature, and atmospheric pressure achieved 97–99% carbonation efficiency across all three feedstocks after water purification, creating high-value precipitated calcium carbonate. This work establishes pH and CFA mineralogy as the two governing parameters for process design, providing a replicable mechanistic framework for converting heterogeneous industrial waste into durable, high-value carbonates without high-pressure or high-temperature processing.

The deployment at scale of indirect carbonation using CFA feedstocks depends on the surrounding resources. Here, the sample sites are used as case studies, where they are mapped along with nearby sources of industrial or atmospheric CO_2 and utilization opportunities in the building and paper industries. This model could be replicated at other CFA disposal locations elsewhere in the United States and globally. When optimized, indirect carbonation of CFA can deliver a triple benefit: achieve the long-term storage of CO_2, remediate hazardous waste, and create a marketable product.