

Study on Carbonation of Ultramafic Tailings

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Abstract. As global industrialization leads to an increase in atmospheric CO₂ concentration, causing environmental issues, reducing CO₂ emissions has become a consensus. CO₂ mineralization storage technology has garnered attention, with ultrabasic tailings being ideal raw materials due to their rich content of magnesium, iron, and other elements. This study centers on the carbonation of ultrabasic tailings, investigating its kinetic mechanisms and optimizing conditions. The research framework encompasses the kinetics of direct aqueous carbonation of olivine under a CO₂ partial pressure of 6.5 MPa, the mechanism of mechanical activation of multiphase ultrabasic tailings, and the feasibility study of ultrabasic mine exploitation in conjunction with CO₂ mineralization storage technology. In terms of research methods, olivine from Washington State, USA, and tailings from northern British Columbia, Canada, were selected as materials. Experiments were carried out using a stirred autoclave manufactured by Parr Instrument Company (USA), with additional equipment such as a laser diffraction particle size analyzer employed for characterization purposes. Quantitative analysis of product composition and chemical kinetic theory were used to analyze the reaction. The research results show that optimizing conditions under low CO₂ partial pressure can significantly enhance the degree of olivine carbonation, and mechanical activation can increase the reactivity of minerals, providing theoretical support for the promotion of ultrabasic mine exploitation in conjunction with CO₂ mineralization storage technology

Keywords: Ultramafic Tailings, CO₂ Mineralization Storage, Mechanical Activation, Carbonation Kinetics, Technical Feasibility

1. Introduction

As global industrialization accelerates, atmospheric CO₂ concentrations continue to climb, culminating in increasingly severe environmental repercussions such as the greenhouse effect and ocean acidification. To mitigate climate change, reducing CO₂ emissions has become an international consensus. In 2016, the United Nations signed the Paris Agreement, aiming to limit the global temperature increase to well below 2°C [1]. CO₂ mineralization storage technology, a form of CCUS (Carbon Capture, Utilization, and Storage), can effectively reduce CO₂ emissions and has garnered widespread attention in recent years. Ultramafic tailings, abundant in elements such as magnesium and iron, have emerged as optimal feedstocks for CO₂ mineralization and storage. This study aims to investigate the kinetic mechanisms and optimization conditions of ultramafic tailings

carbonation, providing theoretical support for the promotion of synergistic technology integrating ultramafic mining and CO₂ mineralization storage. Research on tailings carbonation not only contributes to the efficient utilization of tailings resources and the reduction of environmental pollution and ecological damage, but also advances the development of carbon capture and storage technology to lower carbon emissions in response to climate change. Furthermore, it promotes the development of the circular economy and the green building materials industry, ultimately creating a more environmentally friendly and sustainable living environment. This study will address the following aspects: research on tailings carbonation technology, research content and methodology, current research directions in tailings carbonation, and the current status of CO₂ mineralization and storage development using ultramafic tailings.

2. Overview of research on tailings carbonation technology

2.1. Occurrence status of ultramafic rocks and research progress on accelerated carbonation

Ultramafic rocks are primarily composed of olivine, pyroxene, and their alteration products (e.g., serpentine). Their formation is closely associated with tectonic activity [2], resulting in typically clustered, zonal distributions [Fig. 1]. Studies demonstrate that olivine and serpentine within ultramafic rocks can react with CO₂ under suitable conditions to form carbonate minerals, enabling permanent CO₂ storage [3]. CO₂ mineralization storage technology achieves stable sequestration by converting CO₂ into solid carbonate minerals. This can be implemented via two approaches: In situ carbonation: Direct injection of CO₂ into subsurface ultramafic formations. While cost-effective, this method offers limited control over reaction conditions and is constrained by formation characteristics. Ex situ carbonation: Accelerated carbonation in engineered reactors within industrial processes. This provides superior controllability and can inform the design of in situ methods. Accelerated carbonation processes are further classified into direct and indirect methods, each subdivided into aqueous or anhydrous routes based on water participation: Direct method: CO₂ dissolution, Mg²⁺ leaching, and Mg-carbonate precipitation occur within a single reactor. Indirect method: Mg²⁺ is first extracted from silicates in a separate step before reacting with CO₂ to form Mg-carbonates [4]. Current research on accelerated carbonation of ultramafic rocks predominantly focuses on monomineralic olivine and serpentine systems, while studies on polymineralic ultramafic tailings remain relatively limited.

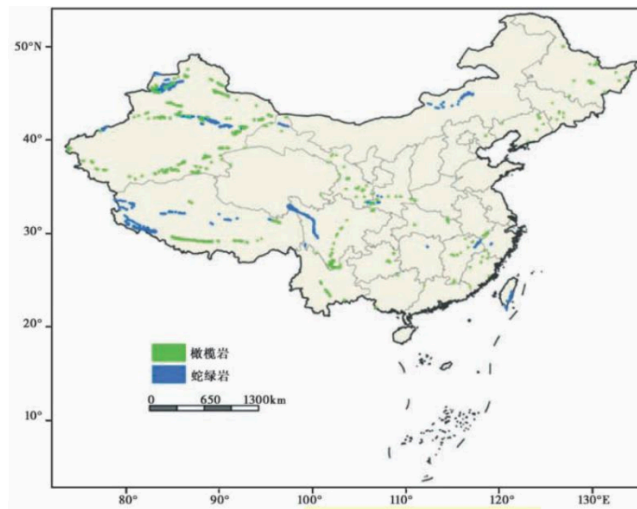


Figure 1. Schematic diagram of the distribution of ultramafic rock bodies in China. Base map data sourced from Google Earth

2.2. Mechanical force activation technology

Mechanical activation is a method that alters mineral structures and properties through physical means. Research on mechanical pretreatment of ultramafic rocks primarily focuses on monomineralic systems [5,6] (e.g., olivine, serpentine), investigating activation mechanisms of surface and lattice structures under varying mechanical energy input [7,8], grinding equipment (such as planetary ball mills, vibratory ball mills, attritor mills, tumbling ball mills, etc.) [6,9], grinding media (water, ethanol, acids) [10,11], atmosphere (e.g., N_2 , CO_2 , air) [12], and temperature [13]. Studies demonstrate that mechanical activation significantly reduces particle size, increases specific surface area, and promotes lattice distortion, thereby enhancing mineral reactivity. In the field of ultramafic tailings carbonation, mechanical activation technology holds broad application prospects.

3. Common research methods

Research on ultramafic tailings carbonation comprises two main components: research content and methodology. For research content: (1) Kinetic studies of direct aqueous olivine carbonation under 6.5 MPa CO_2 partial pressure, aiming to elucidate kinetic mechanisms of direct aqueous olivine carbonation under low CO_2 partial pressure conditions; (2) Investigation of mechanical activation mechanisms in polymineralic ultramafic tailings, examining alterations during mechanical activation and their impact on carbonation efficiency; (3) Feasibility study of integrated ultramafic mining with CO_2 mineralization storage technology, evaluating economic viability and environmental benefits of tailings carbonation. Methodologically: Experimental materials included olivine from the Twin Sister ultramafic deposit (Washington, USA) and tailings from Hard Creek Nickel Corp.'s Turnagain deposit (Northern British Columbia, Canada); Carbonation experiments employed a 100mL stirred Hastelloy-4566C autoclave (Parr Instrument Company, USA); Characterization utilized laser diffraction particle sizing (Mastersizer2000, Malvern, UK), N_2 adsorption-desorption at $-196^\circ C$ (Autosorb-1MP, Quantachrome, USA), X-ray diffraction (Bruker AXS GmbH, Germany), and scanning electron microscopy (Helios Nanolab 650, Eindhoven, Netherlands); Data analysis involved quantifying chemical composition of carbonation products to determine carbonation

efficiency, and applying chemical kinetic theories to solution ion concentrations for reaction mechanism analysis.

4. Current research directions and analysis of tailings carbonation

4.1. Study on the phenolization kinetics of *Canarium album*

Under lower CO₂ partial pressure conditions, olivine carbonation efficiency is influenced by multiple factors. Optimization of the concentration of NaHCO₃/NaCl buffer solutions and carbonation duration markedly enhanced the carbonation efficiency of olivine. The study revealed that under lower CO₂ partial pressure conditions, higher CO₃²⁻ concentration and pH in the solution facilitate carbonate precipitation formation [12].

4.2. Study on the mechanochemical activation mechanism of polymorphic ultrabasic tailings

Mechanical activation can significantly alter the particle size, specific surface area, and microstructure of tailings. Activated tailings exhibit reduced particle size, increased specific surface area, and enhanced lattice distortion in olivine, thereby improving the carbonation reactivity of the tailings. However, mechanical activation has minimal impact on the serpentine lattice [13].

4.3. Technical feasibility analysis

Analysis based on full life-cycle cost modeling and carbon balance modeling demonstrates [14] that under optimized conditions, tailings carbonation technology exhibits low operating costs and significant environmental benefits. At a carbonation tax rate of \$200 per ton of CO₂, the case study mine achieves profitability by implementing tailings carbonation technology.

5. Current development status of CO₂ mineral sequestration using ultramafic tailings

5.1. Ultramafic tailings mineralization carbon sequestration technology

5.1.1. Ultramafic tailings mineralization CO₂ sequestration technology (foreign - UBC)

In-situ carbonation technology in tailings ponds is applicable to both closed and active facilities, with advantages including ambient temperature/pressure operation and low energy consumption. The principle entails the natural weathering of tailings, which gradually undergo carbonation upon exposure to Earth's atmosphere, hydrosphere, and biosphere without substantial human intervention. Concurrently, carbonation can be accelerated via CO₂ injection, optimization of the specific surface area of tailings, periodic water spraying, and microbial augmentation. Currently, this technology remains at theoretical, laboratory, and simulation research stages, with Canada and Australia conducting the most active research in this field.

5.1.2. Ultramafic tailings mineralization CO₂ sequestration technology (domestic - University of Science and Technology Beijing)

The calcium-magnesium-based solid waste block material proposed by University of Science and Technology Beijing utilizes solid wastes with high Ca/Mg content and low hydration activity as cementitious materials, combined with tailings sand as aggregate to form backfill slurry. This slurry is pumped via pipelines to backfill mined-out areas. After sealing the filled zones, CO₂ is injected

for carbonation curing to produce hardened bodies with carbonate binding phases. Real-time monitoring ensures rock mass stability and backfill performance compliance. Currently, this technology is at theoretical, laboratory, and simulation research stages [15].

5.2. Current application status of carbonized building materials

5.2.1. Current application status of carbonized building materials (international)

In the field of carbon capture and utilization technologies, diverse institutions and projects have developed distinctive research and practices: Carbure Technology injects concentrated CO₂ into cement to enhance concrete performance, achieving industrialization; US-based Carbonfree employs aqueous carbonation to convert flue gas CO₂ into precipitated calcium carbonate (PCC), capturing and mineralizing 50,000 tonnes of CO₂ annually; Canada's CO₂MENT Project utilizes solid sorbents to capture CO₂ for steel slag carbonation, demonstrating pilot-scale implementation at a Canadian cement plant; Europe's STEPWISE Project applies aqueous steel slag carbonation for CO₂ capture in the steel industry, validated at pilot-scale in Tata Steel's Dutch facility; UK's Carbon8 Systems treats industrial exhaust gases through aqueous solidification using industrial waste to generate stable carbonates, completing multiple carbonation demonstrations with steel slag and other wastes; Norway's International Mineral Carbonation Research Centre focuses on converting steel industry solid waste into high-strength building materials, currently at laboratory research stage.

5.2.2. Current application status of carbonized building materials (Domestic)

On July 31, 2021, Baotou Steel Group launched Phase 1-Stage 2 of its carbonated steel slag comprehensive utilization project – a 100,000-tonne demonstration industrialization initiative (globally first integrated solid waste and CO₂ mineralization system). On December 25, 2022, Yuancu Technology utilized industrial solid wastes (carbide slag and steel slag) with proprietary cyclic medium solution to react with CO₂, producing high-purity micrometer-grade green calcium carbonate. On July 18, 2023, Jingyun Taibo New Materials Technology Co., Ltd. commissioned the world's first 10,000-tonne industrial pilot plant for direct CO₂ utilization (mineralizing bulk solid wastes like steel slag with flue gas containing $\geq 10\%$ CO₂ to produce mineralized building materials). Given the scarcity of direct flue gas carbonation curing applications for building materials in the steel slag industry, and variations in flue gas composition/concentration across steel plants, investigating the carbonation consolidation mechanisms of steel slag under actual steel plant flue gas conditions provides critical guidance for engineering demonstrations.

6. Conclusions

This paper systematically investigates the mechanochemical activation pretreatment and direct aqueous carbonation kinetics mechanisms of ultramafic tailings through extensive literature review. It conducts a feasibility study on the synergistic technology of ultramafic mining and CO₂ mineralization storage. The research reveals the kinetics mechanism of direct aqueous olivine carbonation under low CO₂ partial pressure and the influence mechanism of mechanochemical activation of multiphase ultramafic tailings on the carbonation degree. A comprehensive literature analysis substantiates the economic viability of tailings carbonation technology.

Future research will further optimize the technical conditions for tailings carbonation and explore its application potential in different mining contexts. Concurrently, studies will strengthen research on the applications of tailings carbonation products to advance the industrialization process of this

technology. Additionally, attention will be given to environmental impact assessments and risk management research during the tailings carbonation process to ensure the sustainable development of the technology. Meanwhile, this study acknowledges its limitations, including the use of a finite number of literature sources and a lack of empirical research. Future studies are encouraged to incorporate a broader range of literature and integrate practical applications to conduct comprehensive and systematic research in this field.

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