

氏 名：姜 天琦

論 文 名：DEVELOPMENT OF BACKFILL MATERIAL FOR CARBON DIOXIDE STORAGE  
BY USING RECYCLED CONCRETE AGGREGATES AT GOAFS IN CHINESE  
UNDERGROUND COAL MINES

(コンクリート再生骨材を用いた中国の坑内掘り石炭鉱山跡地への二酸化炭素  
貯留を目的とした充填材の開発)

区 分：甲

### 論 文 内 容 の 要 旨

After the capture and transportation process, CO<sub>2</sub> can be disposed of in natural locations, such as deep geological storage, mineral carbonization, or ocean storage. Three geological structures are also considered to be the main potential CO<sub>2</sub> sinks: deep saline sediments, depleted oil and gas reservoirs, and unmineable coal seams. In addition, research on the unique properties of micro-nanobubbles (MNBs) has increased significantly, and has attracted extensive attention in advanced scientific and technological fields such as CO<sub>2</sub> enhanced oil recovery and carbon neutrality and carbon storage systems. MNBs exhibit special physical and chemical properties due to their micro/nano-sized diameters, which accelerate the mass transfer efficiency at the gas-liquid interface and promote chemical reactions and physical adsorption. Its preparation materials are simple, low-cost, and have little impact on the environment, which may be a promising solution to the low rate and efficiency of CO<sub>2</sub> mineralization storage. Based on the above background, this study aims to develop a new CO<sub>2</sub> goaf mineralization storage technology based on CO<sub>2</sub> MNB and CO<sub>2</sub> mineralization storage, especially to enhance the mechanical stability of coal pillars and prevent the occurrence of CO<sub>2</sub> MNB leakage disasters. Therefore, this study uses laboratory experiments, numerical simulations, field surveys and analysis methods to study crack development and deterioration characteristics of the coal pillars, development of carbon dioxide storage functional backfill material for coal pillar reinforcement, and the carbon dioxide leakage prevention. The main contents of each chapter are as follows:

**Chapter 1:** Introduction to CO<sub>2</sub> storage technology and existing goaf reuse methods. The history, technology and types of goaf functional transformation are reviewed, and the advantages and application scenarios of different CO<sub>2</sub> underground storage methods are briefly compared and analyzed. In addition, this section also introduces the actual geological conditions of the target coal mine and the introduced CO<sub>2</sub> storage scheme. Finally, the background, motivation and main purpose of this study are explained.

**Chapter 2:** As the CO<sub>2</sub> MNB solution primarily consists of liquid water and gaseous carbon dioxide microbubbles (aligned with a two-phase flow system), its impact on the goaf is largely driven by water-rock interactions. Furthermore, the coal pillar serves as the principal support structure in a room-and-pillar goaf. The mechanical strength and structural integrity of these coal pillars are critical to maintaining long-term stability within the goaf. The periodic recycling of CO<sub>2</sub> MNB solution will also cause the water-rock reaction of coal pillars due to its long-term and periodic immersion, which is also the focus that needs to be analyzed in this study. Therefore, this study focuses on evaluating the stability for coal pillars under long-term, periodic exposure to moisture. The main conclusions are as follows: (1) The water adsorption, uniaxial compressive strength elastic modulus, and stress-strain results indicate that periodic immersion has a more significant impact on the microstructure and macroscopic mechanical properties of coal than long-term immersion. (2) Periodic immersion could induce progressive and irreversible damage to the coal structure. Thus, this study should focus on reducing the risk of instability induced by changes in the water environment that is present in mining engineering applications. (3) In practical engineering contexts, the coal-rock system comprises various interlayered rock (coal) seams, leading to the establishment of a distinct rock-coal structure. The load-bearing framework of the roof-coal pillar exhibits a marked short-slab effect, and the deterioration of the coal pillar compared to the roof or floor presents more considerable risk to the safety of the entire underground CO<sub>2</sub> reservoir.

Through a series of water-coal interaction tests, the mechanical strength and structural integrity of the coal pillars exposed to water for a long time and periodically have been fully analyzed. On this basis, the next chapter focuses on analyzing the effects and differences of pure water and CO<sub>2</sub> MNB solution immersion on the stability of the coal pillars.

**Chapter 3:** To investigate the long-term stability of CO<sub>2</sub> storage through the injection of CO<sub>2</sub> micro-nano bubble solution into coal mine goafs and establish a mechanism for preventing and detecting CO<sub>2</sub> leakage, experimental studies on the deterioration characteristics of coal samples exposed to pure water and CO<sub>2</sub> micro-nano bubble solution for varying soaking periods were conducted. The key findings are summarized as follows: (1) The uniaxial compressive strength and elastic modulus of coal samples decreased as the soaking period increased. During short-term soaking (up to three weeks), the impact of pure water and CO<sub>2</sub> micro-nano bubble solution on coal samples was relatively minor. However, with prolonged soaking (over six weeks), the deterioration of coal samples significantly intensified. Long-term geological storage of CO<sub>2</sub> is shown to cause substantial degradation in the mechanical strength and internal structure of coal seams or pillars. (2) When soaked in a CO<sub>2</sub> MNB solution, the coal samples show varying crack numbers at each loading phase. As the soaking duration extends from zero to three weeks, a corresponding increase in crack development occurs. After six weeks of soaking, the number of cracks within the coal samples rises considerably. The degradation induced by CO<sub>2</sub> micro-nano bubble solution causes the internal structure of the coal sample to become increasingly fragmented, facilitating the formation of seepage channels through which CO<sub>2</sub> can easily leak. Therefore, in actual CO<sub>2</sub> MNB solution storage operations, reinforcement preparation should be limited to three weeks. Prompt or expedited reinforcement helps coal pillars retain high mechanical strength and overburden-bearing capacity.

Considering the degradation effects on coal pillars or coal seams caused by soaking in CO<sub>2</sub> micro-nano bubble solution, further exploration of grouting or backfilling technologies is necessary to study the reinforcement of coal pillars under CO<sub>2</sub> storage effects in following chapters.

**Chapter 4:** Achieving decarbonization targets presents considerable obstacles, particularly given the substantial direct and indirect carbon emissions associated with reinforcement projects. Integrating waste as aggregate offers a promising approach to lessen dependence on newly mined materials in CO<sub>2</sub> storage efforts within coal mine goafs. In this chapter, the backfill reinforcement materials were developed and their mechanical characteristics along with the mechanisms underlying crack development were discussed. The key findings are summarized below: (1) This study provides a comprehensive analysis of the mechanisms for enhancing recycling efficiency and examines the CO<sub>2</sub> storage potential of backfill in goafs under CO<sub>2</sub> MNB immersion. The CaCO<sub>3</sub> content varies with different CO<sub>2</sub> MNB immersion durations. After one day of immersion, the mass loss of CaCO<sub>3</sub> was 0.26%, 0.29%, and 0.11% for backfill specimens with different PGDs (Particle Grade Dimensions). 14 days of immersion, the mass loss increased to 2.03%, 4.02%, and 1.49%, respectively. Long-term CO<sub>2</sub> MNB immersion enhances CO<sub>2</sub> mineralization in the backfill, which can further improve CO<sub>2</sub> uptake efficiency. Immersing the composite backfill in CO<sub>2</sub> MNB enhances its carbon dioxide storage efficiency. (2) After immersing backfill specimens in CO<sub>2</sub> MNB for 14 days, the uniaxial compressive strength of the CO<sub>2</sub> mineralization process enhanced the internal hydration reaction of the backfill specimens, slightly improving their load-bearing capacity. This mineralization also increased the elastic modulus of the specimens. This resistance is crucial for maintaining the long-term performance and durability of the backfill specimens. Additionally, the CO<sub>2</sub> MNB mineralization reduced the compression rate, thereby enhancing the anti-compression performance of the backfill. This improvement ensures that the backfill can maintain a high load-bearing capacity in underground spaces, contributing to the long-term safety of these environments. (3) Considering that concrete weighs approximately 2.4 tons per cubic meter and that cementitious materials constitute 20% to 30% of the total mass, the apparent carbon sequestration for 1 m<sup>3</sup> of backfill with a PGD of 2.2, 2.6, and 2.8 is estimated to be 9.74 to 14.62 kg, 19.30 to 28.94 kg, and 7.15 to 10.73 kg, respectively. Adjusting the PGD of RCA (Recycled Concrete Aggregates) and CO<sub>2</sub> MNB immersion duration can enhance both the recycling rate of waste and the efficiency of CO<sub>2</sub> mineralization and storage.

The designed backfill reinforcement material has good mechanical properties and direct and indirect carbon dioxide emission reduction effects. Backfilling on both sides of the coal pillar can effectively isolate the coal pillar from the CO<sub>2</sub> MNB solution and reduce the deterioration of the coal pillar under the immersion impacts. In actual CO<sub>2</sub> storage projects, the application of this backfill material can be considered to further conduct a detailed analysis of its effect in reinforcing coal pillars in goaf areas.

**Chapter 5:** This chapter concluded with the results of this thesis and provides recommendations for further study.