Electronic supplementary materials

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Effects of ultrasonic treatment on wet mineralization of cement powder

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S1. Wet mineralization and sample preparation methods

S1.1 Ultrasonic-assisted wet mineralization equipment

The ultrasonic-assisted wet mineralization process employs a probe-type ultrasonic transmitter (frequency 20 kHz, model LUIP750, manufactured in China) and with a maximum power output of 750W. A magnetic stirrer (model LC-MSB-HD, manufactured in China) is utilized, featuring a maximum mixing capacity of 5L and a speed range of 100-1600 rpm. The cement mixer is a Hobart mixer.

S1.2 Wet mineralization and samples preparation methods

The production process of ultrasonic-assisted wet mineralization and cement paste preparation is illustrated in Fig.S1. A precise amount of cement powder was weighed into a beaker, followed by the addition of tap water to achieve a water-to-binder ratio of 5:1. The selection of this ratio is based on the results of preliminary research, which can balance mineralization efficiency and avoid excessive temperature rise during the mineralization process. All samples were prepared under identical initial temperature and humidity conditions to ensure experimental consistency. The system remains in an open state. The suspension was thoroughly pre-mixed using a magnetic stirrer (model LC-MSB-HD, manufactured in China) at a speed of 500rpm for 5 minutes. The power was set to 100 w. Excessive power will cause the temperature of the suspension to rise rapidly. Adjust the amplitude to 100%, for which the maximum ultrasonic effect range is achieved at this power level. Continuous ultrasonic mode was selected. A flow rate of 2 L/min was maintained. The gas flow rate was selected according to the value reported in previous research study, which demonstrated high mineralization efficiency. For non-ultrasonic-assisted wet

mineralization, mechanical stirring using a magnetic stirrer replaced the ultrasonic emitter probe. Upon completion of the ultrasonic-assisted wet mineralization process, the beaker containing the suspension was cooled to room temperature using an ice-water bath. This step was implemented to prevent potential flash setting when it was subsequently mixed with the cementitious matrix, owing to its initially elevated temperature. The cooled suspension was then transferred into a Hobart mixer, to which additional cement powder was added, and the water-to-binder ratio was adjusted to 0.4. The mineralized cement powder replaced 8% of the total cement content. The mixture was subsequently cast into molds, demolded and cured at 20 ℃ with 95% RH.

The preparation method of the micro-characterization sample involved fixing wet carbon in a cement suspension, which was subsequently filtered using 3μm filter paper at a controlled rate. The filtered cement particles were then transferred to anhydrous ethanol to stop hydration, followed by low temperature drying.

S2. Energy Efficiency Evaluation of Ultrasonic Action

To quantify the relationship between ultrasonic energy consumption and the net CO_2 capture, an ultrasonic mineralization energy efficiency factor (UMEE) was introduced. The UMEE is defined as the amount of CO_2 sequestered per unit of ultrasonic energy consumed.

$$\mathrm{UMEE} = \frac{m_{CO_2}}{E_{\mathrm{ultrasonic}}}$$

The calculated UMEE values are summarized in Table S2. The UC-2-5 group exhibited the highest UMEE value of 544 kg/(kW ħ), which is ascribed to the rapid consumption of portlandite (CH) generated during pre-mixing within the first five minutes of the ultrasonic-assisted wet mineralization process. In contrast, the UC-2-25 group showed the lowest UMEE, indicating that prolonged mineralization leads to diminished energy efficiency, likely due to the progressively limited reaction kinetics under extended treatment conditions.

S3. Industrial Application Prospects and Technical Challenges

At the practical application level, this study proposes an industrial solution suitable for precast component plants — the Ultrasonic Assisted Mineralization Circulation System. Designed based on the synergistic enhancement of cement-based material strength and carbon sequestration efficiency through ultrasonic wet mineralization technology, the process flow is illustrated in Fig.S5. The system comprises three core units: an ultrasonic mineralization reactor, a temperature control unit, and a concrete mixing unit.

The ultrasonic mineralization reactor is equipped with multiple sets of ultrasonic probes and precision temperature management systems to maintain the reaction system within the optimal temperature range to facilitate efficient mineralization, thereby preventing heat accumulation from adversely affecting the mineralization process. Temperature control units are outfitted with stirring devices and temperature management systems. These units are interconnected via tubing peristaltic pumps, forming a complete circuit for material circulation and temperature control.

CO_2 gas is introduced into the ultrasonic mineralization reactor, where it undergoes

mineralization with cement powder under ultrasonic action. At periodic intervals in the mineralization process, Pump A diverts a portion of the suspension to the temperature control unit, thereby preventing flash setting during subsequent blending with the cementitious matrix due to elevated temperatures. Finally, the suspension is pumped by Pump B to the concrete mixing unit for uniform blending with the cementitious materials and supplementary mixing water. The coordinated operation of the two peristaltic pumps maintains synchronization across all three process stages, thereby enabling continuous functioning of the circulatory system. It should be noted that this integrated process currently exists as a conceptual design and has not yet been physically implemented.

While ultrasonically assisted wet mineralization technology has demonstrated promising carbon sequestration effectiveness and mechanical property enhancement in laboratory settings, its industrial application faces several key challenges: (1) Equipment Durability Issues Caused by Ultrasonic Cavitation Erosion. The ultrasonic cavitation effect, while promoting the reaction, continuously erodes the probe surfaces, leading to gradual material loss and the formation of cavitation pits. This ultimately compromises acoustic performance and shortens the service life. In continuous industrial production, frequent probe replacement would significantly increase equipment maintenance costs. (2) Impact of Alkalinity Reduction in Mineralized Products on Durability. The mineralization process continuously consumes alkaline oxides within the system, resulting in a decreased paste pH. This may weaken the passivating protection of steel reinforcement in concrete, thereby increasing the risk of corrosion. This issue directly affects the long-term durability of structures and represents a critical materials science challenge that must be addressed for successful technology scaling.

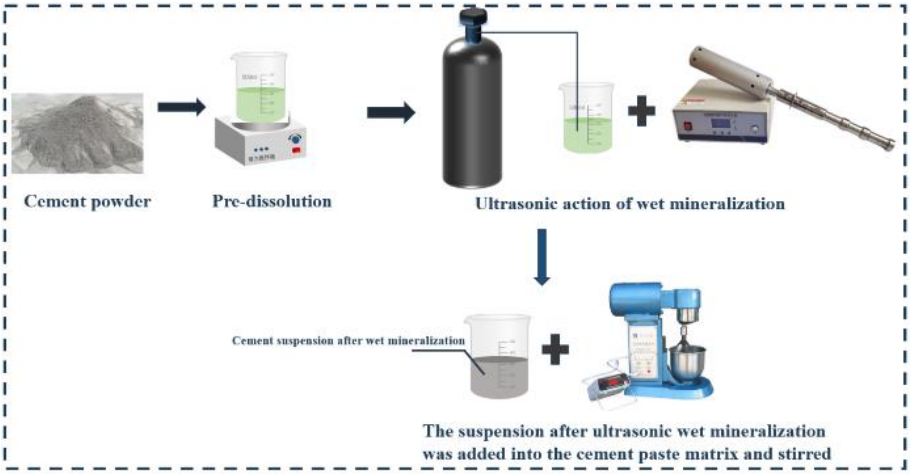

Fig. S1 Ultrasonic-assisted wet mineralization and specimens making process

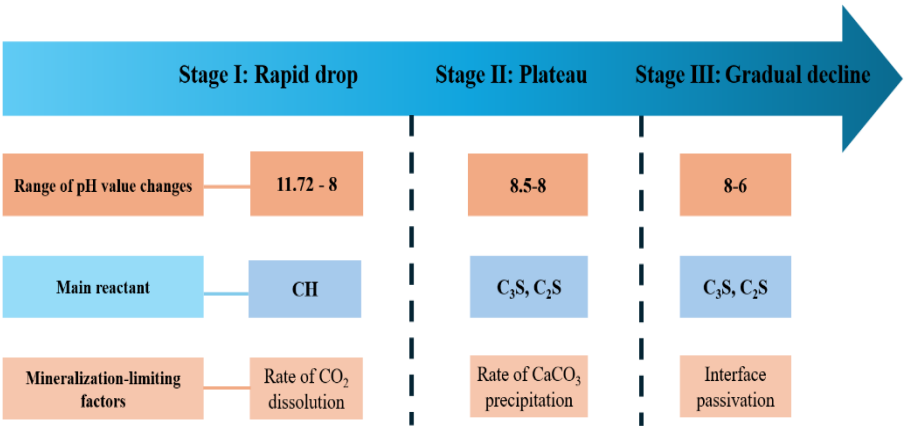

Fig. S2 The pH value variation process of ultrasonic-assisted wet mineralization cement suspension

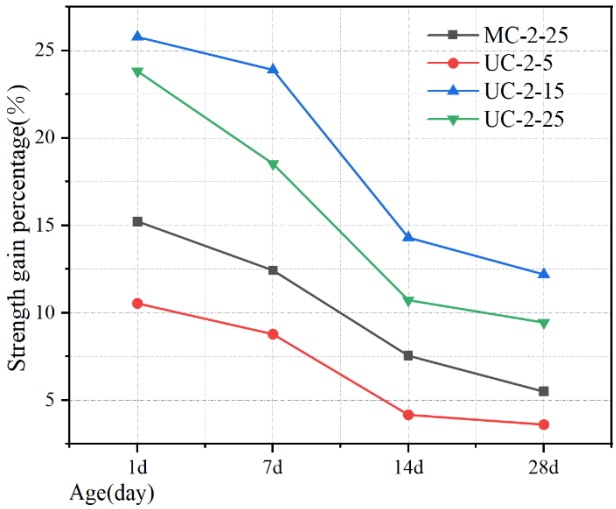

Fig. S3 Comparison of strength gain percentage relative to control

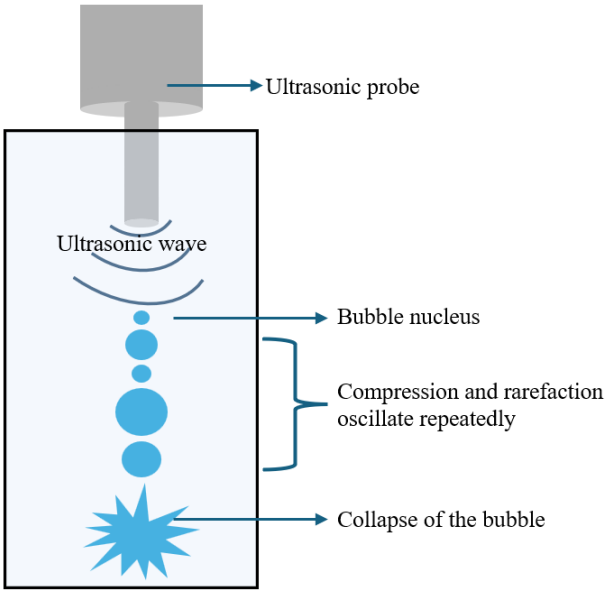

Fig. S4 Formation process of ultrasonic cavitation bubbles

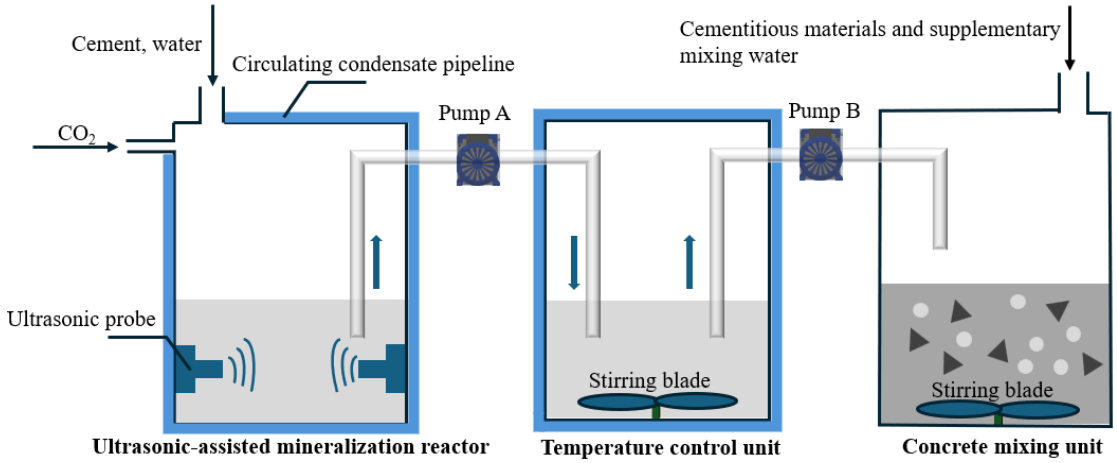

Fig. S5 Schematic diagram of the industrial process of the ultrasonic-assisted mineralization circulation system

Table S1 Comparison of setting time, standard deviation and the rate of change relative to the control

Sample name	Reduction relative to control (%)	Reduction relative to control (%)
N(control)	0	0
MC-2-25	-10.27	-8.25
UC-2-5	-6.48	-5.89
UC-2-15	-17.29	-11.50
UC-2-25	-19.46	-12.98

Table S2 Comparison of UMEE

Sample name	UMEE (kg/kW ħ)
UC-2-5	544.32
UC-2-15	94.80
UC-2-25	61.44