Final Report

STORING CO_2 IN BUILT INFRASTRUCTURE: CO_2 CARBONATION OF PRECAST CONCRETE PRODUCTS

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Abstract

The overall objective of the proposed study was to advance the technical understanding of CO_2 incorporation into novel cementitious materials for the development of high value products that provide a net reduction in carbon emissions. This project combined two primary phases of research that addressed technical barriers related to (i) optimizing CO_2 storage capability of cementitious materials, (ii) evaluating and enhancing physical properties of novel carbonated materials, and (iii) assessing the reductions in life cycle CO_2 emissions attributed to CO_2 carbonation of precast cementitious materials. Engineered cementitious composites (ECC) are a class of highly ductile concrete composites that have been shown to be very durable when used in the built environment. CO_2 carbonation of ECC was examined in this study and it was found that precast ECC specimens could sequester up to 35% CO_2 by cement mass after 24 hours of curing at a CO_2 pressure of 0.5 MPa and 23°C and had a strain capacity of 3%. Carbonation conditions were optimized at the bench-scale and then utilized to create full-scale CO_2-cured ECC railroad ties that were field tested on a train track. Rail ties were selected for this initial assessment of CO_2 storage in precast concrete materials due to the large market for concrete ties in the railroad industry. Although the full-scale rail ties passed all of the required American Railway Engineering and Maintenance-of-Way Association qualifying mechanical tests, on-track testing of the CO_2-cured ECC rail ties was unsuccessful due to fiber alignment in the ECC the during the rail tie casting process which prevented the material from achieving the expected level of strain capacity. This result highlights the challenge in scaling up bench-scale processes to full-scale product manufacturing and requires additional investigation into the casting process of large-scale infrastructure elements using ECC combined with carbonation curing. Life cycle assessment of a CO_2-cured ECC rail tie versus a traditional concrete rail tie indicates that the ECC tie can have lifecycle carbon savings of between 11% and 51% depending on how much longer its useful lifetime is compared to traditional concrete rail ties. Both carbon and cost savings are driven by a reduction in the need to replace broken rail ties, so the key factor is the extent to which a CO_2-cured ECC rail tie will have increased lifetime durability compared to alternative rail ties.

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1. INTRODUCTION

The U.S. National Academy of Engineering (National Academy of Engineering 2008) has recognized the need to mitigate greenhouse gas emissions as one of the grand challenges facing our society during the next century. For over the past two decades, research efforts have explored ways to capture CO_2 from point source emissions sites and sequester it safely, thereby preventing further carbon emissions to the atmosphere. A vast majority of this work as focused on geologic storage of the captured CO_2, however, this type of storage faces challenges regarding leakage risk of stored CO_2 and lacks market incentives for geologic storage outside of using the CO_2 for enhanced oil recovery. CO_2 sequestration efforts will therefore be supported if the stored CO_2 is utilized to develop high value, saleable products. Mineral carbonation of the sequestered CO_2 will further support the successful deployment of carbon mitigation strategies, as it provides a long-term secure storage mechanism for CO_2 emissions in the form of stable carbonate mineral precipitates.

Concrete is the single most utilized construction material in the build environment and cement manufacturing is responsible for approximately 8% of global CO_2 emissions (Ellis et al. 2020). Concrete also has the potential to sequester CO_2 in the form of stable carbonate precipitates, thereby lowering the carbon footprint of the built environment if CO_2 is incorporated into concrete materials. Engineered cementitious composites (ECC) are a novel fiber-reinforced cement that has demonstrated excellent durability in the built environment due to its intrinsically tight crack width and the ability to self-healing via mineral carbonation reaction (Zhang, Yu, et al. 2020). ECC shows extreme tensile ductility (Li, Wang, and Wu 2001; Li 2008), about 500 times that of normal concrete, which makes ECC a potential material choice for carbon storage without danger of embrittlement resulting from the carbonation process without any steel reinforcement. The potential for avoiding having to use steel reinforcement also mitigates the concern for steel corrosion in typical reinforced concrete elements associated with a reduction in pore water pH due to carbonation. The superior mechanical performance and durability of ECC make it a competitive candidate for further investigation in this study.

A goal of this work is to promote the development of concrete materials that have a long service life and directly sequester CO_2 such that they will reduce the life cycle CO_2 emissions of the built environment. Precast concrete elements, such as railroad ties, offer great potential to be a marketable product that is also a sink for coal-fired powerplant CO_2 emissions via mineral carbonation. Successful integration of precast concrete product sequestration of CO_2 emissions into existing power plant infrastructure will require advancements in our understanding of mineral carbonation chemistry, carbonated material properties, and life cycle CO_2 emissions reductions. Market adoption of these new materials will also require a thorough investigation of product durability and price competiveness. This study sought to undertake this challenge in an effort to develop economically viable, secure CO_2 storage technologies. This project evaluated the CO_2 sequestration potential of ECC materials used for the production of railroad ties, as the rail tie market presents a substantial opportunity for growth in the use of carbonated precast concrete materials and will serve as a fertile proving ground for this nascent transformative CO_2 sequestration technology.

2. EXPERIMENTAL METHODS

A brief overview of the experimental methods and testing protocols used in the project are provided in this section. The specific experimental methods are described in detail in the peer-reviewed publications that resulted from this project, with publications clearly noted where applicable. Readers are referred to the published works for additional details.

2.1 Carbonation of Engineered Cementitious Composites (ECC)

Three different bench-scale carbonation reactors were utilized in this study. The first is a chamber that operates under ambient pressure and temperature conditions. This sealed chamber (45.5 × 36.5 × 114 cm) was built with acrylic sheets and contains fans at both the top and bottom to ensure the chamber is well-mixed throughout the carbonation experiments (Neves Junior et al. 2019). A humidifier is used to provide controlled humidity to the chamber. The second reactor is a 58 cm long and 20 cm diameter cylindrical chamber capable of maintaining CO_2 pressures up to 0.9 MPa and is suitable for the carbonation of precast ECC specimens (Wu et al. 2018). The reactor is customized with the capability of monitoring the temperature. Relative humidity can be controlled via a saturated salt solution. The third reactor is a 12 cm height and 8.1 cm diameter 600 ml high-pressure batch reactor (Parr Instruments) (Adeoye et al. 2019). The high-pressure capability of this reactor (up to 48 MPa) and a broad temperature range from 25 to 160ºC allowed for parametric studies on the carbonation reactivity of solid wastes and alternative binders.

A large-scale carbonation chamber was constructed by Shandong Chonhunteda Composite Co., Ltd and is 3 m in length and has a diameter of 1.5 m. This large-scale chamber can hold a pressure up to 1.45 MPa and was used to CO_2-cure the full-size ECC railway ties in this project. The CO_2 gas is continuously replenished to maintain a constant pressure of 0.5 MPa over the carbonation period. A broad range of carbonation conditions were examined (Zhang, Li, and Ellis 2018; Neves Junior et al. 2019; Adeoye et al. 2019; Zhang et al. 2021) and the following 4-step process was found to optimize CO_2 carbonation efficiency of ECC materials. The purpose and operation of each step of carbonation curing is illustrated as follows:

1. Step 1: In-mold conditioning was designed to achieve sufficient early strength for demolding. The trowel-finished surface was exposed to room conditions (temperature of 23±2°C and relative humidity of 55±5%) for 18 hours.
2. Step 2: Specimens were demolded and transferred to the step of de-mold conditioning. The purpose of de-mold conditioning was to create an optimal condition in the ECC specimens for the impending carbonation reaction. A fan drying method was applied to remove excess free water in ECC's microporous space to facilitate the diffusion of CO_2 gas into the ECC specimens. Fan drying for approximately 4-hours was found to be effective in achieving desirable carbonation efficiency.
3. Step 3: Carbonation was performed with pure CO_2 gas at a pressure of 0.5 MPa for 24 hours. This elevated CO_2 pressure environment was designed to achieve a maximum degree of carbonation in the dogbone-shaped specimen.
4. Step 4: Post-carbonation hydration of cement was enabled due to the remaining free water in ECC after early-age carbonation. Specimens were kept at room condition until 28 days.

2.2 Mechanical Testing

Direct uniaxial tension test

In this project, a Material Testing Systems (MTS) loading frame with a 100 kN capacity was used. Uniaxial tension tests were conducted on the dogbone-shaped specimens with displacement control at a rate of 0.5 mm/min following JSCE. The deformation was measured by two linear variable displacement transducers (LVDT) with a gauge length of 80 mm. The geometry of the tension test and sample dimensions are described in Hu et al. (2023).

Compressive strength test

50 × 50 × 50 mm cube specimens were prepared to observe the compressive strength. The compression test was conducted using a Forney loading machine at a loading rate of 0.5 MPa/s following ASTM C109.

Fracture toughness test

Matrix fracture toughness experiments were conducted using a 3-point bending configuration recommended by ASTM E399. Specimen dimensions were 305 × 76 × 38 mm. Beam specimens were pre-notched at the tension surface and the notch depth to beam height ratio was kept 0.4 for all specimens. Carbonation occurs to the greatest extent on surface of a given sample and the lowest in core of the material due to slow diffusion of CO_2 into the cement matrix. To ensure the material adjacent to the notch tip was carbonated effectively, matrix specimens were pre-notched before carbonation. The loading process was displacement-controlled at a rate of 0.5 mm/min.

Static flexural strength test

Four-point flexural static tests were conducted under displacement control to determine the flexural strength of ECC specimens following the ASTM D6272. All specimens had a span length (L) of 254 mm with two-point loads at one half of the span. An Optotrak Certus from NDI was installed to measure the midspan deflection of specimens by captured the displacement of each marker at 40 Hz. Further detail on testing setup and the calculation method for determining midspan deflection can be found in Hu et al. (2023).

Fatigue performance test

Flexural fatigue tests were carried out using sinusoidal cyclic load at 8Hz (Hu et al. 2023). The maximum to minimum fatigue load ratio was 0.1. The fatigue test began with a ramp up force to maximum fatigue load at a rate of 100 N/sec followed by the sinusoidal waveform fatigue loading. The fatigue tests stopped either when the specimen failed or after it reached three million cycles. An Optotrak Certus from NDI was also used to measure the midspan deflection of specimens.

Single fiber pull-out test

A single fiber pull-out test was used to determine the interfacial bonding between fibers and the cementitious matrix. Fibers are glued onto the steel plate and connected to an actuator. Pull-out curves can be obtained after the fibers pull out of the sample. Furthermore, the chemical debonding energy value, G_d, and frictional bond strength, τ_0, can be calculated according to the single fiber pull-out curves (Wu et al. 2018; Zhang et al. 2021).

2.3 Material Characterization

Mineralogical characterization via x-ray diffraction and scanning electron microscopy

X-ray diffraction (XRD) was performed using powder samples on a Rigaku SmartLab XRD with CuKα radiation. The X-rays were generated at 40 mA and 45 kV. The powdered samples were thoroughly mixed with 10% lithium fluoride (LiF) as an internal reference for conducting quantitative XRD. The diffraction pattern was collected in the range of 5°–70° 2θ with a step size of 0.02° 2θ. Phase quantification was carried out using MDI Jade 2010. A JEOL IT500 scanning electron microscope (SEM) was used to examine the surface morphology of samples at 5 kV accelerating voltage. The SEM was also used to image the sample using backscattered electron (BSE) and energy dispersive spectroscopy (EDS). The combination of BSE/EDS imaging coupled with known mineralogy from XRD can be used to track mineral phases at high resolution across the surface of the ECC matrix.

Mass gain method and thermogravimetric analysis (TGA) for CO_2 uptake

For the mass gain method, CO_2 uptake per cement mass is determined following equation where m_2 and m_1 are the sample masses weighed before and after carbonation, respectively:

$$CO_2 \text{ uptake } (\%) = \frac{m_2 + m_{water} - m_1}{m_{cement}} \times 100\%$$

Thermogravimetric analysis (TGA) is also used to determine carbonation efficiency. For TGA, approximately 100 g of the carbonated sample is heated from 20 ˚C to 950 ˚C. The mass change between 20 ˚C 450 ˚C is associated with dehydration of the sample, while the mass change from 550 ˚C and 950 ˚C is assumed to be the mass of CO_2 released from the sample. The amount of CO_2 sequestered in the sample can thus be obtained through use of this method. Further details regarding this method and how it was used to determine CO_2 carbonation efficiency are presented in (Neves Junior et al. 2019).

2.4 Design of Life Cycle and Technoeconomic Analyses

The study by Lim et al. (2023) provides a detailed description of the life cycle assessment (LCA) approach and technoeconomic analysis (TEA) used in this study to determine net carbon emissions reductions and cost estimates associated with switching from the state-of-the-art concrete rail tie with prestressing wires to a CO_2-cured ECC rail tie without steel wire reinforcement. The scope of the LCA and TEA incorporates the lifecycle of rail ties as well as train operation on a track built with either concrete or CO_2-cured ECC rail ties. A functional unit of providing train service on 1-km of track section in the U.S. for 100 years was chosen for the LCA study. Rail tie manufacturing and disposal processes are modeled based on literature review and commercial processes in the U.S. A stochastic use-phase model was developed to evaluate systems-level impacts associated with rail tie failures, replacements and subsequent delays in train travel associated with the stochastic nature of rail tie failures. A total of 2,000 realizations were completed to determine use-phase greenhouse gas emissions and lifecycle costs from tie failures. Results were generated through iteratively running the model while randomizing key parameters related to tie degradation, tie replacement (and method of replacement), and train operation.

The stochastic failure of rail ties was estimated following a Weibull distribution that included parameters for average service life of a tie and temporal distribution of failures. Weibull parameter values are estimated from available historic data on the failure and replacement of about 11 million concrete railroad ties installed in the U.S. The average expected service lifetime for concrete ties was between 40-60 years, while the expected lifetime of ECC ties ranged from 40-100 years. A rail track is installed with new ties, either concrete or CO_2-cured ECC ties, at year 1 and each installed tie independently follows a Weibull distribution. When ties fail in the model they can either (i) be replaced on an individual tie basis (spot replacement), or (ii) be replaced with use of a track laying system (TLS) that replaces all ties within a given section of track regardless of individual tie degradation status, or (iii) cause a slow order when maximum replacement capacities are reached, thereby reducing the train speed within a given section of track. The degree of the train slow order is based on incumbent safety regulations (Cloutier 2014; Federal Railroad Administration 2012) as well as the acceleration profile of trains. Replacing a tie with TLS is cheaper than spot replacement but at a risk of prematurely replacing adjacent ties. The flexibility of the model enables exploring different tie replacement strategies relevant to real world scenarios, such as preemptive replacement of aging ties or prioritizing between spot replacement and TLS.

A detailed cost and process model was constructed based on available industrial data (Grimbergen 2007; Precast/Prestressed Concrete Institute 2021) with a model concrete rail tie plant operating non-stop outputting 250,000 rail ties per year. CO_2-cured ECC ties will require a longer time to complete due to the drying and CO_2-curing stages; as such, the equivalent ECC rail tie facility was assumed to produce 180,000 ties per year. At its end of life, the concrete tie is crushed to be recycled or landfilled. ECC ties are also assumed to be landfilled. Materials, energy, and operations/maintenance costs used in the TEA and LCA are provided in Lim et al. (2023). For the LCA, the Ecoinvent 3 database and Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts (TRACI) methodology were used to evaluate the life cycle emissions and energy use associated with other materials used across the respective rail tie life cycles (Wernet et al. 2016). Mid-point indicators for the LCA include global warming potential (GWP), ozone depletion potential (ODP), acidification potential (AP), and eutrophication potential (EP). A discount rate of 5% is used for all of the economic analyses.

3. RESULTS AND DISCUSSION

The following sections present a summary of the main findings from this study and the subsequent implications for developing CO_2-cured precast ECC infrastructure materials. Peer-reviewed publications submitted by the project team contain detailed presentation of the results and further discussion; each section references the respective publications where the reader can find further information on specific results from this project.

3.1 Carbonation Efficiency

Zhang et al. (2021) reported on how the carbonation curing process described in Section 2.1 was optimized for maximal CO_2 storage through extending carbonation time, elevating CO_2 partial pressure and tailoring binder composition. Further, Zhang et al. (2018) discovered a new parameter that potentially impacts CO_2 storage efficiency as well as governs mechanical integrity of the cured cementitious materials. The new parameter is named as the “pre-hydration period”, which represents the hydration age of Portland cement (PC) when intentionally exposed to high-purity CO_2 carbonation. The experimental results suggest that the CO_2 uptake clearly decreased with a longer pre-hydration period (Zhang, Li, and Ellis 2018). This agrees with the declining trend of the carbonation-induced water evaporation, suggesting the carbonation intensity reduces as the prehydration time is extended. With the assistance of thermal analysis, it was concluded that the calcium carbonate precipitated from CO_2 carbonation could potentially induce nucleation seeding effect by depositing sub-micron calcite solids and therefore accelerate the development of post-carbonation binding property.

Bench-scale carbonation tests were performed using the three different experimental chambers described in Section 2.1 (Adeoye et al. 2019; Neves Junior et al. 2019; Zhang et al. 2021). One chamber allowed for carbonation at ambient pressure and temperature for controlled gas composition and relative humidity; a second chamber allowed for carbonation at ambient temperature and PCO_2 pressures up to 0.9 MPa; a third chamber allowed for carbonation of small sample sizes under elevated temperature and pressure, up to conditions of 48 MPa and 160ºC. Various combinations of temperature and P_{CO2} pressures were evaluated and CO_2 carbonation efficiency was not found to be substantially improved at high CO_2 partial pressures beyond 0.5 MPa or at elevated temperatures beyond 23ºC. As such, P_{CO2} of 0.5 MPa and 23ºC were chosen as the conditions for continued assessment in this project.

Zhang et al. (2021) presented results of carbonation curing of two conventional PC-based ECC versions, i.e., M45 and M45-high volume fly ash (HVFA), were developed and optimized. M45 has a fly ash-to-cement ratio of 1.2, while M45-HVFA has a ratio of 2.2. More detailed mix proportions can be found in Zhang et al. (2021). All mixtures were cast into dogbone-shaped specimens, which were treated with a 4-step of carbonation curing process as described in Section 2.1. The mechanical properties and CO_2 uptake of the CO_2-cured ECC samples were then evaluated.

After a 24-hour period of carbonation, the averaged CO_2 uptake for M45 and M45-HVFA ECC mix proportion attained around 35% CO_2 sequestered by cement mass for dogbone-shaped specimens. It is important to note that this high degree of carbonation efficiency was achieved for thin samples (13 mm). When sample volume is increased, carbonation efficiency decreases due to slow diffusion of CO_2 into the cement matrix.

Based on the mass gain method and TGA results, carbonation-cured ECC can achieve 21.2% and 14.5% CO_2 uptake by cement mass for cube and beam specimens, respectively, after a 24-hour carbonation curing period which can lower the embodied carbon footprint of typical M45 ECC from 600 kg CO_2/m^3 to 472 kg CO_2/m^3 and 513 kg CO_2/m^3. However, this CO_2 uptake is limited to laboratory-scale material. When scaling up to field-scale dimensions, i.e., full-size railway ties, the amount of CO_2 sequestered per cement mass through carbonation curing will significantly decrease due to diffusion limitations of CO_2 into larger volume materials. Considering that the cross-sectional area for full-scale railway ties is twenty times that of the beam specimen, a 10-inch cubic sample was made and tested within the carbonation reactor under varying degrees of drying and times of CO_2 exposure. Under the same conditions for which 14.5% CO_2 uptake was achieved in the beam specimen, the cubic specimen reached a carbonation depth of only 5 mm (6 mm decrease compared to the beam specimen), and the CO_2 uptake also decreased to 3.2 %. It was thus necessary to modify the carbonation process to improve the carbonation depth and CO_2 uptake for full-scale railway ties.

Drying time (i.e., 8 hours and 32 hours) and carbonation time (i.e., 24 hours, 48 hours, and 144 hours) were investigated to find a suitable duration for full-size railway tie CO_2-curing. With 32 hours of drying and 48 hours of carbonation, the 10-inch cubic specimen can achieve approximately 8.4 % CO_2 uptake for the railway ties (i.e., 9 kg CO_2 capture). Phenolphthalein was used to evaluate the depth of carbonation on a cross section of the CO_2-cured cube. The carbonated region was found to be of uniform thickness. XRD analysis confirmed the presence of calcite in the carbonated region (gray color after phenolphthalein exposure) and no calcite was observed via XRD in the uncarbonated region (pink color after phenolphthalein exposure). The results suggest that if the carbonation time is long enough (e.g., 48 hours), the drying time will be the main factor impacting CO_2 uptake for the full-size railway ties. The reason is that the carbonation of the ECC is highly dependent on the mixture's water content. According to T.C. Power's empirical formula for component volume changes during cement hydration, a longer drying time allows more water in capillary pores to evaporate from the mixture. Therefore, there is more contact surface area for the mixture and CO_2 gas to react, resulting in more CO_2 uptake and greater depth of carbonation. Since no significant CO_2 uptake increase was observed when the carbonation time was extended from 48 hours to 144 hours, 32 hours of drying and 48 hours of carbonation time were chosen for carbonation of the full-scale rail ties in this study.

In order to sequester a higher amount of CO_2 into larger volume ECC specimens, ground granulated blast-furnace slag (GGBFS) was selected as a candidate material for pre-carbonation and incorporation into the ECC mixture prior to casting and CO_2-curing. GGBFS contains nearly 50% of CaO and MgO, which is available for CO_2 sequestration before concrete mixing. The GGBFS is first mixed with water and put into a chamber with 0.5 MPa CO_2 pressure for 24 hours. After 24 hours of carbonation, GGBFS is put into an oven to dry before use. According to test results using phenolphthalein indicator, GGBFS shows possible carbonation after curing. XRD analysis was used to verify that the CO_2 was sequestered as $CaCO_3$ and $MgCO_3$ after pre-carbonation treatment of GGBFS. The test results indicate that the carbonation product for pre-carbonated GGBFS can reach as high as 20% weight CO_2 per GGBFS mass; the weight percentage of calcite, nesquehonite, hydromagnesite, and dypingite is 11.2, 6.8, 2.3, and 1 wt%, respectively. This pre-carbonated GGBFS was tested as a substitute for 30% of cement weight in the M45 mix proportion. The CO_2 uptake for this pre-carbonated GGBFS ECC increased from 21.2% to 44.5 % per cement mass, significantly reducing the embodied CO_2 footprint of typical M45 ECC from 600 kg CO_2/m^3 to 209 kg CO_2/m^3. The carbonation depth also increased from 11mm to 20 mm.

3.2 Mechanical Performance of CO_2-cured ECC at the Bench-scale

M45 can achieve a tensile strain capacity of 3% (meeting the criteria of 2% of tensile strain) after carbonation curing at 28 days and a 5 MPa tensile stress. M45-HVFA also achieves a tensile strain capacity of 3%. However, its tensile stress decreased to 3 MPa after carbonation curing. A similar performance drop can be found in the fracture toughness and compressive strength of the M45-HVFA matrix.

The fracture toughness of ECC matrix materials was measured following ASTM E399. Carbonation curing was found to slightly decrease the fracture toughness at 28 days for the M45-HVFA mixture but did not induce significant impacts on the M45 mixture. The fracture toughness of M45 is around 0.426 $MPa \cdot m^{1/2}$ and 0.415 $MPa \cdot m^{1/2}$ before and after carbonation curing. The results of the compressive strength test also show that the 28-day strengths were close between the carbonation-cured (54 MPa) and air-cured M45 specimens (52.6 MPa) but were found comparatively lower for the M45-HVFA specimens subjected to carbonation curing (36.1MPa compared to air-cured 42.7 MPa). The lower 28-day strength of the carbonation-cured M45-HVFA matrix is due to the lessened pozzolanic reaction of fly ash. With a fly ash to cement mass ratio of 1.2, the pozzolanic reaction of fly ash was eliminated after 24-hour CO_2 carbonation. The low portlandite intensity in the carbonated system, even after 28-day subsequent hydration, impeded the fly ash reactions. The low reaction degrees of fly ash led to a relatively coarse microstructure, which could be densified by calcium carbonate precipitation during the CO_2 carbonation. Therefore, carbonation curing led to a similar compressive strength and fracture toughness for M45. As the fly ash content increases and cement content decreases (HVFA ECC), the densification effect by carbonation curing poses a reduced impact with respect to the effect of the fly ash reaction. In this regard, carbonation curing is more favorable for applications on the typical M45 ECC than the high-volume fly ash ECC.

Additional ECC mixtures were tested without clinker and with the addition of MgO. The clinker-free ECC achieved a greater tensile strain capacity of 4.6% for air-cured and 4% for carbonation cured compared to PC-based ECC (M45 and M45-HVFA), however, it only attained a CO_2 uptake of 7% based on the mass of FA-1 fly ash, which is significantly lower than the PC based ECC. Incorporations of MgO into the PC-free ECC mixtures improved CO_2 sequestration potential (Wu et al. 2018; Zhang, Wu, et al. 2020). However, these mixtures were unsuccessful due to the substantial reduction in tensile strain capacity (less than 2%). When GGBFS was incorporated into ECC the tensile performance exceeded 3% of tensile strain and 4 MPa of tensile stress after carbonation-curing, showing comparable mechanical performance to typical M45 ECC.

Additional results regarding the mechanical performance of carbonate ECC are given in Hu et al. (2023) and discussed here. To determine the mechanical behavior, including tensile, flexural, and fatigue performance of ECC after carbonation curing, carbonation-cured ECC samples were fabricated following the carbonation process presented in Zhang et al. (2021) with fan drying after demolding and then cured under 0.5 MPa CO_2 pressure for 24 hours. Flexural performances, including static and fatigue load conditions, are the critical requirements for railway ties. Despite the presence of a body of literature on the flexural performance of water or air-cured ECC, the flexural performance of carbonation-cured ECC has not been studied. It was necessary to conduct experiments to examine carbonation-cured ECC's flexural/fatigue performance before scaling up to the full-size railway ties. A typical M45 PVA ECC mix with a 1.2 fly ash-to-cement ratio was used in this research. A total of sixty 30.5 × 7.6 × 3.8 cm beam specimens were prepared to examine the mechanical performance of carbonation-cured ECC. Five single fiber pull-out test specimens were used to evaluate the fiber bridging and interfacial bond between fibers and the cementitious matrix as affected by the carbonation curing. Finally, three 2.54 × 2.54 × 2.54 cm cube and dogbone-shaped specimens were made to investigate the CO_2 uptake of carbonation-cured ECC by mass gain method and thermal gravimetric analysis (TGA).

The results showed that the first crack strength of carbonation-cured ECC increased by 27% from 5.66 MPa to 7.17 MPa compared to air-cured ECC specimens. Under static loading, the ultimate flexural strength increased by 32% from 10.01 MPa to 13.18 MPa. CO_2 curing also improved the deformation capacity with a 69% midspan deflection increase. During fatigue loading conditions, carbonation-cured ECC also demonstrated more durable and ductile behaviors than air-cured ECC. The fatigue life as a function of flexural stress suggests that carbonation curing had a minimal impact on ECC's fatigue behaviors at a low stress (< 8 MPa) or stress level (< 0.5) but significantly enhanced the fatigue life when imposed by higher flexural stress. The carbonation-cured ECC exhibited a maximum crack opening of 76 μm, whereas non-carbonated

ECC showed a broad range of crack widths up to 148 μm. Also, the average crack width decreased from 62 μm to 30 μm after carbonation curing.

The results indicate that carbonation curing strengthened ECC's crack width control capability under fatigue conditions. The failure type for both carbonated and air-cured ECC fibers was also examined. Compared to air-cured ECC, PVA fibers for carbonation-cured ECC tend to have more rupture failure which is attributed to the increased interfacial bond between PVA fibers and carbonated ECC matrix after carbonation curing based on the single fiber test results. The average values of chemical debonding energy G_d and frictional bond strength τ_0 increased from 0.37 J/m^2 to 2.42 J/m^2 and from 1.32 MPa to 2.24 MPa, respectively, indicating a stronger resistance to fiber debonding and slippage.

3.3 Mechanical Performance of Carbonated ECC Railroad Ties

Considering CO_2 sequestration efficiency and mechanical performance, the M45 mix proportion following the optimized carbonation curing process was chosen as the optimal composition for this project. This project selected railway ties as a test case product for storing CO_2. The rail industry has been transitioning from wood ties to concrete ties over the last thirty years. This transition is driven in large part by the increasing difficulty in acquiring quality wood and the promise of a significant increase in service life by concrete ties of up to fifty years. Unfortunately, the experience so far has been uneven. Some concrete ties fail within the first few years due to cracking. The solution so far has been the use of a large amount of steel prestressing, which introduces another problem given the tendency for bond failure under dynamic rail loading. A CO_2-sequestrated and steel prestressing-free railway tie that can meet all the American Railroad Engineering and Maintenance-of-Way Association (AREMA) standards may be one of the solutions to achieve a more durable concrete rail tie (AREMA 2019). Therefore, the carbonation-cured M45 ECC mix proportion was chosen to build the real-size ECC railway ties in this project.

The dimension of the full-size rail tie for this research is 7'9" × 12" × 10", which meets the American Railway Engineering and Maintenance-of-Way Association (AREMA) standard for concrete railway ties. The relevant equipment necessary for full-size rail tie construction includes (but is not limited to) a rail fastener system, cast-in shoulders, concrete/ECC molds, and vibration tools. Based on the literature review and iterations of communication with industrial specialists, the E-Clip fastener (donated by Pandrol) was selected for this project. This type of fastener system has been widely adopted in North America and can be easily installed and removed without special tools. Helser Industry built a custom mold due to the unique design of our railway ties (no prestressed wires). The rail that will be used for the qualification lab tests before in-track testing of the carbonated ties is AREMA 141 RE rail.

Sixteen carbonation-cured ECC railway ties were prepared. Before Transportation Technology Center, Inc. (MxV Rai, TTCI) provided the single tie push test (STPT), the following prerequisite laboratory tests were conducted according to AREMA standards to a preliminary assess the railway tie's performance before installation in track (AREMA 2019). Three of the sixteen full-scale ECC railway ties were prepared and examined following the AREMA standards:

1. Test 1A: Bending-Rail seat Positive
2. Test 1C: Bending-Center Negative

3. Test 5A: Fastener Uplift Test
4. Test 6: Tie and Fastener System Wear/Deterioration Test
5. Test 7: Fastener Electrical Impedance Test

According to AREMA standards, the ECC railway ties passed tests 1A and 1C since no crack was observed after MTS held the required load P for 3 minutes. For test 5A, ECC railway ties also passed the standard that no inserts were pulled out or loosened in the tie, and no components of the fastening system were fractured. Finally, for test 7, the railway ties will meet the requirements if a minimum impedance value of 20,000 ohms has been measured. The impedance value for carbonation-cured ECC railway ties exceeded 200,000 ohms. The main reason for this is that there are no prestressed wires in our railway ties. Carbonation-cured ECC also passed Test 6. Only 1 marker reached 0.52 cm at 2.7 million cycles which is acceptable for the wear/deterioration test, showing promising wear resistance. All laboratory test results were reviewed and accepted by MxV Rail engineers.

After passing all the prerequisite laboratory tests, the remaining 13 carbonation-cured ECC railway ties were sent to TTCI to conduct STPT on the High Tonnage Loop (HTL) at the Facility for Accelerated Service Testing (FAST) located in Pueblo, Colorado and included the following activities:

1. A test section with 13 concrete ties was established on the HTL at FAST.
2. Single tie push testing at 0 million gross tons (MGT) was conducted to characterize the track lateral stability of the concrete ties.
3. Inspections during and after the train operation were performed to document the conditions of the ECC tie section.

After the ties arrived, they were unloaded and visually inspected. The visual inspection showed the ties were intact and in good physical condition. A hydraulic tie inserter was used to install the ties in the track. The claw of the tie inserter caused the tie surface indentation/damage. Therefore, the installation method was changed to push the tie ends laterally using a speed swing to avoid tie damage. One tie experienced breakage during the speed swing push. Based on the observations made during the installation, to ensure the ties' in-track performance, one tie was selected to conduct AREMA bending tests 1A and 1C at the MxV Rail's Component Testing Laboratory. Both tests met the AREMA test recommendations and the tie section remained installed in the track for the duration of the testing.

After the installation of the 13 ties in the test zone, STP testing in accordance with the AREMA Manual for Railway Engineering, Chapter 30, Part 2, Test 8 – Single Tie Lateral Push (STP) Test1 was performed at 0 MGT (AREMA 2019). In the test, the force required to push the tie laterally through the ballast section was measured as a function of the tie's lateral displacement. The hydraulic piston was attached to the tie that was then pushed against the rail. The STP tests were conducted for 0 MGT only. Every other tie in the 13-tie section was pushed for a total of six pushed ties. During tie pushing, the push forces and tie lateral displacement for each of the six ties was recorded. In general, the peak forces between 0 and 0.64 cm of the tie lateral displacement are tracked and evaluated at FAST. The average value of the peak forces was 2.76 kips, an amount above the required tie lateral resistance of 2 kips for FAST.

After verifying that the carbonated ECC railway ties passed the STP standard, the train operation began. The track condition, including tie surface condition and track gauge, was monitored, especially during the first 40 train laps. Several observations were made during this early inspection: hairline cracks were detected near the rail seat and center of ties and the cracks grew broader as tonnage accumulated. One tie pad and one insulator were broken in the first 10 laps of train travel. Track gauge was monitored during the entire operation. The track gauge was maintained within 56.5–57.0 inches in the test section. Thirteen concrete ties were able to hold the track for the first night. After the first night of operation, however, 12 of the 13 ties had cracks, 8 of the 12 were fully cracked, and 4 of the 12 were half-cracked. Due to safety concerns related to the presence of these cracks, the entire test section had to be removed from the track and the STP test at 10 MGT was canceled. Upon its removal, the tie section had accumulated 2.5 MGT.

The carbonated ECC railway ties met the test requirements of the AREMA laboratory design tests for prestressed concrete ties. Based on the test results, the AREMA tests for prestressed concrete ties are not indicative of the in-track performance of ECC railway ties. Therefore, post-test observations were made to clarify the problems with the carbonation-cured ECC railway ties. The cross-sectional area of the critical crack showed an unexpected non-uniform fiber dispersion where the fibers were aligned in the same direction as the casting; this limited the performance of the full-scale railway ties and prevented the ECC from being able to control crack width as it is designed to do. When scaled up to the larger structural elements, this remains a technical challenge that may need to be solved in the future. Also, when scaling up to field-scale dimensions, the amount of CO_2 sequestrated per cement mass will likely be lower. Though we can achieve 10 % CO_2 uptake for full-size railway ties (i.e., 12 kg of CO_2 capture) by adjusting the carbonation process, there is still a need to find a way to enhance the carbon sequestration to further decrease the CO_2 footprint for larger ECC precast elements. One solution to achieving greater CO_2 sequestration capacity in the carbonated rail ties would be to directly incorporate pre-carbonated aggregate materials into ECC such as GGBFS (see section 3.1). Since CO_2 sequestration capacity can be limited by slow diffusion into the material, this strategy would allow for the storage of CO_2 via mineral carbonation of the aggregate further into large-volume specimens.

3.4 Life Cycle Assessment and Technoeconomic Analyses

Manufacturing cost of a traditional prestressed steal wire concrete tie is estimated to \$41/tie. This estimate includes cost of raw materials, wages, and manufacturing overhead, with the primary factor being cost of raw materials (75% total cost). The manufacturing cost of a CO_2-cured ECC rail tie is estimated to be \$67/tie and is higher due to the greater amount of PC used in ECC and the addition of PVA fibers. Assuming a cost of CO_2 of \$440/tCO_2 based on cost of recovery and transport from industrial processes, 11% of the total cost of the CO_2-cured ECC tie is due to purchasing CO_2. Estimated cost uncertainty is within 30% and is due mostly to material cost uncertainties (Lim, Ellis, and Skerlos 2023).

The environmental impact of both the concrete tie and the ECC tie is due primarily to the amount of PC used in the production of each tie. Across all four mid-point impact categories (GWP, ODP, AP, EP), the environmental impact increased with ECC by 17-68% due to the higher amount of PC used in manufacturing of the ECC ties. If CO_2 were captured directly from the air versus from the flue gas emissions of a coal-fired powerplant, only 11.4 kgCO_2eq can be offset by incorporation of 12 kg of air-sourced CO_2 into the ECC tie; this results in CO_2-cured

ECC ties using air-sourced CO_2 still having 38% greater GWP than a traditional concrete tie. concrete tie when use-phase benefits are not accounted.

Switching to CO_2-cured ECC ties is estimated to require 45% fewer ties over a 100-year time horizon based on the median values of the 2,000 iterations for tie failure (see Section 2.4). Use of fewer ties over this 100-year period would result in an 11% reduction in GWP for ECC ties compared to concrete ties. These results support prior work that demonstrated the majority of CO_2 savings (or emissions reduction) associated with CO_2-curing of concrete materials is due to changes in the physical properties of the material and not the amount of CO_2 actually stored (Lim, Ellis, and Skerlos 2019). Although the cost per tie for ECC is 63% higher than that of a concrete tie, the total cost of building and maintenance on a 100 km of track made with ECC ties is 12% less than that when using concrete ties over 100 years. Reductions in cost and CO_2 emissions are driven by enhanced durability and longer service life of ECC ties, which require fewer ties over the lifetime of the track operation.

Given that tie longevity is the key parameter driving potential benefits of using ECC for rail tie construction, a sensitivity analysis was conducted to test uncertainties around the assumption that ECC ties will be more durable in the field. Additional sets of results (1,000 iterations per analysis) were conducted where the average lifespan of ECC ties was continually decreased from the initially assume 60-year average service life to the point where the lifecycle cost and GWP of CO_2-cured ECC ties becomes higher than that of the concrete ties. It was found that the ECC ties need to last 25% longer than the concrete ties (60 years average lifespan for ECC ties vs 40 years average lifespan for concrete ties) in order to achieve a net reduction in CO_2eq emissions. If the CO_2-cured ECC ties are able to last an average of 100 years while in service on the track, there could be a reduction in GWP of 51% and cost savings of 24% over 100 years of service if ECC ties were used instead of traditional concrete ties (Lim, Ellis, and Skerlos 2023).

Market assessment

North American railroads purchase about 24 million new rail ties each year mainly to replace degraded ties on the track. According to a recent survey that collected purchase information of 15 million ties in 2017, about 95% of the purchased ties were wood ties. Prestressed concrete ties represented only 4.7% of the total purchase. Applying this to 24 million total new tie purchases yields 1.1 million concrete tie sales. Assuming a \$100 purchase price of a concrete tie, the annual concrete tie market in the U.S. can be approximated as 110 million USD/year. On the other hand, Amtrak has recently announced its plan to replace all wood ties in its busiest northeast corridor with concrete ties as a part of its infrastructure upgrade efforts. Amtrak currently has approximately 2.9 million concrete ties installed along its 2,364 track miles (Amtrak 2020). Assuming 2,640 concrete ties are needed to sustain a mile of track, Amtrak is expected to purchase 3.3 million new concrete ties to replace its current wood ties in the coming decades. This new demand from Amtrak is driven by multiple factors including rising cost of wood ties and superior quality of concrete ties. If ECC ties can be proven in the field for its enhanced durability and become available for mass productions, ECC ties can potentially replace not only concrete but also wood ties. If all 24 million ties annually being purchased in North American market can be replaced with ECC ties, the total market size would be about 1.67 billion USD/year assuming 126 \$/tie for an ECC tie and accounting for reduced tie needs due to its superior durability.

According to the TEA/LCA model used developed in this project, designing a track with CO_2-cured ECC ties would require 45% less ties over 100 years compared to concrete ties due to their enhanced lifespan. If ECC ties were to be mass-produced to replace new concrete tie demand for NEC, about 1.8 million ECC ties would be needed. Manufacturing this amount of ECC ties would permanently sequester 22,000 tonnes of CO_2 through carbonation curing. Taking 126 $/tie as a manufacturing cost of an ECC tie, replacing wood ties in the North East Corridor with ECC ties would cost 2.8 million USD, which is 16.5% lower than the expected cost with concrete ties. If ECC ties can be scaled to target the entire North American tie market, nearly 288,000 tonnes of CO_2 would be sequestered.

Key performance metrics

CO_2 utilization intensity = $\frac{12\ kg\ CO2\ utilized}{379.6\ kg\ ECC\ tie} * 100 = 3.16\%$

CO_2 integration reaction rate = $\frac{180\ ties * \frac{12\ kgCO2}{tie} * \frac{1000\ mol}{44\ kgCO2} * \frac{1\ lb-mol}{453.59\ mol} * \frac{1}{48\ hr}}{70.7\ m3 * \frac{264.172\ gal}{1\ m3}} = 1.21 * 10^{-4}\ \frac{lb-mol/hr}{gal}$

- CO_2 integration reaction rate is based on CO_2 curing of 180 ECC ties over 48 hours inside a single carbonation chamber whose interior volume is 70.7 m^3.

CO_2 energy utilization = $\frac{22\ kW}{80\ ties * \frac{0.012\ tCO2}{tie} * \frac{1}{48\ hr}} = 1100 \frac{kW}{tonnes/hour\ CO2\ utilized}$

- CO_2 energy utilization only accounts for energy required to recover residual CO_2 inside the carbonation chamber upon completion of curing using a vacuum pump. This value represents maximum energy requirement from vacuum pump operation.

Product marketability = $\frac{100\frac{\$}{tie}*(1-0.1)+(67-41)\frac{\$}{tie}}{100\frac{\$}{tie}+(67-41)\frac{\$}{tie}} * 100 = 92\%$

- The manufacturing cost and market value of a conventional tie are estimated as 90 $/tie and 100 $/tie assuming 10% profit margin ("Benchmarking Report: Precast Industry Grew by 5% in 2016 - NPCA" n.d.). Note that a 10% profit margin is chosen as a point of reference and may not represent the margin of the actual ECC plant.
- The manufacturing cost and market value of an ECC tie are estimated by adding manufacturing cost difference of an ECC tie relative to a concrete tie to the manufacturing and market value of a concrete tie, respectively.

Incremental cost reduction = 41 – 67 $/tie = -26 $/tie

- Incremental cost reduction is based on manufacturing a single tie only, not accounting for its use-phase cost reduction opportunities highlighted in the main text of Lim et al. (2023). Negative value indicates that CO_2-cured ECC tie costs more to manufacture compared to a concrete tie.

CO_2 emissions reduction is +60% during manufacturing, meaning 60% more CO_2 is emitted when manufacturing an ECC tie compared to a concrete tie; however, CO_2 emissions reduction is -12% over the lifecycle of ties due to enhanced lifespan of an ECC tie.

Technology gap analysis

The process of creating a CO_2-cured concrete rail tie does not currently exist in the market and was tested in this study for the first time. ECC was used as the material of the rail tie and its unique strain-hardening properties and ability to control crack width allowed for the creation of a concrete rail tie without the integration of pre-stressing steel wires. As noted in the results of the LCA and TEA (Lim, Ellis, and Skerlos 2023), the carbonated ECC tie has environmental benefits over traditional steel-reinforced ties if it is able to have a useful median lifetime of 40% longer than that of a traditional concrete tie. Bench-scale results for the carbonated ECC showed a high degree of durability with strain capacity of 3%, which is orders of magnitude higher than traditional concrete, suggesting the ECC ties may outperform concrete ties in the field from the standpoint of durability. However, on-track testing of the carbonated ECC ties demonstrated that the ties were unable to perform as expected and thus additional experimentation is needed to develop a casting procedure that prevents fiber alignment during pouring of the ECC. This is not an insurmountable challenge, as there are large-scale demonstrations where ECC has been used in buildings and in roadways, however, the lack of commercial-scale mixing equipment hindered the ability to achieve a randomized homogeneous distribution of the fibers within the ECC matrix of the precast rail ties in this study. Further, full-scale testing of CO_2-cured precast materials that incorporate pre-carbonated aggregate materials is needed to achieve a higher amount of sequestered CO_2 in large volume infrastructure elements.

There is also a remaining challenge with coupling CO_2-curing of precast concrete elements with flue gas gathered directly from a coal-fired powerplant. Pure CO_2 was used in this study and it was found that carbonation was enhanced up to a CO_2 partial pressure of 0.5 MPa. Lower P_{CO2} would slow the rate of CO_2 ingress into the cement matrix and result in either a lower total amount of sequestered CO_2 or require longer exposure (curing) times for the precast materials. In this study, interaction between ECC and gas mixtures that may be more representative of untreated flue gas streams were not tested in the carbonation experiments. The presence of gases such as SO_2 may lead to lower cement porewater pH values and increased concentrations of SO_4^{2-} which could simultaneously inhibit carbonate precipitation (due to lower pH) and potentially increase formation of deleterious products such as ettringite (Zhang, Li, and Ellis 2019). These challenges will need to be addressed in further research.

4. Conclusions

This project sought to address technical barriers related to (*i*) optimizing CO_2 carbonation of cementitious materials, (*ii*) evaluating physical properties of novel carbonated materials, (*iii*) assessing the reductions in life cycle CO_2 emissions attributed to CO_2 carbonation of precast cementitious materials. The research approach combined a series of experimental investigations that examined and sought to optimize the extent of CO_2 mineral carbonation of ECC materials. Incorporation of other industrial solid waste products, including fly ash and ground-granulated blast furnace slag, into carbonated ECC mixtures was evaluated in an effort to further reduce the embodied life cycle CO_2 emissions of the precast concrete end products. An extensive suite of characterization techniques was employed to determine if carbonated ECC products will meet or exceed required industry standards. Based on bench-scale experimental results, an ECC mixture and optimized carbonation procedure were used to cast full-scale rail road ties that were lab-tested to meet American Railway Engineering and Maintenance-of-Way Association standards and tested on an actual train track to evaluate performance field performance. At the bench-scale, CO_2 sequestration in ECC could achieve 35% CO_2 stored per cement mass, while in larger volume precast products only 8.4% CO_2 sequestered per cement mass. This is due to diffusion limitations of CO_2 into the cement matrix and densification of the matrix associated with precipitation of $CaCO_3$. The CO_2-cured ECC retained the durability characteristics of air-cured ECC with a strain capacity of 3% and ultimate tensile strength of 5 MPa. It also reached a compressive strength of 54 MPa.

Net CO_2 emission reductions attributable to CO_2 storage in the novel ECC precast rail ties were evaluated through completion of a full life cycle analysis. Depending on the lifespan of the CO_2-cured ECC ties, there is a potential for net CO_2eq emissions of between 11% and 51% over a 100-year time period. Cost reductions are estimated to be on the order of 12% to 24% if ECC ties are used in place of traditional concrete ties. Tie durability is the key parameter driving CO_2 and cost reductions; the ECC ties would have to have an average service life that is a minimum of 25% longer than the concrete ties in order to achieve net CO_2 reductions and cost savings over a 100-year period. The CO_2-cured ECC ties produced and tested on-track as part of this study lacked the expected durability due to the occurrence of fiber alignment during tie casting. These ties were not able to represent the performance characteristics of ECC materials and highlight challenges with scaling production from bench-scale to full-scale infrastructure elements. Future efforts will need to overcome this challenge in order to test the true viability of this novel cementitious material for rail tie manufacturing.

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