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# Mineral Carbonation of Mixed Industrial Residues for Enhanced CO<sub>2</sub> Sequestration and Environmental Stabilization: The Role of Slag Composition and Industrial Symbiosis

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## ABSTRACT

The increase in greenhouse gases (GHGs), particularly carbon dioxide (CO<sub>2</sub>), represents one of the major environmental challenges of recent decades. Carbon capture, utilization, and storage technologies are essential for deep decarbonization, especially in hard-to-abate sectors. Among these, CO<sub>2</sub> mineralization using alkaline industrial wastes is a promising approach, enabling carbon sequestration and waste valorization within a circular economy framework. In this study, the efficiency of mineral carbonation is evaluated as an integrated strategy for permanent CO<sub>2</sub> sequestration and stabilization of hazardous components in combustion residues. Fly ash (FA), bottom ash (BA), and slag are investigated due to their strong reactivity toward carbonation. The process promotes the formation of stable carbonate phases, ensuring long-term carbon storage under ambient conditions. Additionally, carbonation enhances the immobilization of heavy metals like Pb and Zn, while contributing to chlorine stabilization through the formation of Friedel's salt. The influence of industrial wastes and by-products on CO<sub>2</sub> capture capacity is systematically analyzed, emphasizing the role of BA in improving stabilization mechanisms. Additionally, carbonated FA was valorized as a filler in high-density polyethylene-based composites. The results demonstrate that mineral carbonation represents an effective and sustainable solution for simultaneous CO<sub>2</sub> sequestration and waste management, contributing to environmental protection and resource efficiency.

## 1 | Introduction

One of the major global challenges of recent decades is the increase in greenhouse gas (GHG) emissions. Carbon dioxide (CO<sub>2</sub>) accounts for approximately 72% of total emissions [1] and reached an atmospheric concentration of 426.48 ppm in November 2025 [2]. This results in an approximately 0.8 °C increase in the global mean temperature, intensifying climate change [3]. In the broader context of sustainability, innovative

materials play a key role in achieving the Sustainable Development Goals (SDGs), which require a multidisciplinary approach due to their interconnected nature. Recent studies highlight the importance of materials science in supporting environmental solutions [4]. A long-term solution to address CO<sub>2</sub> emissions is represented by carbon capture, utilization, and storage (CCUS). The International Energy Agency (IEA) highlights that CCUS is critical for deep decarbonization, especially in hard-to-abate sectors like cement, steel, and chemicals. While costs

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vary widely, the technology is already competitive in some industries. Transport and storage costs are often low, and overall CCUS costs are expected to decline with scale, experience, and stronger policy support. This technology can potentially prevent the emission of eight billion tonnes of CO<sub>2</sub> by 2050, which is around one-quarter of today's annual energy-sector emissions [5].

A promising approach for the development of CCUS technologies is represented by CO<sub>2</sub> mineralization and utilization using alkaline solid wastes, which simultaneously contribute to mitigating global warming and solid waste stabilization. An effective option for CO<sub>2</sub> reduction involves its sequestration through adsorption onto suitable materials, a process that can be carried out via the carbonation of ashes. Thanks to their high content of alkali and alkaline earth metals, these materials are well suited for CO<sub>2</sub> capture. Mineral carbonation of CO<sub>2</sub> mimics the natural chemical weathering of rocks, in which dissolved CO<sub>2</sub> forms carbonic acid that reacts with alkaline minerals to produce stable carbonate phases [6–8].

The European Commission has established the criteria and requirements for GHGs to be considered stably chemically bound in a product. To provide a climate benefit, CO<sub>2</sub> must remain permanently incorporated into the atmosphere for at least several centuries. The permanence of the carbon stored in a product depends on the chemical bond, product use, and end-of-life treatment, as carbon may be released through combustion or incineration. Mineral carbonates, such as calcium carbonate (CaCO<sub>3</sub>) and magnesium carbonate (MgCO<sub>3</sub>), form strong and stable chemical bonds so that the carbon can be considered permanently chemically bound unless exposed to extreme conditions like elevated temperatures or strong acids.

Mineral carbonation of alkaline residues can be performed through different process routes. Direct aqueous carbonation involves the reaction between CO<sub>2</sub> and alkaline solids suspended in water, where the carbonation efficiency is controlled by the dissolution of Ca- and Mg-bearing phases and the subsequent precipitation of carbonate minerals [9]. Unlike extraction-based carbonation processes, direct aqueous carbonation does not require pH adjustment or controlled Ca<sup>2+</sup> and Mg<sup>2+</sup> concentrations since carbonation proceeds using the intrinsic alkalinity of the investigated residues under fixed operating conditions. More recently, pH swing processes and calcination–dissolution techniques have been proposed to enhance carbonation kinetics by separating the dissolution and precipitation steps. In these approaches, acidic or alkaline extracting media are employed to selectively dissolve Ca<sup>2+</sup> and Mg<sup>2+</sup> ions, while subsequent pH adjustment promotes carbonate precipitation. The effectiveness of these processes strongly depends on solution pH, Ca and Mg concentrations, and the balance between dissolution and precipitation kinetics [10].

Although pH adjustment and extraction-based approaches can significantly increase carbonation efficiency by enhancing Ca<sup>2+</sup> and Mg<sup>2+</sup> availability, they generally require additional chemical reagents, multiple processing steps, or thermal treatments, which increase process complexity and energy demand. In contrast, the present work focuses on the direct carbonation of mixed industrial residues under fixed operating conditions,

exploiting the intrinsic alkalinity of the investigated residues without additional pH control. The objective is to evaluate whether the strategic combination of complementary industrial wastes can improve the carbonation performance and contaminant stabilization within an industrial symbiosis framework.

Therefore, mineral carbonation enables long-term carbon retention under normal environmental conditions in accordance with the current European regulatory framework [11]. However, although the present work demonstrates the formation of stable carbonate phases, the product use phase and end-of-life scenarios were not investigated and therefore remain beyond the scope of this study and the subject of future work.

Recent studies conducted on ashes demonstrated their reactivity with CO<sub>2</sub>, highlighting their use as a sequestering agent [12]. In particular, fly ash (FA) can react with CO<sub>2</sub> in aqueous environments, leading to the precipitation of carbonates with a significant uptake of CO<sub>2</sub>, thereby emphasizing the high reactivity of these materials toward carbonation [1, 8, 13–15].

Furthermore, since FA originates from processes that generate large amounts of CO<sub>2</sub>, using the solid waste as a raw material for the capture and storage of carbon dioxide can be considered a sustainable method for limiting the emission of this gas [8]. It is known that ash exhibits a highly variable composition and, consequently, diverse physicochemical properties; for this reason, carbonated ash can be directed toward different industrial applications, including the production of construction materials, soil amendments, and fertilizers, as well as the recovery of valuable components and elements [1]. The ash from the incineration of municipal solid waste is composed of bottom ash (BA) and FA. BA represents 30% of the total and is considered a by-product. It does not pose significant environmental risks and can be recycled as a source of metals or as a substitute for quartz sand in the production of cement. On the contrary, FA accounts for between 1% and 3%, and it is a volatile residue rich in heavy metals, chlorine, and salts; therefore, it is considered a hazardous product. Hence, FA must be treated before reuse or disposal in landfills. As previously said, FA contain heavy metals, as, Zn, Pb, Ni, Cu, As, Hg, Cr, and Cd, that can be volatilized during combustion, consequently exposed to atmospheric agents, and transported by wind or leached by rain, potentially spreading into the surrounding environment [7, 8, 16, 17]. Recent studies have demonstrated that carbonation promotes the stabilization of heavy metals in FA, particularly lead and zinc. The incorporation of BA further enhances their immobilization through carbonation and pozzolanic reactions [18–25]. In addition to heavy metals, FA typically contains significant amounts of chlorine, which represents a further environmental concern. Carbonation processes can contribute to chlorine immobilization through the formation of hydrocalumite ([Ca<sub>2</sub>Al(OH)<sub>6</sub>]Cl·2H<sub>2</sub>O), commonly referred to as Friedel's salt. In the presence of aluminum, chlorides can react with calcium to form stable chloroaluminate phases that structurally incorporate Cl<sup>−</sup> ions, thereby reducing the availability and mobility of chlorine within the material [22–25].

Within this context, the concept of industrial symbiosis plays a key role as it enables the combination of residues from different industrial processes to optimize their chemical reactivity and

functional performance [3]. The combined use of different industrial by-products, such as FA, BA, and steel slags, allows the optimization of chemical composition and reactivity, enhancing both CO<sub>2</sub> capture efficiency and environmental performance. These materials, originating from different industrial processes, can be strategically combined to exploit complementary properties, such as the high calcium content of slags, the presence of reactive phases in FA, and the silica and alumina contribution of BA.

This approach enables the design of tailored mixtures in which carbonation reactions and stabilization mechanisms occur simultaneously, maximizing both CO<sub>2</sub> sequestration efficiency and environmental performance, yielding materials suitable for further valorization [1].

The present study aims to evaluate the efficiency of mineral carbonation as an integrated strategy for CO<sub>2</sub> sequestration and environmental stabilization, with particular emphasis on the role of mixed industrial residues.

Unlike most previous studies, which optimize carbonation through chemical extraction and/or pH control to increase Ca<sup>2+</sup> and Mg<sup>2+</sup> availability, the novelty of the present work lies in designing tailored mixtures of industrial residues while maintaining identical carbonation conditions, as shown in Table 1. This approach enables the influence of the waste composition and mineralogical reactivity to be isolated without introducing additional process variables. Although more energy-efficient extraction routes are currently being investigated, they still require additional processing stages.

Specifically, the influence of slag composition on CO<sub>2</sub> uptake is investigated, together with the contribution of BA to the immobilization of heavy metals and chlorine. Moreover, this work explores the potential use of carbonated industrial residues as a reinforcing filler for thermoplastic composites. Particularly, high-density polyethylene (HDPE)-based composites were produced via melt compounding and characterized to understand how filler content and particle size affect their mechanical performance. This approach provides new insights into the potential of industrial symbiosis for the development of sustainable materials based on mineral carbonation, while future life cycle assessments will be required to evaluate their overall carbon footprint. Therefore, the proposed strategy should be regarded as a waste valorization process assisted by mineral carbonation rather than as a process specifically designed to maximize Ca and Mg extraction efficiency.

## 2 | Materials and Methods

### 2.1 | Starting Materials

This work considered the following industrial secondary products: FA and BA, provided by the waste-to-energy plant in Brescia (A2A), and Desulfurized Fly Ash (FDG), obtained from the Lamarmora thermal power plant in Brescia (A2A). FA is a dangerous volatile by-product rich in heavy metals, whereas BA is not classified as environmentally hazardous. The steel slags investigated were Blast Furnace (BF), Ladle Furnace (LF), Argon Oxygen Decarburization (AOD), and Electric Arc Furnace (EAF). BF refers to blast furnace slag formed during the initial stage of ironmaking, when iron ore is reduced using coke and limestone. Non-metallic impurities combine with fluxes to form molten slag, which is subsequently separated from the hot metal. LF slag is formed during ladle refining, where additives such as lime and dolomite are used to adjust the chemical composition and temperature of the steel. Two batches of LF slag from different industrial sources were considered and are hereafter denoted as LF.1 and LF.2. AOD slag is generated during the refining of stainless steel when oxygen and inert gases are blown into the molten metal to reduce its carbon content. This process leads to the formation of slag rich in lime and other oxides. EAF slag is produced in electric arc furnaces, where metal scrap or directly reduced iron is melted using high-voltage electric arcs. Steelmaking slag is formed from fluxes added to remove impurities and protect the molten steel from oxidation. As for the LF slag, two different batches were also considered in this case and are hereafter referred to as EAF.1 and EAF.2.

The results of chemical analysis of the slag are reported in the Supporting Information (Table S1) [26].

The thermoplastic matrix used in this study was a high-density polyethylene (HDPE) ELTEX A4040P provided by INEOS Olefins & Polymers Europe (Rolle, Switzerland) in powder form with a density of 0.94 g/cm<sup>3</sup> and a melting temperature of 128 °C.

### 2.2 | Accelerated Carbonation Test

The samples underwent an accelerated carbonation process carried out in a Berghof autoclave, which allows for control of pressure and temperature. In a preliminary phase, blank tests were conducted to evaluate the CO<sub>2</sub> absorption capacity of water used as the solvent in the subsequent experiments. Since the blank pressure drop was found to vary slightly with the external

**TABLE 1** | Comparison of the main mineral carbonation approaches reported in the literature and the strategy adopted in the present work, highlighting the underlying principles, advantages, and main limitations.

| Carbonation route       | Main principle                                       | Advantages                                   | Limitations                |
|-------------------------|------------------------------------------------------|----------------------------------------------|----------------------------|
| Direct carbonation [9]  | CO <sub>2</sub> reacts directly with alkaline solids | Simple                                       | Slower kinetics            |
| pH swing [10]           | Extraction + precipitation                           | High conversion                              | Additional chemicals       |
| Calcination–dissolution | Thermal activation + carbonation                     | High Ca availability                         | High energy demand         |
| This work               | Direct carbonation of mixed residues                 | Industrial symbiosis, no additional reagents | Lower process optimization |

**TABLE 2** | Composition of the samples FA and the investigated mixtures used for the carbonation tests.

| Sample ID | Fly ash (FA), % | Bottom ash (BA), % | Additional ash type |
|-----------|-----------------|--------------------|---------------------|
| FA        | 100             | —                  | —                   |
| MIX FDG   | 65              | 9                  | FDG—26%             |
| MIX BF    | 65              | 9                  | BF—26%              |
| MIX EAF.2 | 65              | 9                  | EAF.2—26%           |
| MIX AOD   | 65              | 9                  | AOD—26%             |
| MIX EAF.1 | 65              | 9                  | EAF.1—26%           |
| MIX LF.1  | 65              | 9                  | LF.1—26%            |
| MIX LF.2  | 65              | 9                  | LF.2—26%            |

Note: All mixture formulations contain the same proportions of fly ash (FA, 65 wt%) and bottom ash (BA, 9 wt%), while only the type of steel slag (26 wt%) was varied to evaluate its influence on CO<sub>2</sub> uptake and contaminant stabilization.

temperature, multiple blank values were determined under different conditions. These values, reported in the Supporting Information, were subtracted from the total pressure drop to obtain the effective amount of CO<sub>2</sub> absorbed by the samples. This procedure follows the methodology proposed by G. Montes-Hernandez et al. [9], allowing the contribution of CO<sub>2</sub> dissolution in water, minor temperature variations, and instrumental effects to be corrected through blank experiments performed under identical operating conditions.

Subsequently, the samples composed as shown in Table 2 were analyzed. Each mix was tested in four to five independent replicate experiments.

The carbonation tests were carried out by mixing 35 g of the selected sample with 45 mL of Milli-Q water at a constant temperature of 35 °C under magnetic stirring at 30 rpm for 24 h. In each experiment, the initial pressure was set to 15 bar, and the pressure drop was monitored. At the end of the experiment, the samples were filtered to separate the liquid fraction and solid fraction. The overall accelerated carbonation workflow is illustrated in Figure 1.

### 2.3 | Leaching Test

To evaluate the heavy metal immobilization, leaching tests were performed. 20 g of selected samples were mixed with 200 mL of

Milli-Q water, with a weight ratio of 1:10. The reaction mixture was mechanically stirred for 24 h at room temperature, followed by vacuum filtration through 0.45 µm pore-size membranes. The liquid fraction was then analyzed using total reflection X-ray Fluorescence (TXRF).

### 2.4 | XRD Analyses

The starting materials and the carbonated products were analyzed by performing an X-ray diffraction analysis (XRD) to identify the crystalline phases in the samples. The analysis was carried out using a PANalytical X'Pert PRO diffractometer equipped with a Cu Kα (1.5406 Å) radiation source and operated at 40 kV and a current of 40 mA, with the scans (2θ) from 10° to 90° having a step interval of 0.0167°. To identify the mineralogical composition, two different software programs were employed, namely, the software X'Pert HighScore Plus (version 2.1.0, PANalytical B.V., 2004) [27] and the open-source software Profex (version 5.2.2, Nicola Döbelin, 2003–2026) [28].

### 2.5 | TXRF Analyses

The elemental chemical analyses were carried out using a Horizon TXRF spectrometer (GNR S.r.l., Italy) equipped with a Mo anode (40 kV, 15 mA). Gallium was used as an internal standard to calculate the concentrations of the heavy metals present in the samples; a 1 g/L Ga solution in nitric acid stock solution (Gallium Standard for ICP Solution, Fluka, Sigma Aldrich, Saint Louis, MO, USA) was employed for this purpose. The samples for the analyses were prepared by weight to obtain solutions with 1 ppm of Ga and subsequently homogenized using a vortex shaker. A 10 µL drop of sample was deposited on a plexiglass reflector and dried on a hot plate at 50 °C. The residues were measured, and the spectra were analyzed with the instrumental software using routine deconvolution based on mono-element profiles to evaluate the peak areas.

### 2.6 | Thermoplastic Composites Preparation and Characterization

Before processing, all materials were dried in an oven at 50 °C for 48 h. The most promising carbonated product was then passed through different stacked sieves (600, 300, 100, 80, 30 µm) and sieved using a vibrating plate Endecotts Sieve Shaker E.V.S.1 (Endecotts Ltd. England) for 10 min. At the end, two size ranges



**FIGURE 1** | Schematic representation of the accelerated carbonation process.

were selected: 100–600  $\mu\text{m}$  and 30–100  $\mu\text{m}$ . HDPE was melt-compounded with the ash using a Thermo Haake Rheomix 600 internal mixer equipped with counter-rotating rotors, operating at 150 °C and 60 rpm for a total mixing time of 5 min. The filler was incorporated after 2 min of HDPE pre-melting at loadings of 5, 10, 20, and 30 wt%, and mixing was continued for an additional 3 min. The resultant compounds were compression-molded in a Carver hot-plate press at 150 °C for 8 min under an applied pressure of 4.9 MPa. In this way, square sheets 100 × 100 × 2 mm<sup>3</sup> were prepared. Volumetric fractions were calculated via the mixture rule starting from the nominal mass fractions and the gravimetric density values. The density of the powder filler and the matrix pellets was measured with a helium pycnometer, AccuPyc1330 (Micromeritics Instrument Corporation, GA, USA). The resulting density of the ash in the range of 30–100  $\mu\text{m}$  was equal to 2.63 g/cm<sup>3</sup>, and that of the ash in the range of 100–600  $\mu\text{m}$  was equal to 2.58 g/cm<sup>3</sup>.

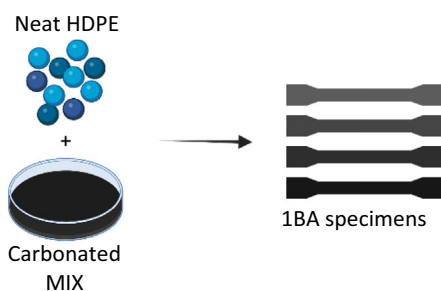
Table 3 reports the list of the prepared samples with the nominal mass composition and calculated volumetric composition.

Tensile tests were performed on the produced thermoplastic composite samples using an Instron 5969 (Instron, MA, USA)

**TABLE 3** | List of the prepared thermoplastic composite samples for mechanical characterization.

| Sample         | Neat HDPE, % wt | Granulometry of Ash, $\mu\text{m}$ | Ash, % wt | Ash, % vol |
|----------------|-----------------|------------------------------------|-----------|------------|
| HDPE           | 100.0           | —                                  | —         | —          |
| PE_5%_30-100   | 95              | 30–100                             | 5         | 1.9        |
| PE_10%_30-100  | 90              | 30–100                             | 10        | 3.8        |
| PE_20%_30-100  | 80              | 30–100                             | 20        | 8.2        |
| PE_30%_30-100  | 70              | 30–100                             | 30        | 13.3       |
| PE_5%_100-600  | 95              | 100–600                            | 5         | 1.9        |
| PE_10%_100-600 | 90              | 100–600                            | 10        | 3.9        |
| PE_20%_100-600 | 80              | 100–600                            | 20        | 8.3        |
| PE_30%_100-600 | 70              | 100–600                            | 30        | 13.5       |

Note: The most promising carbonated product was used as filler (here reported as ash).



testing machine equipped with a load cell of 50 kN and operating at a crosshead speed of 20 mm/min, for the determination of elastic modulus ( $E$ ), maximum tensile stress ( $\sigma_{\text{max}}$ ), and elongation at break ( $\epsilon_b$ ).  $E$  was calculated as the secant modulus considering the stress levels associated with strain values of 0.05% and 0.25%. The tensile tests were carried out on ISO 527 1BA specimens. The materials used for specimen preparation and the tensile testing setup are shown in Figure 2.

## 2.7 | Statistical Analysis

Data were reported as mean  $\pm$  standard deviation. Carbonation experiments were performed in four or five independent replicates, whereas tensile tests were performed on at least five specimens per composition. Mechanical data were analyzed using one-way ANOVA, followed by Tukey's post hoc test ( $\alpha = 0.05$ ). No data transformation or outlier removal was applied.

## 3 | Results and Discussion

### 3.1 | Accelerated Carbonation Tests

Multiple experiments were performed for each mix. Table 4 presents the mean values of the carbonation tests for each sample, showing that the mix containing the AOD slag exhibits the highest CO<sub>2</sub> absorption. Complete tables with all carbonation test results, along with the associated pressure drop graphs, are provided in the Supporting Information (Tables S2–S11).

The results show that the mixture containing AOD slag exhibited the highest CO<sub>2</sub> uptake, whereas the mixture containing EAF.2 slag showed the lowest CO<sub>2</sub> uptake. For comparison, the CO<sub>2</sub> uptake of the individual BA and AOD samples is reported in Table 4. Since all MIX formulations contain the same proportions of FA (65 wt%) and BA (9 wt%), while only the slag type (26 wt%) varies, the observed differences in carbonation behavior cannot be attributed to variations in the FA or BA contents. Instead, they are mainly associated with the nature of the added slag and its mineralogical characteristics.

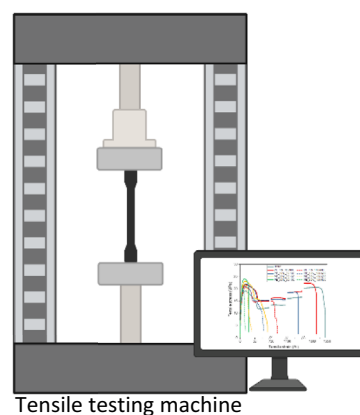


**FIGURE 2** | Materials used for specimen preparation and experimental setup for tensile testing.

**TABLE 4** | Mean  $\pm$  standard deviation values of the carbonation test results for each mix and their comparison with the FA sample.

| Sample    | P <sub>initial</sub> , bar | P <sub>final</sub> , bar | Pressure drop, bar | n, mol | Absorbed CO <sub>2</sub> , g | % Absorbed CO <sub>2</sub> vs. FA |
|-----------|----------------------------|--------------------------|--------------------|--------|------------------------------|-----------------------------------|
| FA        | 15.20                      | 6.69                     | 4.92               | 0.068  | 3.00 $\pm$ 0.32              | —                                 |
| MIX FDG   | 15.24                      | 6.44                     | 4.91               | 0.068  | 3.00 $\pm$ 0.38              | 0%                                |
| MIX BF    | 15.34                      | 6.89                     | 5.02               | 0.070  | 3.08 $\pm$ 0.36              | 3%                                |
| MIX AOD   | 15.35                      | 6.13                     | 6.61               | 0.092  | 4.04 $\pm$ 0.24              | 35%                               |
| MIX EAF.1 | 15.43                      | 7.34                     | 5.50               | 0.076  | 3.35 $\pm$ 0.27              | 12%                               |
| MIX EAF.2 | 15.35                      | 6.89                     | 4.09               | 0.057  | 2.49 $\pm$ 0.62              | −17%                              |
| MIX LF.1  | 15.39                      | 6.58                     | 6.22               | 0.086  | 3.79 $\pm$ 0.43              | 26%                               |
| MIX LF.2  | 15.48                      | 6.98                     | 5.65               | 0.078  | 3.45 $\pm$ 0.14              | 15%                               |
| AOD       | 15.44                      | 1.71                     | 10.65              | 0.133  | 5.86 $\pm$ 0.29              | 95%                               |
| BA        | 15.71                      | 8.02                     | 4.62               | 0.058  | 2.54 $\pm$ 0.23              | −15%                              |

Based on the carbonation tests performed on the individual components, the weighted-average CO<sub>2</sub> uptake expected for the MIX AOD is 3.70  $\pm$  0.26 g CO<sub>2</sub>. The experimentally measured value (4.04  $\pm$  0.24 g CO<sub>2</sub>) is slightly higher than this predicted value, indicating a positive deviation from the expected additive behavior. Although this observation suggests that the mixed formulation may favor carbonation beyond the weighted contribution of its individual components, further investigation is required to determine whether this difference is statistically significant.

To better understand the origin of this behavior, further analyses were carried out on the samples exhibiting the highest and lowest carbonation performance. Accordingly, XRD analyses were performed on FA, AOD slag, MIX AOD, MIX EAF.2, and their corresponding carbonated products. Furthermore, to evaluate whether carbonation promotes heavy metal and chloride immobilization, TXRF analyses were performed on the leachates obtained from the investigated samples.

### 3.2 | XRD Analyses

The XRD patterns of the raw (FA) and carbonated (C-FA) samples are reported in Figure 3.

The FA sample is mostly composed of alkaline and Ca-rich phases (CaClOH, portlandite, and tobermorite) together with soluble salts (sylvite and sodium chloride) and anhydrite. After the carbonation process, the disappearance of portlandite and CaClOH is observed; these phases react efficiently with CO<sub>2</sub> to give calcium carbonate (calcite and vaterite) [29]. Similarly, tobermorite is not detected in the carbonated sample, suggesting its transformation into carbonates and amorphous siliceous products [30, 31]. Anhydrite and sodium chloride, on the other hand, are essentially inert with respect to the process and persist in the carbonated sample.

Subsequently, the XRD patterns of the AOD slag and carbonated AOD (C-AOD) were compared (Figure 4).

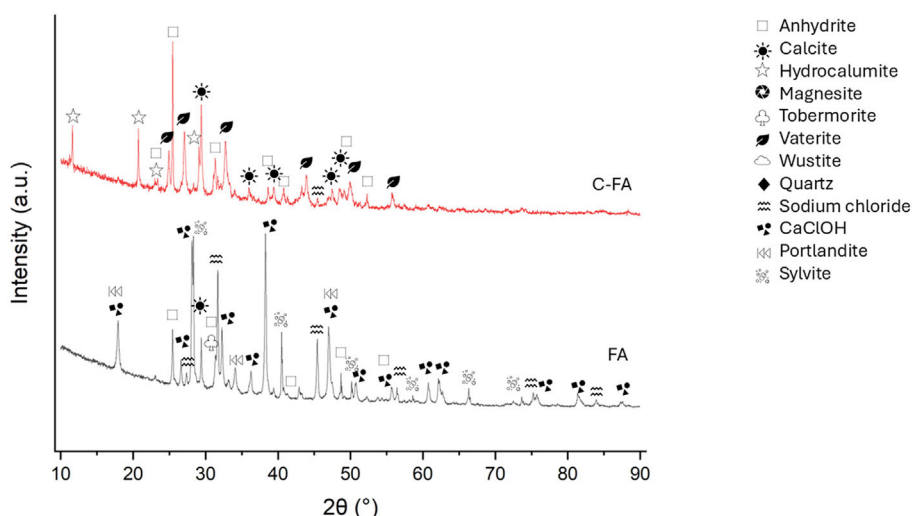


**FIGURE 3** | Comparison of XRD patterns. C-FA (red): calcite (CaCO<sub>3</sub>), anhydrite (CaSO<sub>4</sub>), Vaterite (CaCO<sub>3</sub>), sodium chloride (NaCl), and hydrocalumite ([Ca<sub>2</sub>Al(OH)<sub>6</sub>]Cl·2H<sub>2</sub>O). FA (black): anhydrite (CaSO<sub>4</sub>), CaClOH, sylvite (KCl), sodium chloride (NaCl), portlandite (Ca(OH)<sub>2</sub>), and tobermorite (Ca<sub>4</sub>Si<sub>6</sub>O<sub>17</sub>(H<sub>2</sub>O)<sub>2</sub>·(Ca·3H<sub>2</sub>O)).

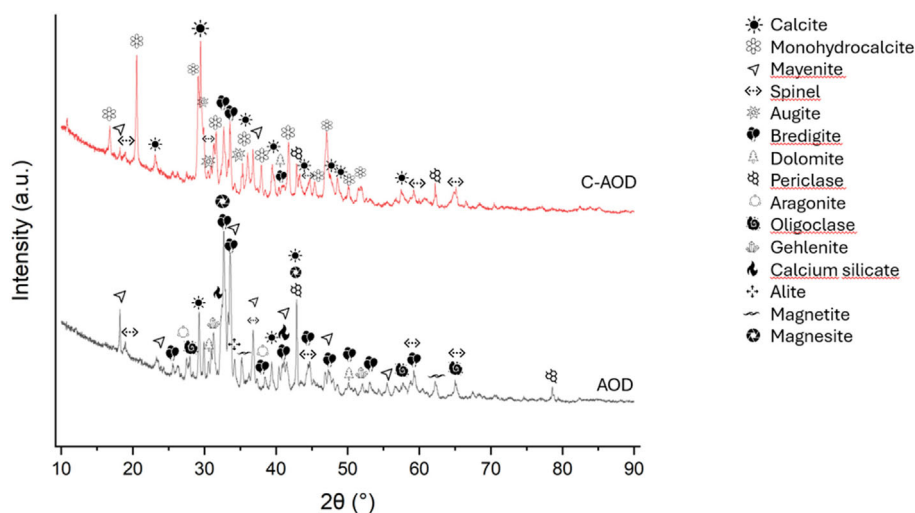


**FIGURE 4** | Comparison of XRD patterns. C-AOD (red): calcite ( $\text{CaCO}_3$ ), monohydrocalcite ( $\text{CaCO}_3 \cdot \text{H}_2\text{O}$ ), bredigite ( $\text{Ca}_7\text{Mg}(\text{SiO}_4)_4$ ), mayenite ( $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ ), spinel ( $\text{MgAl}_2\text{O}_4$ ), periclase ( $\text{MgO}$ ), augite ( $(\text{Ca,Mg,Fe})_2\text{Si}_2\text{O}_6$ ), and dolomite ( $\text{CaMg}(\text{CO}_3)_2$ ). AOD (black): calcite ( $\text{CaCO}_3$ ), alite ( $(\text{CaO})_3 \cdot \text{SiO}_2$ ), bredigite ( $\text{Ca}_7\text{Mg}(\text{SiO}_4)_4$ ), magnesite ( $\text{MgCO}_3$ ), mayenite ( $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ ), spinel ( $\text{MgAl}_2\text{O}_4$ ), aragonite ( $\text{CaCO}_3$ ), oligoclase ( $(\text{Na,Ca})(\text{Al,Si})_4\text{O}_8$ ), calcium silicate ( $\text{Ca}_2\text{O}_4\text{Si}$ ), periclase ( $\text{MgO}$ ), dolomite ( $\text{CaMg}(\text{CO}_3)_2$ ), and gehlenite ( $\text{Ca}_2\text{Al}(\text{AlSi})\text{O}_7$ ).

In these experiments, bredigite showed the most significant conversion to carbonates, mainly monohydrocalcite, and amorphous siliceous products. Periclase was converted into dolomite and hydrated magnesium carbonates [32]. Another very reactive phase is represented by mayenite, which is not present as a crystalline phase in the carbonated product, meaning that it underwent complete transformation into  $\text{CaCO}_3$ , possibly in the hydrated form, along with hydrated or amorphous aluminum phases. In contrast, spinel exhibited negligible involvement in the carbonation process.

The comparison between the XRD patterns of C-FA and MIX AOD is then examined (Figure 5).

Carbonation was found to be more efficient in MIX AOD than in FA, as shown by the greater formation of calcium carbonate

in the final phases. MIX AOD contains numerous oxide and silicate phases that are highly reactive, while the FA sample is mainly composed of saline and hydroxide phases ( $\text{CaClOH}$ ,  $\text{NaCl}$ , and  $\text{KCl}$ ) and of tobermorite; as a result, fewer phases actively participate into carbonation reactions. After the experiment, in MIX AOD, phases like calcite and vaterite become predominant, indicating a broad conversion of calcium in carbonate phases; on the other hand, the FA sample shows limited carbonation [29, 32].

The carbonation of the sample containing BF showed the worst results. BF exhibits low carbonation, which can be attributed to its high amorphous fraction, which limits the availability of reactive Ca-bearing phases and reduces their effective participation in carbonation reactions (Figure S11) [33].

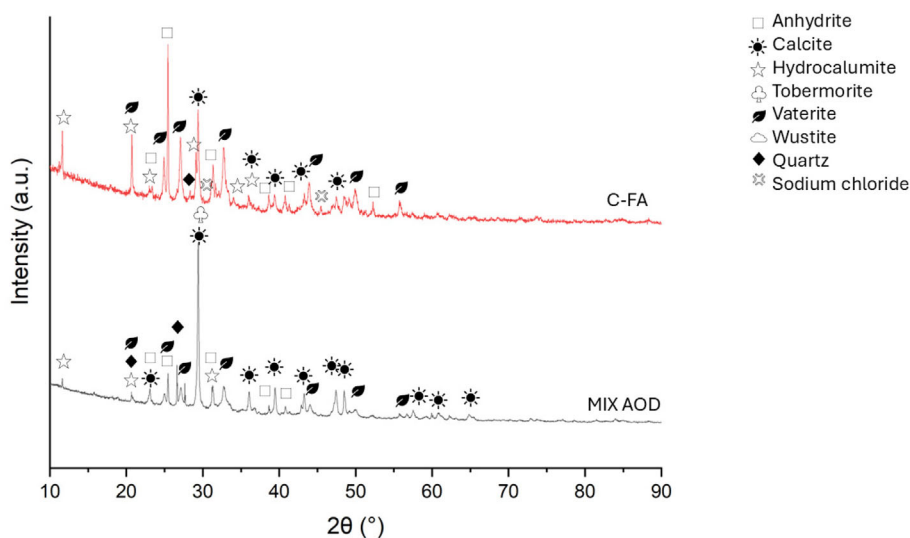


**FIGURE 5** | Comparison of XRD patterns. C-FA (red): calcite ( $\text{CaCO}_3$ ), anhydrite ( $\text{CaSO}_4$ ), vaterite ( $\text{CaCO}_3$ ), sodium chloride ( $\text{NaCl}$ ), hydrocalumite ( $[\text{Ca}_2\text{Al}(\text{OH})_6]\text{Cl} \cdot 2\text{H}_2\text{O}$ ). MIX AOD (black): calcite ( $\text{CaCO}_3$ ), anhydrite ( $\text{CaSO}_4$ ), vaterite ( $\text{CaCO}_3$ ), tobermorite ( $\text{Ca}_4\text{Si}_6\text{O}_{17}(\text{H}_2\text{O})_2 \cdot (\text{Ca} \cdot 3\text{H}_2\text{O})$ ), quartz ( $\text{SiO}_2$ ), hydrocalumite ( $[\text{Ca}_2\text{Al}(\text{OH})_6]\text{Cl} \cdot 2\text{H}_2\text{O}$ ).

For completeness, an additional comparison between the XRD patterns of MIX AOD and MIX EAF.2 was carried out and is available in the Supporting Information (Figure S12).

### 3.3 | TXRF Analyses

TXRF was used to determine the elemental composition of the solutions obtained from the leaching of FA, C-FA, MIX AOD, and MIX EAF.2. Figure 6 displays the results obtained from the analyses of the samples. The spectrum of each sample is reported in the Supporting Information (Figures S13–S16).

The carbonation process contributes to the stabilization of heavy metals, as evidenced by the results of the TXRF analyses performed on FA and C-FA. FA exhibits high concentrations of calcium (6800 ppm) and chlorine (12,800 ppm), together with significant amounts of heavy metals: in particular, lead is present at a concentration of 70 ppm, while zinc reaches 4.1 ppm. After carbonation, the lead concentration decreases to 8 ppm, while zinc increases to 1 ppm, indicating a substantial reduction in their leachability. The treated sample also shows lower concentrations of calcium (2800 ppm) and chlorine (4200 ppm). The reduction in calcium concentration is related to its consumption during carbonation reactions and to its reaction with chlorine, leading to the formation of hydrocalumite ( $[\text{Ca}_2\text{Al}(\text{OH})_6]\text{Cl}\cdot 2\text{H}_2\text{O}$ ), a Friedel's salt. Aluminum helps the reaction between calcium and chlorides that leads to the precipitation of stable chloroaluminate phases, which structurally incorporate  $\text{Cl}^-$  ions into their lattice.

Consequently, chlorine is partially immobilized through its incorporation into Friedel's salt and related chloroaluminate phases, leading to a reduction in its free and mobile form, as confirmed by the decrease in the chloride concentration observed in the leaching tests [23, 25].

In MIX AOD and MIX EAF.2, zinc and lead concentrations were below the limit of detection (LOD:  $\text{Zn} = 0.9$  ppm and  $\text{Pb} = 2.5$  ppm), and therefore no detectable traces of either element were observed [34]. This is due to the composition of the MIX since BA was added to the samples with the purpose of further stabilizing the heavy metals [18, 20]. BA provides amorphous silica and alumina [17], which, under highly alkaline conditions and in the presence of calcium ions, promote pozzolanic reactions. These reactions lead to the formation of cementitious phases such as calcium silicate hydrates (C–S–H), detected by XRD as tobermorite in MIX AOD, and calcium aluminate hydrates (C–A–H). Such phases are known to incorporate or adsorb heavy metals within their structure, thereby significantly reducing their leachability [21]. The combined use of FA, BA, and slag results in enhanced carbonation and stabilization performance, demonstrating the potential of industrial symbiosis for optimizing waste valorization processes.

### 3.4 | Proof-of-Concept Application of Carbonated Residues in HDPE Composites

The objective of this section is not to develop an optimized HDPE composite but to demonstrate that the carbonated residue obtained after  $\text{CO}_2$  sequestration can be successfully reused as a functional filler, thereby closing the material loop within an industrial symbiosis framework.

Tensile tests were performed on neat HDPE and on the corresponding composites for their mechanical characterization (Figure 7).

The results of the tensile test highlight a clear effect of the filler addition on the mechanical behavior of HDPE. As shown in Figure 7a, the incorporation of MIX AOD generally increases the elastic modulus compared to that of neat HDPE, indicating

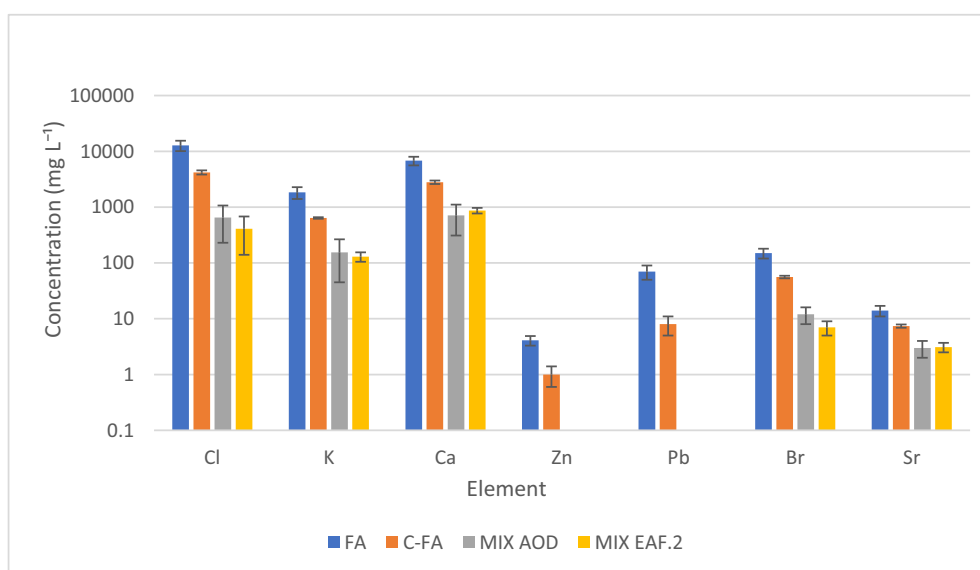


**FIGURE 6** | Elemental composition of leached solutions. The y-axis is shown on a logarithmic scale. Data are reported as the average  $\pm$  standard deviation of three TXRF measurements. Cr, Ni, and V were also analyzed in the leaching tests. However, their concentrations were below the analytical limits of detection for all samples (LOD:  $\text{Cr} = 5.0$  ppm,  $\text{V} = 5.6$  ppm;  $\text{Ni} = 1.5$  ppm); [34] therefore, these results are not reported.

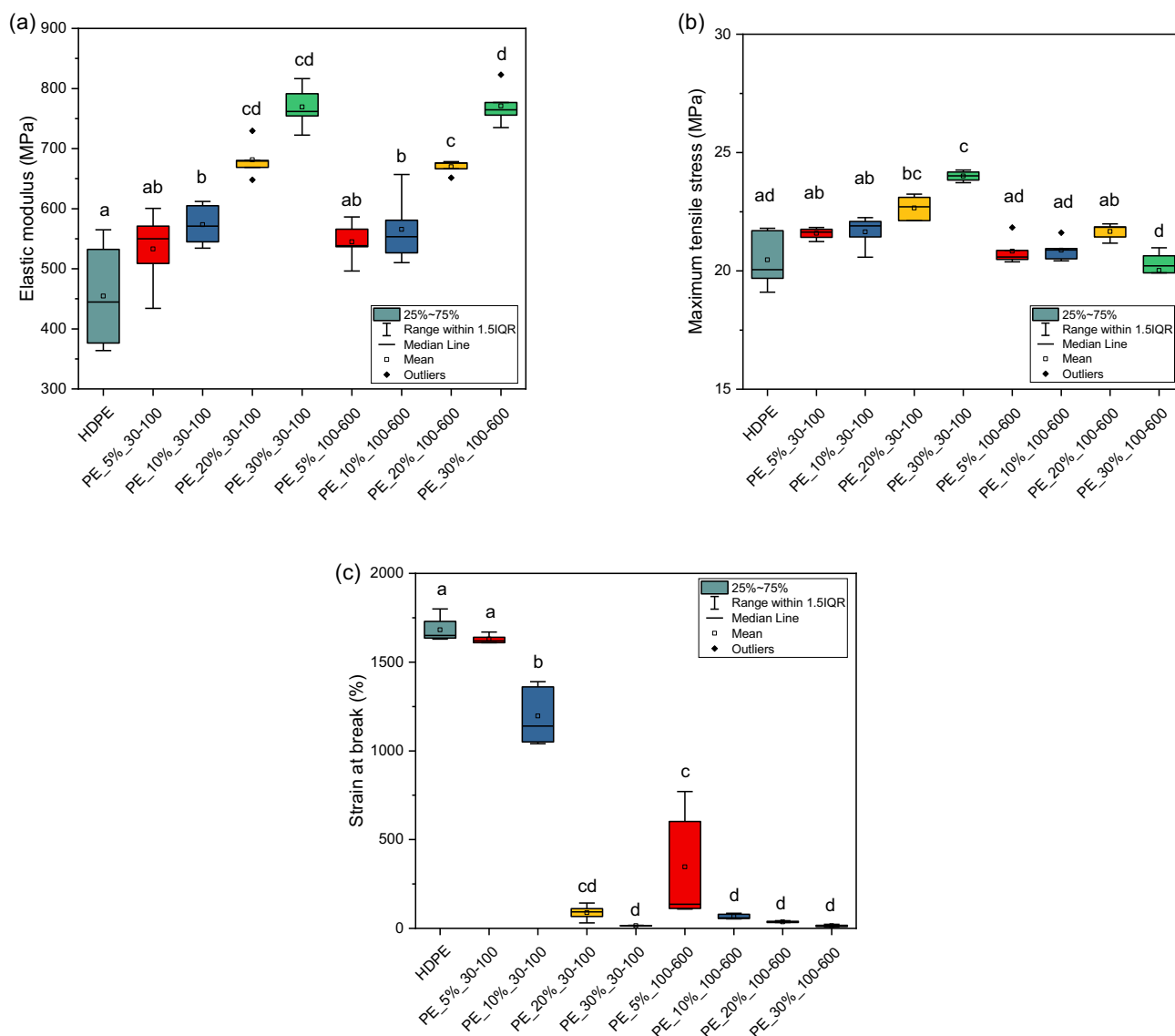


**FIGURE 7** | Results of quasi-static tensile tests on neat HDPE and the relative composites. (a) Elastic modulus; (b) maximum tensile stress; and (c) strain at break. Different letters indicate that results are statistically different ( $p < 0.05$ ).

a stiffening effect of the inorganic filler. This improvement becomes progressively more pronounced with increasing filler content (+70% for the samples with 30 wt% of MIX AOD); however, it appears to be only marginally influenced by the filler particle sizes. The maximum tensile stress results (Figure 7b) show a significant increase for composites containing finer particles, with an increase of approximately 17% for the sample with 30 wt% of MIX AOD. This behavior can be attributed to the improved dispersion of smaller particles within the polymer matrix, which promotes more efficient stress transfer and reduces the presence of critical flaws and defects [35]. Conversely, no substantial increase in tensile strength, compared to that in neat HDPE, is observed for composites reinforced with coarser particles. This behavior may be associated with particle agglomeration and weaker filler/matrix interfacial interactions, which generate stress concentration effects [36, 37].

Additionally, an interesting observation emerges for the sample containing 10 wt% of 30–100  $\mu\text{m}$  particles, which exhibits

mechanical properties comparable to those of neat HDPE. Unlike other composites, which show a marked reduction in elongation at break, this formulation maintains the ductility of the unfilled polymer. This suggests that up to 10 wt% of polymer can be effectively replaced with this filler fraction, potentially reducing both material costs and environmental impact without compromising the mechanical performance.

Finally, the strain at break (Figure 7c) decreased significantly with the FA addition, reflecting reduced ductility and increased brittleness of the composites. The sample reinforced with 5 wt% of finer particles shows no statistically significant difference compared to neat HDPE. However, as the filler content increases, the elongation at break drops substantially, with a reduction of approximately 28% at 10 wt% and up to 99% at 30 wt%. An even more pronounced embrittlement is observed in composites filled with coarser particles, where the strain at break decreases by about 79% already at 5 wt%, reaching a reduction of 99% at 30 wt%. These reductions can be attributed to the fact that the

addition of MIX AOD constraints plastic deformation mechanisms and limits the ability of HDPE chains to undergo large-scale molecular rearrangements [38].

Figure 8 shows the variation of tensile strength as a function of Young's modulus, allowing a direct comparison among the investigated materials and literature data.

In the tensile strength–Young's modulus diagram, the developed materials based on MIX AOD-filled HDPE are located in the low-to-intermediate modulus region ( $\approx 0.5$ – $0.9$  GPa) and exhibit tensile strength values in the range of  $\sim 18$ – $25$  MPa. This region is consistent with that typically associated with polyolefin-based polymers while showing some relevant improvements. Although the developed composites exhibit lower tensile strength and Young's modulus compared to most PP-based systems, they show a simultaneous increase in both properties with increasing filler content, indicating effective reinforcement and a progressive shift toward more competitive mechanical performance.

Overall, the developed composites exhibit a clear trade-off between stiffness and ductility. The incorporation of carbonated MIX AOD increases Young's modulus and, for fine particles, also improves tensile strength, whereas elongation at break decreases with increasing filler content. Therefore, these composites are particularly suited for applications requiring dimensional stability and rigidity rather than high deformability, such as rigid building components, non-structural construction products, urban furniture, drainage systems, and technical packaging.

Although the mechanical characterization is not intended to optimize the composite formulation, these results demonstrate that the carbonated residues obtained through mineral carbonation can be successfully valorized as functional fillers, providing a potential end use for the  $\text{CO}_2$ -sequestering material.

### 3.5 | Overall Data Discussion

The carbonation results highlight a strong dependence of  $\text{CO}_2$  uptake on both the chemical composition and the mineralogical nature of the investigated mixtures. Among the analyzed samples, MIX AOD exhibits the highest carbonation efficiency, with a  $\text{CO}_2$  uptake increase of approximately 34% compared to FA, while MIX EAF.2 shows the lowest performance. The carbonation experiments performed on the individual FA, BA, and AOD (see Table 4) samples also allowed the expected  $\text{CO}_2$  uptake of MIX AOD to be estimated from the weighted average of its components ( $3.70 \pm 0.26$  g  $\text{CO}_2$ ). The experimentally measured value ( $4.04 \pm 0.24$  g  $\text{CO}_2$ ) was slightly higher than this prediction, indicating a positive deviation from the expected additive behavior. Although this observation cannot be considered conclusive evidence of a synergistic effect, it suggests that the mixed formulation may favor carbonation beyond the weighted contribution of its individual components.

The mineralogical transformations observed by XRD, namely, the formation of calcite, vaterite, and monohydrocalcite together with the disappearance of reactive Ca-bearing phases, confirm

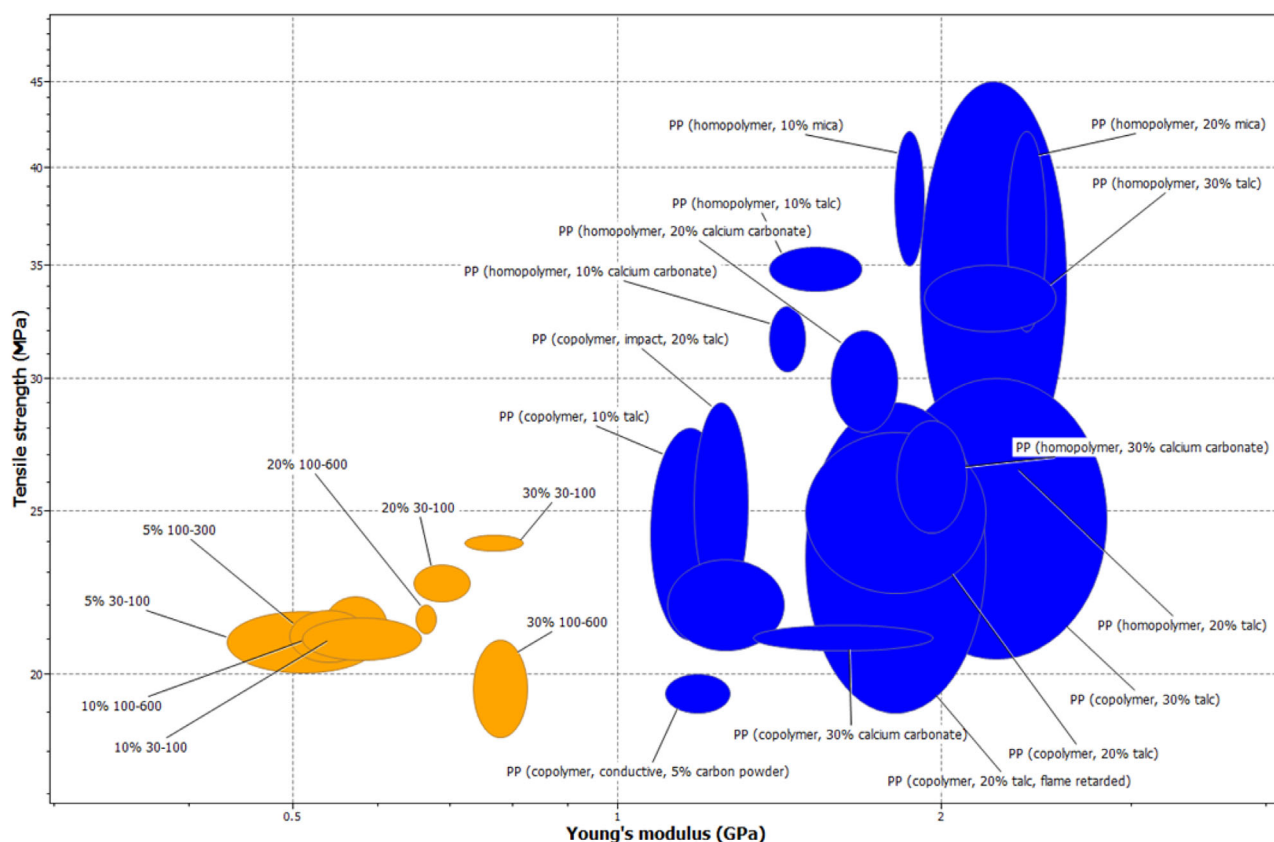


**FIGURE 8** | Tensile strength as a function of Young's modulus for polyethylene-based composites. Literature data for different material systems are reported for comparison with the material developed in this work. Orange circles denote samples prepared in this study.

that the corrected pressure drop corresponds to effective mineral carbonation rather than only physical CO<sub>2</sub> dissolution.

The chemical composition of the slags, reported in the Supporting Information (Table S1), provides key insights into this behavior. In particular, AOD slag is characterized by a high content of CaO and MgO combined with a very low Fe<sub>2</sub>O<sub>3</sub> fraction. This results in a high (CaO + MgO)/Fe<sub>2</sub>O<sub>3</sub> ratio, which correlates well with the enhanced carbonation performance (Figure 9). In contrast, EAF slags contain a significantly higher Fe<sub>2</sub>O<sub>3</sub> content (up to ~50 wt%), which reduces the relative fraction of reactive Ca- and Mg-bearing phases and negatively affects CO<sub>2</sub> sequestration. Figure 9 provides further insight into the factors controlling carbonation efficiency. A general increase in CO<sub>2</sub> uptake is observed with increasing (CaO + MgO)/Fe<sub>2</sub>O<sub>3</sub> ratio, indicating that the relative abundance of alkaline oxides with respect to Fe-rich phases strongly influences carbonation performance. However, the relationship is not perfectly linear. In particular, BF slag exhibits one of the highest (CaO + MgO)/Fe<sub>2</sub>O<sub>3</sub> ratios but only a marginal increase in the CO<sub>2</sub> uptake. This behavior demonstrates that the bulk chemical composition alone cannot explain the carbonation performance. Instead, mineralogical characteristics and phase accessibility were considered. As confirmed by the XRD analyses, BF is characterized by a very high amorphous fraction, which likely limits the accessibility of reactive Ca-bearing phases. EAF-based mixtures contain a higher proportion of stable or already partially carbonated phases, such as magnesite and Fe-rich minerals, which show limited reactivity and therefore reduce the overall carbonation efficiency. Conversely, AOD exhibits both favorable chemical composition and highly reactive crystalline Ca- and Mg-bearing phases, resulting in the highest carbonation efficiency among the investigated mixtures.

The leaching results provide further evidence of the effectiveness of the carbonation process. A significant reduction in the concentration of heavy metals, particularly Pb and Zn, is observed after the carbonation of FA, confirming the stabilization effect of the process. This behavior can be attributed to the formation of carbonate phases and secondary mineral phases capable of immobilizing contaminants.

In addition, a marked decrease in the chlorine concentration is observed after carbonation, which can be linked to the formation

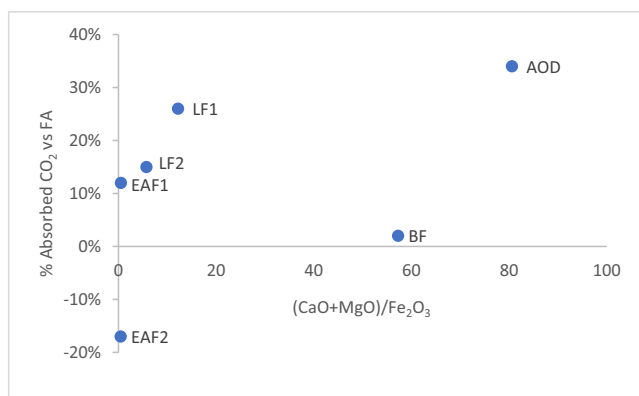


**FIGURE 9** | Correlation between (CaO + MgO)/Fe<sub>2</sub>O<sub>3</sub> ratio and CO<sub>2</sub> uptake.

of chloroaluminate phases such as Friedel's salt. The presence of aluminum and calcium promotes the incorporation of chloride ions into stable crystalline structures, reducing their mobility. This effect is further enhanced in mixtures containing BA, which provides additional silica and alumina, promoting pozzolanic reactions and the formation of C-S-H and C-A-H phases that contribute to contaminant immobilization.

Overall, the results demonstrate that CO<sub>2</sub> uptake is governed by the interplay between chemical composition, mineralogical phase distribution, and phase reactivity. In particular, high carbonation efficiency is achieved when reactive Ca- and Mg-bearing phases are abundant and not diluted by nonreactive components such as iron oxides or poorly reactive amorphous phases. The results demonstrate that carbonation efficiency is governed by the interplay between chemical composition, mineralogical structure, and phase accessibility, rather than by bulk composition alone.

In addition to the carbonation efficiency and stabilization performance, the results also have implications for the mechanical behavior of the developed materials. The mineralogical transformations induced by carbonation, including the formation of calcium carbonate phases and amorphous silica-rich products, may contribute to the modification of particle surface characteristics and interfacial interactions with the matrix. These changes can influence the dispersion and bonding of the filler within the polymer, resulting in improved stiffness and, in some cases, enhanced tensile strength.

In particular, the observed increase in the elastic modulus and tensile strength for composites containing finer particles can be linked to the improved distribution and higher surface reactivity of the carbonated material. This confirms that the carbonation process not only enables CO<sub>2</sub> sequestration and contaminant stabilization but also contributes to the development of value-added materials with tailored mechanical performance, characterized by increased stiffness and tensile strength at the expense of ductility, making these materials suitable for specific semi-structural applications. These findings further support the potential of integrated waste valorization strategies within an industrial symbiosis framework.

## 4 | Conclusions

This study demonstrates that mineral carbonation of mixed industrial residues represents an effective and integrated strategy for simultaneous CO<sub>2</sub> sequestration and environmental stabilization. The combined use of FA, BA, and steel slags exploits their complementary chemical and mineralogical properties, enhancing both carbonation efficiency and contaminant immobilization within a circular economy framework.

The carbonation performance was strongly influenced by both the chemical composition and mineralogy of the investigated mixtures. In particular, the AOD-based mixture exhibited the highest CO<sub>2</sub> uptake, with an increase of approximately 35% compared to fly ash alone. The experimentally measured CO<sub>2</sub> uptake of MIX AOD also seems slightly higher than the value predicted

from the weighted contribution of its individual components, suggesting that the mixed formulation may favor carbonation beyond a purely additive behavior, although this trend requires further statistical confirmation.

This behavior is attributed to the high availability of reactive Ca- and Mg-bearing phases combined with a low content of iron-rich components. Conversely, slags characterized by a high  $\text{Fe}_2\text{O}_3$  content showed reduced carbonation performance due to the predominance of inert phases that dilute the reactive fraction.

The study also demonstrates that  $\text{CO}_2$  sequestration cannot be predicted solely based on bulk CaO and MgO content. Instead, carbonation efficiency depends on the accessibility and reactivity of the mineral phases, with amorphous or poorly reactive phases, as observed for BF slag, limiting the availability of reactive species.

Carbonation also significantly improved the environmental performance of the materials. Leaching tests showed marked reductions in Pb and Zn concentrations, together with lower chlorine mobility associated with the formation of stable chloroaluminate phases such as Friedel's salt. The addition of BA further enhanced these effects by supplying silica and alumina, promoting pozzolanic reactions and the formation of secondary phases such as C-S-H and C-A-H, which contribute to contaminant immobilization.

Beyond environmental benefits, the results indicate that carbonation-induced mineralogical transformations also influence the performance of the resulting materials. The formation of carbonate phases and amorphous silica-rich products contribute to increased stiffness and, in some cases, enhanced tensile strength in polymer-based composites. However, this improvement was accompanied by a reduction in ductility, whose extent depended on the filler content and particle size.

Overall, the combined use of FA, BA, and slag enhances both carbonation and stabilization, demonstrating the potential of industrial symbiosis for optimizing waste valorization processes. This approach represents a robust pathway for the development of sustainable materials based on permanently mineralized  $\text{CO}_2$ , while future life cycle assessment studies will be required to quantify their overall carbon footprint and verify their potential classification as carbon-negative materials.

From an industrial perspective, the proposed approach is fully consistent with the principles of cleaner production and industrial symbiosis since it simultaneously promotes  $\text{CO}_2$  sequestration, hazardous waste stabilization, and the production of secondary raw materials. Rather than treating FA, BA, and steel slags as independent waste streams, the process integrates them into a single valorization pathway, reducing landfill disposal while increasing resource efficiency.

Nevertheless, several challenges remain before their industrial implementation. Future research should address process scale-up, continuous reactor operation, optimization of  $\text{CO}_2$  supply, economic feasibility, and comprehensive life-cycle assessment. In particular, the long-term environmental performance and end-of-life behavior of the carbonated products

should be further investigated to fully quantify their contribution to sustainable industrial production.

Despite these remaining challenges, the present results demonstrate that combining complementary industrial residues through mineral carbonation represents a promising strategy for supporting cleaner production practices and circular economy implementation in industrial districts where multiple alkaline waste streams are simultaneously available.

## Author Contributions

**Erica Pietrobelli:** data curation, formal analysis, investigation, and writing – original draft. **Matteo Giosuè Vezzuto:** data curation, formal analysis, investigation, and writing – original draft. **Alessandra Zanoletti:** formal analysis, investigation, visualization, and writing – review and editing. **Alessandro Sorze:** data curation, investigation, and writing – original draft. **Giulia Fredi:** data curation, investigation, visualization, and writing – review and editing. **Elza Bontempi:** conceptualization, formal analysis, funding acquisition, investigation, methodology, project administration, resources, supervision, validation, visualization, and writing – original draft.

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## Conflicts of Interest

The authors declare no conflicts of interest.

## Data Availability Statement

Data are reported in Supporting Information.

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### Supporting Information

Additional supporting information can be found online in the Supporting Information section.