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# Electrochemical analysis of EAF slag-cement hydration and role of TIPA and glycine

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**Title:** Electrochemical analysis of EAF slag-cement hydration and role of TIPA and glycine.

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## **Abstract**

This study employs electrochemical analysis using alternating current impedance spectroscopy to investigate electric arc furnace slag (EAF) slag-cement hydration kinetics through organic ligand coordination with triisopropanolamine (TIPA) and glycine (Gly) in Portland cement blends containing carbonated and non-carbonated EAF. The electrochemical monitoring was correlated with calorimetry, thermogravimetric analysis, X-ray diffraction, setting measurements, and electrical conductivity to examine coordinating effects on hydration behavior. Results demonstrated that organic ligand coordination significantly influences hydration kinetics, with carbonated mineral phases showing greater electrochemical response than non-carbonated slag. TIPA and glycine coordination enhanced fluidity, accelerated setting time, increased compressive strength, and improved hydration efficiency by promoting phase transformation into calcium silicate hydrate. Electrochemical analysis revealed that ligand affects dissolution kinetics, phase formation, and microstructure development, influenced by EAF slag type. Impedance spectroscopy effectively monitored changes in electrical conductivity during hydration, correlating with microstructure resistivity and network connectivity evolution. This electrochemical approach highlights the complementary roles of impedance spectroscopy and calorimetry in monitoring organic ligand effects on cement hydration kinetics.

**Keywords:** Electrical conductivity, hydration, electric arc furnace slag, organic ligands, carbonation

## **1. Introduction**

The process of Portland cement production requires significant quantities of raw materials and energy while releasing substantial amounts of greenhouse gases into the atmosphere during manufacturing [1]. CO<sub>2</sub> emissions from cement production are estimated at approximately 7-10% of

global emissions [2]–[4]. Population growth drives increased demand in the construction sector, while more stringent environmental regulations compel cement industries to reduce pollution levels, as limestone decarbonization generates approximately 480 kg of CO<sub>2</sub> per tonne of clinker [5]. To address these challenges and comply with regulations, cement industries have begun modernizing by incorporating alternative materials such as metakaolin, fly ash, and industrial byproducts as clinker replacements to produce blended cements [2], [6]–[8].

Slags are byproducts of iron and steel manufacturing and the four main types are: blast furnace slag (BFS), basic oxygen furnace (BOF) slag, electric arc furnace (EAF) slag, and ladle furnace (LF) slag. Global production of these slag wastes is estimated at 190–290 million tonnes, with EAF slag representing 15–20% [9], [10]. The composition of these different steel slags varies depending on the manufacturing process, the type of furnace, and the type of iron ore used. It has primarily been used as an aggregate in concrete and for the construction of pavements and roads [11]. EAF slag as a supplementary cementitious material (SCM) has been challenging due to its low cementitious reactivity and due to the content of heavy metals [10].

Lee et al., [12] demonstrated that a 15% replacement of EAF slag in PC blends did not impact compressive strength after 28 days of hydration, while Bernado et al., [13] and Muhmood et al., [10] showed that addition up to 20% improved phase formation and microstructure. However, the hydration mechanism remains poorly understood due to phase products from the blended cement hydration and the crystalline nature of EAF slag. Specific elements in EAF slag, such as excess free calcium, influence the solidification process at significant replacement rates [9]. Carbonation effectively reduces free calcium content and provides volumetric stability for calcium carbonate and magnesium carbonate, as demonstrated by Li et al. [14]. This technique, traditionally employed for CO<sub>2</sub> sequestration, offers advantages in reactions with dicalcium silicates present in EAF slag to form carbonate compounds, promoting dissolution of Ca and Mg ions and accelerating reaction rate of EAF slag [15], [16]. Various characterization methods have been employed to understand the influence of EAF slag on reaction kinetics and microstructure development. Techniques such as calorimetry and rheology provide essential information during early reaction stages but fail to characterize sedimentation [17], [18]. On the other hand, while microscopy techniques are generally difficult to apply to fresh cement paste before final setting, X-ray diffraction (XRD) can be performed during this early stage either through in situ measurements on fresh paste or through ex situ analysis after stopping hydration at selected time intervals.

Earlier research has shown that increasing fineness and using chemical activators (NaOH, Na<sub>2</sub>SiO<sub>3</sub>, etc.) can enhance EAF slag reaction kinetics [19]. Additionally, chelating agents such as organic ligands can complex metal ions in the cement matrix pore solution, potentially increasing their solubility and reactivity [20]. Ligands such as triisopropanolamine (TIPA), triethanolamine

(TEA), glycine, ethylenediaminetetraacetic acid (EDTA), and citric acid have demonstrated potential effectiveness in Portland cement systems and blast furnace slag-based cements [21][22][23], though their influence on microstructural evolution in EAF slag-blended systems remains unclear. The method most frequently used to understand ion behavior in cement-based materials is ICP-OES (Inductively Coupled Plasma Optical Emission Spectroscopy) analysis by extracting liquid from the pore solution. This technique provides indirect insights into reaction kinetics within the interstitial pore solution by offering information on ionic concentrations of various species (Ca, K, Na, Al, Mg, S, Fe) [23][24], [25]. However, due to the rapid nature of chemical reactions occurring in hydrating cement pastes, precise sampling at desired time intervals is challenging, and instrumental limitations exist when analyzing highly alkaline fluids [26], indicating that certain characterization techniques have limitations in fully elucidating cement hydration processes.

Impedance spectroscopy is a promising technique for monitoring and providing information on the electrical mobility of ions in pore solution and the microstructure of various materials. This characterization method applies alternating current across a well-defined frequency range (10 Hz to 100 MHz) [26] [28], which enables separate resolution of the bulk ionic response of the pore solution at high frequencies and the electrode paste interfacial response at low frequencies; this dual sensitivity allows the technique to track both the ionic mobility of fresh fluid paste and the evolving resistive capacitive behaviour of the developing solid microstructure [29], making it uniquely suited to monitoring cement paste continuously from mixing through final hardening. In this analysis, the cement paste is considered as a circuit model where the electrical behavior depends on the resistance and reactance response of the material [28]–[30]. In this system, high-frequency impedance is considered as the overall response of the material, and at low frequency, it is considered as the material-electrode interface response. Historical limitations in applying this technique included measurement errors affecting impedance response, such as parasitic errors from cables, wires, and electrode-sample contacts [27]. With the advancements of electrochemical science, many solutions have been addressed to limit these various errors. Recent work with this technique has focused more on the hydration process of Portland cement to limit parasitic interference during the early stages of hydration. However, questions remain about whether alternating current impedance spectroscopy (ACIS) can effectively distinguish the influence of carbonated versus non-carbonated EAF slag and its reactivity during hydration while detecting the impact of organic ligand admixtures on electrical conductivity and curing at both early and later stages of these composite binders

The objective of this study is to employ electrical conductivity measurements to better understand the influence of non-carbonated and carbonated EAF slag on blended cement and to investigate the reaction mechanisms of organic ligands such as TIPA and glycine (Gly) on reactivity and hydration.

Additionally, correlations will be established between setting time, isothermal calorimetry, thermogravimetric analysis, and electrical conductivity.

## 2. Materials and methods

### 2.1. Raw materials

Four types of raw material were used in this study, CEM I type cement from Finland (PC), EAF from Sweden (EAF1), EAF from India (EAF2) and EAF from Poland (EAF3). The chemical composition of all the raw materials used is illustrated in Table 1 and the X-ray diffraction pattern were shown in Fig. 1. The particle size distribution was determined by using a Beckman Coulter LS 13320 and the results are illustrated in Fig. 2. The  $d_{10}$ ,  $d_{50}$ , and  $d_{90}$  of PC and different EAF slag differ from each other (PC ( $d_{10}=0.91\ \mu\text{m}$ ,  $d_{50}=9.73\ \mu\text{m}$ ,  $d_{90}=29.06\ \mu\text{m}$ ) EAF1 ( $d_{10}=0.42\ \mu\text{m}$ ,  $d_{50}=4.50\ \mu\text{m}$ ,  $d_{90}=35.39\ \mu\text{m}$ ), EAF2 ( $d_{10}=0.73\ \mu\text{m}$ ,  $d_{50}=7.60\ \mu\text{m}$ ,  $d_{90}=27.12\ \mu\text{m}$ ) EAF3 ( $d_{10}=0.58\ \mu\text{m}$ ,  $d_{50}=5.20\ \mu\text{m}$ ,  $d_{90}=46.22\ \mu\text{m}$ )). All EAF slag samples were received pre-ground with fine particle size, except EAF3 slag, which presented a larger particle size. Therefore, the EAF3 slag was milled for 6 hours using a stainless steel ball mill; then, after particle size analysis was determined, it offered less reactive behaviour compared to EAF1 and EAF2, as demonstrated by Elijan et al.,[31].

Table 1: Chemical composition of PC and different EAF slag.

| Oxides (%) | CaO  | SiO <sub>2</sub> | Al <sub>2</sub> O <sub>3</sub> | Fe <sub>2</sub> O <sub>3</sub> | Na <sub>2</sub> O | K <sub>2</sub> O | MgO  | P <sub>2</sub> O <sub>5</sub> | TiO <sub>2</sub> | SO <sub>3</sub> | Cl  |
|------------|------|------------------|--------------------------------|--------------------------------|-------------------|------------------|------|-------------------------------|------------------|-----------------|-----|
| PC         | 64.6 | 19.2             | 4.5                            | 4                              | 0.6               | 0.86             | 2.9  | 0.07                          | 0.2              | 2.7             | 0.1 |
| EAF1       | 35   | 15               | 5.7                            | 34                             | 0.2               | 0.1              | 11.2 | 0.6                           | 2.1              | -               | -   |
| EAF2       | 37.5 | 17.2             | 10.1                           | 24.2                           | 0.1               | 0.1              | 5.2  | 0.8                           | 0.6              | 0.9             | 0   |
| EAF3       | 26.7 | 17.5             | 10.3                           | 34.1                           | 0.1               | 0                | 3.6  | 0.3                           | 0.5              | 0.4             | 0   |

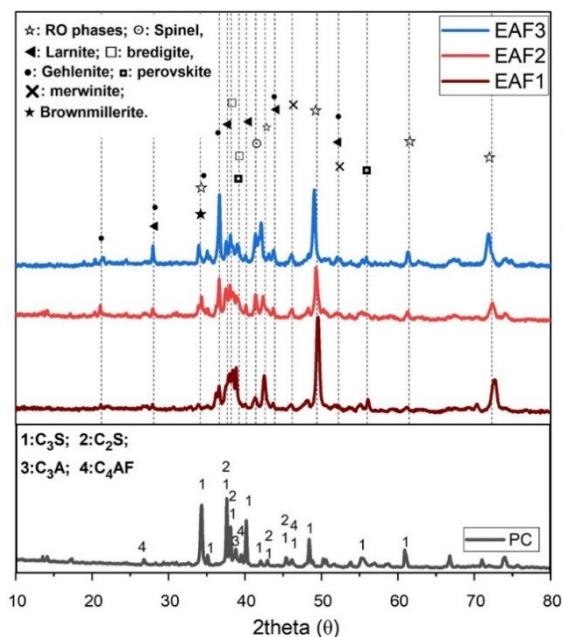


Figure 1: XRD patterns of PC and different EAF slags (RO phases,  $(\text{Ca,Fe,Mn,Mg})\text{O}$ : PDF number-01-089-2468; Spinel,  $\text{Fe}(\text{Fe}_2\text{O}_4)$ : PDF number-01-084-6700; Gehlenite,  $\text{Ca}_2\text{Al}(\text{AlSiO}_7)$ : PDF number-01-073-6601; Perovskite,  $\text{CaTiO}_3$ : PDF number-00-042-0423; Brownmillerite  $\text{Ca}_2(\text{Al,Fe}_{+3})_2\text{O}_5$ : PDF number-00-030-0226;  $\text{Ca}_2(\text{Al,Fe}_{+3})_2\text{O}_5$ ; Bredigite  $\text{Ca}_7\text{Mg}(\text{SiO}_4)_4$ : PDF number 00-027-1060; Larnite  $\text{Ca}_2\text{SiO}_4$ : PDF number-00-033-0302; Merwinite  $\text{Ca}_3\text{Mg}(\text{SiO}_4)_2$ : PDF number-04-011-6738.

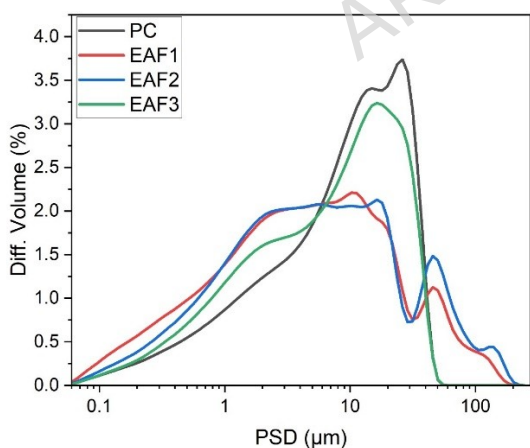


Figure 2: Particle size of PC and different EAF slags used in this study.

## 2.2. Experimental procedure

In this study, samples were prepared by mixing Portland Cement (PC) with 20% Electric Arc Furnace slag (EAF slag) at a water-to-cement ratio of 0.4. EAF slag samples were investigated using two distinct approaches: untreated and carbonation-pretreated. The carbonation pretreatment was conducted in a controlled environment with 20% CO<sub>2</sub> concentration, 85% relative humidity, and a temperature of 50°C for 24 hours. The thermal gravimetry of each EAF slag is illustrated in the appendix, showing the uptake of 20% carbonation with more effect on EAF1 and EAF2. The selection of 20% EAF slag incorporation into PC was substantiated by a comprehensive literature review, which consistently indicates an optimal replacement threshold for supplementary cementitious materials between 15-20% slag content [10][13][32]. A dosage of 0.1 wt.% organic ligands (Glycine [GLY] and Triisopropanolamine [TIPA]) was incorporated into different PC-EAF blends to investigate their effects on reactivity and hardening. Sample nomenclature was systematically developed to differentiate between control and ligand-incorporated specimens, with samples categorized into distinct series: sample names with Cab denote EAF carbonated, for instance, PC-EAF1 Cab denotes cement composite with carbonated EAF1, while PC-EAF1 Cab Gly denotes cement composite PC-EAF1 Cab with Gly as ligand.

Tables 2 delineate the comprehensive sample preparation protocol for this experimental investigation. For each mix, 450 g of raw materials were precisely weighed, comprising 360 g of PC and 90 g of EAF slag. The preparation procedure involved manual homogenization for 2 minutes to ensure uniform distribution of constituents, followed by mechanical mixing with water for 3 minutes to achieve optimal paste consistency. The resulting paste was processed into two distinct specimen geometries: 230 g was molded into cylindrical specimens for electrical impedance spectroscopy, with careful vibration to eliminate potential air entrainment. The remaining paste was cast into 2 × 2 cm cubic molds for compressive strength evaluation. All specimens were cured under standardized conditions with a constant relative humidity of 70% and a temperature of 21 ± 3°C. Mechanical characterization was systematically conducted at predetermined age intervals of 1, 3, 7, 14, and 28 days to enable comprehensive assessment of material performance evolution.

Table 2: Mix Design for Blended PC with Treated and Untreated EAF Slag and Ligands design

| Blended PC without ligand |        |          |              |          |              |              |          |           | Blended PC with ligand |             |                  |          |
|---------------------------|--------|----------|--------------|----------|--------------|--------------|----------|-----------|------------------------|-------------|------------------|----------|
| sample code               | CE M 1 | EAF1 [g] | EAF1 cab [g] | EAF2 [g] | EAF2 cab [g] | EAF3 cab [g] | EAF3 [g] | water [g] | sample code            | Glycine [g] | sample name      | TIPA [g] |
| PC                        | 450    |          |              |          |              |              |          | 180       | PC GLY                 | 0.45        | PC TIPA          | 0.45     |
| PC-EAF1                   | 360    | 90       |              |          |              |              |          | 180       | PC-EAF1 GLY            | 0.45        | PC-EAF1 TIPA     | 0.45     |
| PC-EAF1 Cab               | 360    |          | 90           |          |              |              |          | 180       | PC-EAF1 Cab GLY        | 0.45        | PC-EAF1 Cab TIPA | 0.45     |
| PC-EAF2                   | 360    |          |              | 90       |              |              |          | 180       | PC-EAF2 GLY            | 0.45        | PC-EAF2 TIPA     | 0.45     |
| PC-EAF2 Cab               | 360    |          |              |          | 90           |              |          | 180       | PC-EAF2 Cab GLY        | 0.45        | PC-EAF2 Cab TIPA | 0.45     |
| PC-EAF3                   | 360    |          |              |          |              | 90           |          | 180       | PC-EAF3 GLY            | 0.45        | PC-EAF3 TIPA     | 0.45     |
| PC-EAF3 Cab               | 360    |          |              |          |              |              | 90       | 180       | PC-EAF3 Cab GLY        | 0.45        | PC-EAF3 Cab TIPA | 0.45     |

## Characterization methods

### Setting time and compressive strength

The initial and final setting times were determined by applying 86 penetrations during the first 15 hr after mixing, using a Vicatronic apparatus (Matest, E044N) by adapting the EN 196-3 standard due to different water/binder ratios and room humidity. Compressive strengths for the paste samples were tested at 1, 3, 14, and 28 days, using a Zwick testing instrument with a maximum loading of 100 kN. and a loading capacity at a constant displacement rate of 1.8 mm/min. Four different samples were used, and the average strength was reported as compressive strength.

### Isothermal calorimetry and impedance spectroscopy

Isothermal calorimetry tests were conducted during the first 3 days after mixing using an 8-channel TAM Air isothermal calorimeter (TA Instruments) at 23°C. A plastic ampoule holder was used for samples and deionized water was used as a reference. The samples were first prepared by mixing for 3 min at 800–1000 rpm and then transferred to the plastic ampoule, which was then loaded into the calorimeter. Reported results were normalized to the CEM I (clinker) content of each sample to isolate the contribution of the reactive clinker fraction and allow direct comparison of hydration kinetics across blends with varying EAF slag content. Results were additionally adjusted by cutting the first 45 min after loading the sample, due to the time device adjustment before detecting the reaction.

ACIS device named VIONIC powered by INTELLO from Metrohm AutoLab was used to measure the electrical conductivity of blended cement with EAF. The paste as mentioned was moulded into a plastic mould of 6(diameter)\*12(height)mm with electrodes made of stainless steel embedded on both sides according to the model proposed by Provis and Sosa Gallardo [28] and Tchio et al [23]. The frequency range was 10MHz to 10Hz and the sinusoidal amplitude was 0.1V [33], [34]; during the first 24 hr, the measurement was carried out at 30min, 1h30min, 3, 6 and 24hr after that measurements were conducted at 3, 7, 28 days. To minimise the parasitic effects associated with the cell components and leads, the impedance of the cell in a short-circuit arrangement without a sample was measured before the samples were tested at each frequency and fitted with a circuit model. The impedance of the cell in a short-circuit arrangement was measured at each frequency prior to sample testing and vectorially subtracted from the sample impedance, effectively correcting for the inductive and resistive contributions of leads and connections, following the approach validated by Sosa Gallardo and Provis [28]. The equivalent circuit model illustrated in Appendix A was selected based on the accuracy of fitting using NOVA software [35] with the chosen model providing the best agreement with experimental data and the lowest fitting errors among the circuits evaluated. The equivalent circuit consists of  $R_s$ , representing the bulk ionic resistance of the pore solution; a parallel  $R_p$ –CPE combination, capturing the non-ideal electrode–paste interfacial polarisation arising from the heterogeneous and continuously evolving pore structure; and a capacitance  $C$ , accounting for the geometric capacitance of the stainless-steel electrode cell. The constant phase element (CPE) [33] replaces an ideal capacitor to represent distributed relaxation times within the cement paste microstructure, consistent with established practices in impedance modelling of cementitious materials [23]. The bulk resistance was obtained according to the parameter of the circuit model that was used to fit the impedance response at the high-frequency arch of the Nyquist plot by ACIS measurement [23][35]. Conductivity measurements were obtained from the resistivity of each cementitious paste by dividing the sum of the bulk resistance into a cell constant, which was obtained by measuring different concentrations of NaOH solutions of known conductivity [36].

#### *Phase assemblage*

At 1 and 28 days of hydration, the solvent exchange drying method was opted for stopping the hydration reaction. For this, the crushed samples (around 3–5 g) were dipped in isopropanol (60–100 ml) and stirred slowly for 20 min and filtered using 2–5  $\mu\text{m}$  particle retention Whatman paper. The filtered solids were then added to fresh isopropanol again and after 1 d filtered and dried. This procedure was carried out to stop hydration process prior to analysis. After the samples are completely dried under atmospheric air, they were ground into a fine powder using an agate mortar and pestle. The grinding time was uniform to all samples. The ground powder under 63  $\mu\text{m}$  of blended cement with EAF was then analysed by X-ray diffraction (XRD) using Rigaku SmartLab with

a Co source lamp of 40 kV and 135 mA. External standard ( $\text{SiO}_2$ ) was measured along the samples for the quantitative phase analysis. The phases were identified using HighScore software version 5.1 (Malvern PANalytical) with the ICDD PDF5+ 2024 database. The thermogravimetry analysis and differential thermogravimetry (TGA/DTG) analysis were carried out using a simultaneous thermal analyzer (SDT 650, TA instruments). Powder samples placed in platinum holder were heated from 30 to 1015°C at 10°C/min in  $\text{N}_2$  atmosphere.

### 3. Results

#### 3.1. Setting time and compressive strength

##### *Effect of carbonation treatment and ligands*

Blended cement samples exhibited varying setting times based on EAF slag content (Fig. 3a). Pure PC achieved initial and final setting at 3.6 h and 5 h, respectively. Non-carbonated EAF blends showed extended setting times, while carbonated EAF blends (EAF1, EAF2, EAF3) demonstrated significantly longer setting times compared to both PC and non-carbonated blends. This indicates carbonated EAF slag decelerates the setting, suggesting slower hydration and hardening processes, due to the low reaction kinetic activity of EAF slag or calcite and carbonate phase formation that modifies cement hydration mechanisms.

Compressive strength increased over time across all samples as shown in Fig. 3b. Both non-carbonated and carbonated EAF slag blends showed distinct strength development compared to PC. While initial strength was lower than the reference, significant increases occurred after 3 and 28 days. Carbonation significantly influenced strength development, with PC-EAF1I Cab, PC-EAF2 Cab, and PC-EAF3 Cab exhibiting higher strengths at 3, 7, 14, and 28 days compared to non-carbonated counterparts. PC-EAF1 Cab notably achieved 64 MPa at later stages, suggesting carbonation promotes enhanced later-strength development. EAF-EAF1 superior compressive strength among EAF slags likely stems from the higher CaO content in EAF1 which also showed higher heat release during the  $R^3$  test as reported by Adesanya et al, [31] which favor the formation of amorphous silicates that react in contact with cement pore solution.

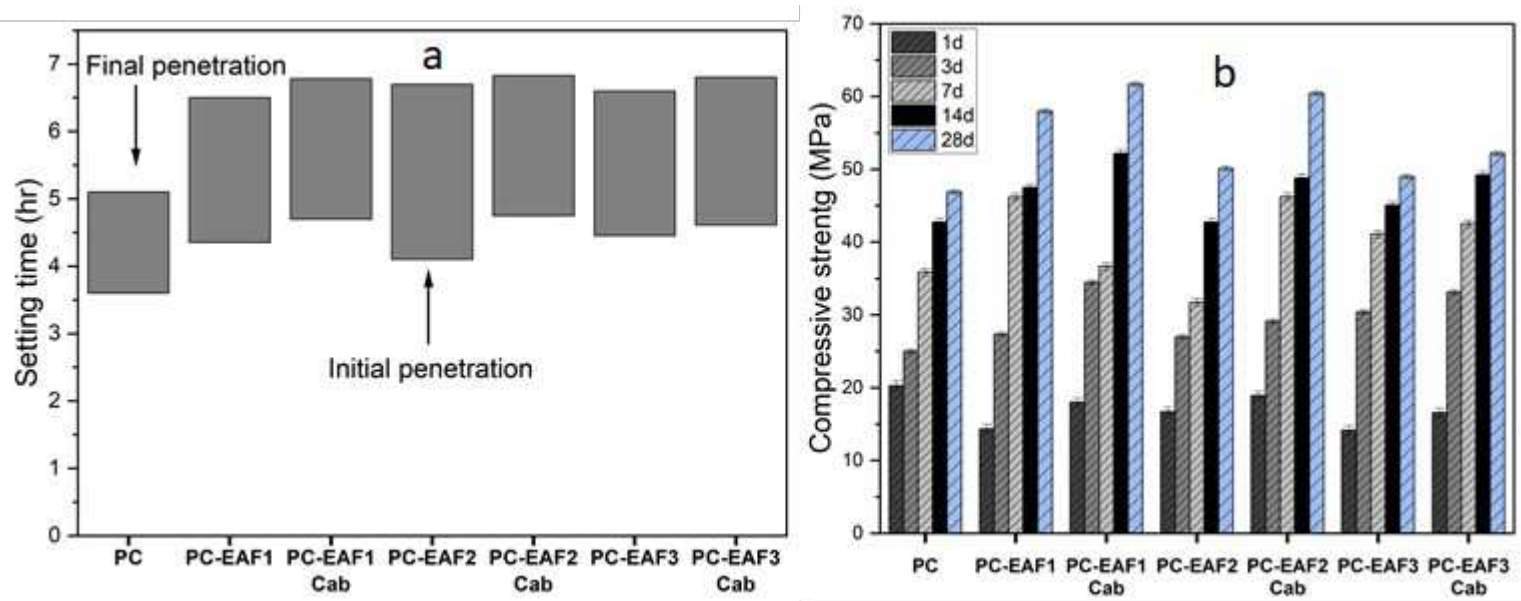


Figure 3: Setting time (3a) and compressive strength (3b) of blended cement

The addition of Gly and TIPA ligands influenced initial and final setting times in cement and blended systems (Fig. 4) through variations in penetration depth. Both ligands accelerated initial setting times to 3-3.25 hr in PC and non-carbonated EAF blends (Fig. 4a), while carbonated EAF slag blends showed 4hr initial setting (Fig. 4b) compared to 5hr in ligand-free blends (Fig.3a). Final setting times averaged 6.25-6.5hr across all samples except PC-Gly and PC-TIPA. In the literature, it has been noted that adding Gly ligand accelerated the initial setting time in the Portland system and extended the final [37], this seems to be the case with the addition of carbonated and non-carbonated blended EAF slag in PC system, similar trend with the TIPA ligand but with a shorter final setting time [38]. However, the same mechanism for setting time acceleration was observed in this study despite the inclusion of non-carbonated and carbonated EAF slag. Compressive strength increased over time with ligand addition (Fig. 4c and 4d), with TIPA showing greater enhancement in a blended system compared to Gly. PC with either ligand maintained slightly higher strength than blended systems from 1-14 days, except for PC-EAF1, PC-EAF1 Cab-TIPA, which achieved comparable or higher 28-day strengths.

Both ligands decreased early strength in non-carbonated EAF slag blends (<20 MPa at 1 day, <40 MPa at 3 days) compared to ligand-free blends. However, carbonated EAF slag blends with ligands achieved >20 MPa at 1 day and >30 MPa at 3 days, indicating distinct hydration mechanisms between carbonated and non-carbonated systems with ligands.

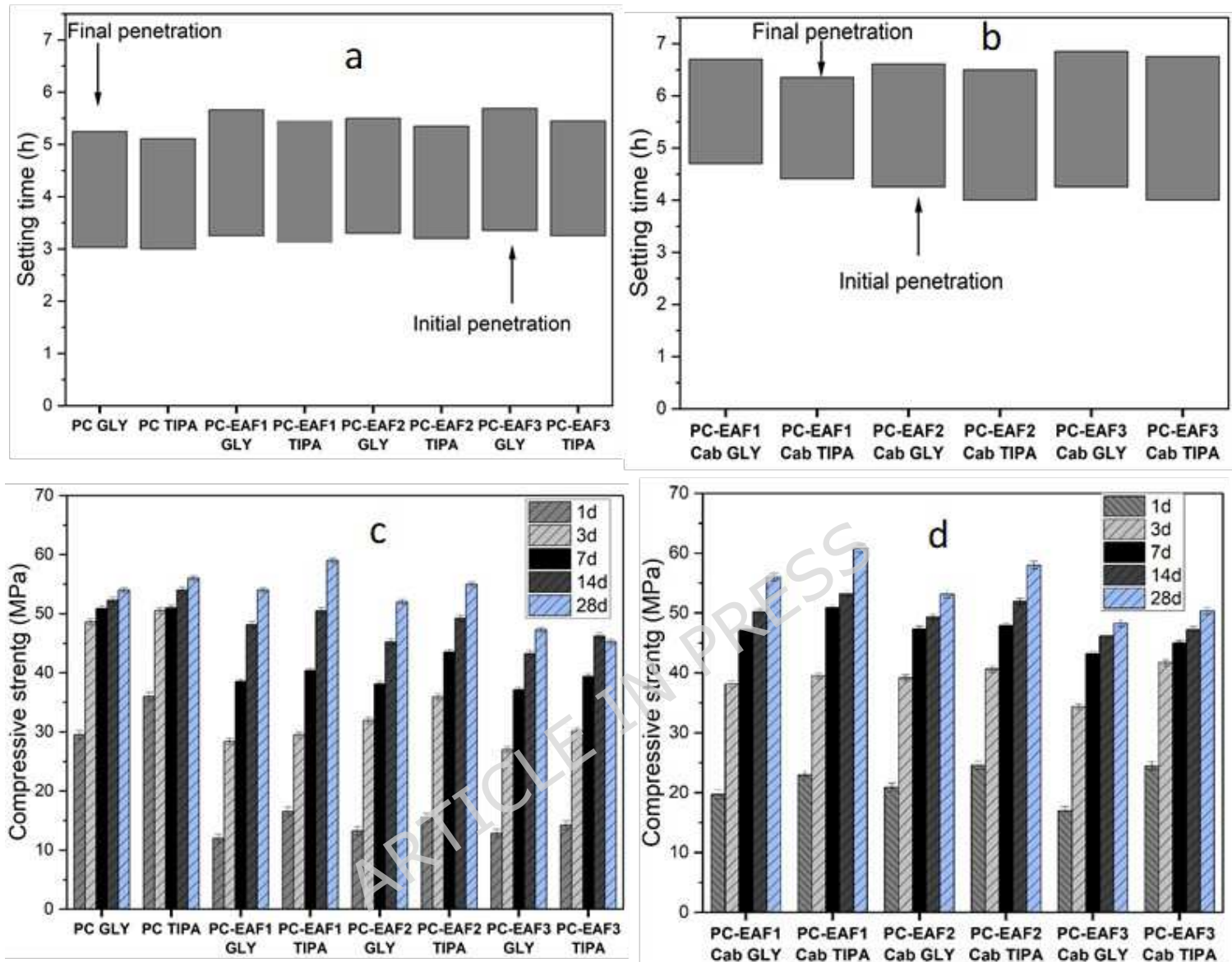


Figure 4: Setting time and compressive strength after including TIPA and Gly ligands.

### 3.2. Hydration kinetics

#### Effect of carbonation treatment and ligands

The heat flow and cumulative heat release of blended PC with untreated and pretreated EAF slags (Figs. 5–8) were normalized to the CEM I content of each blend rather than to the total binder mass. This choice is justified by the low intrinsic hydraulic reactivity of EAF slag at early age: normalizing by total binder mass would dilute the heat signal proportionally to the 20 wt.% substitution level,

masking differences in clinker hydration kinetics between mixes, whereas normalization to CEM I content isolates the clinker contribution and ensures direct comparability across all blends. It should be noted that total binder normalization would uniformly reduce heat values by a factor of 0.8 without altering any trend, ranking, or conclusion.

In non-carbonated EAF blend systems (Fig. 5), PC-EAF1 and PC-EAF3 exhibited higher accelerated initial hydration peaks during 12-15hr compared to the control PC 24hr peak, this peak is attributed to the formation of the main hydration product in cement (C-S-H) which may indicate higher formation of C-S-H in the composite cement. PC-EAF2 showed delayed but sharp hydration peaks at 18hr, with significantly lower heat release than PC-EAF1 and PC-EAF3, indicating distinct hydration kinetics and lower formation of C-S-H phases. In carbonated EAF blend systems, blended PC with carbonated EAF slag (PC-EAF1 Cab, PC-EAF2 Cab, and PC-EAF3 Cab) demonstrated uniform heat flow patterns, characterized by accelerated, synchronous peaks around 15hr. These carbonated EAF blend systems displayed more uniform and synchronized peak intensities among themselves compared to non-carbonated counterparts not in comparison to PC, whose main hydration peak occurs later (24 hr) suggesting that carbonation of calcium silicate phases homogenizes the reactive phase assemblage across different EAF slag sources. Specifically, the conversion of free CaO and MgO into  $\text{CaCO}_3$  and amorphous silicate during carbonation results in a more consistent reactive surface available to participate in hydration, regardless of the original slag composition, explaining the convergent heat flow behaviour observed across PC-EAF1 Cab, PC-EAF2 Cab, and PC-EAF3 Cab. Heat release patterns in carbonated systems showed similar evolution during the first 50h, contrasting with non-carbonated EAF blend distinct peak intensity, it can then appear that the mechanism that leads to the carbonation of EAF slag where the capture of  $\text{CO}_2$  by  $\text{C}_2\text{S}$ , free CaO, and MgO to form  $\text{CaCO}_3$  and  $\text{MgCO}_3$  follow the same process whatever the type of slag and consequently improve in the same reaction the volumetric stability of the EAF slag. This similarity trend in carbonated blend suggests that amorphous silicates,  $\text{CaCO}_3$  and potentially  $\text{MgCO}_3$  in EAF slag present a closer reaction mechanism for the formation of phase product. However, when comparing our results to those of older studies, it can be pointed out that carbonation can further promote the hydration of the steel slag phase due to further increase in  $\text{CaCO}_3$ ,  $\text{MgCO}_3$  that are more beneficial to react [14] [39], [40].

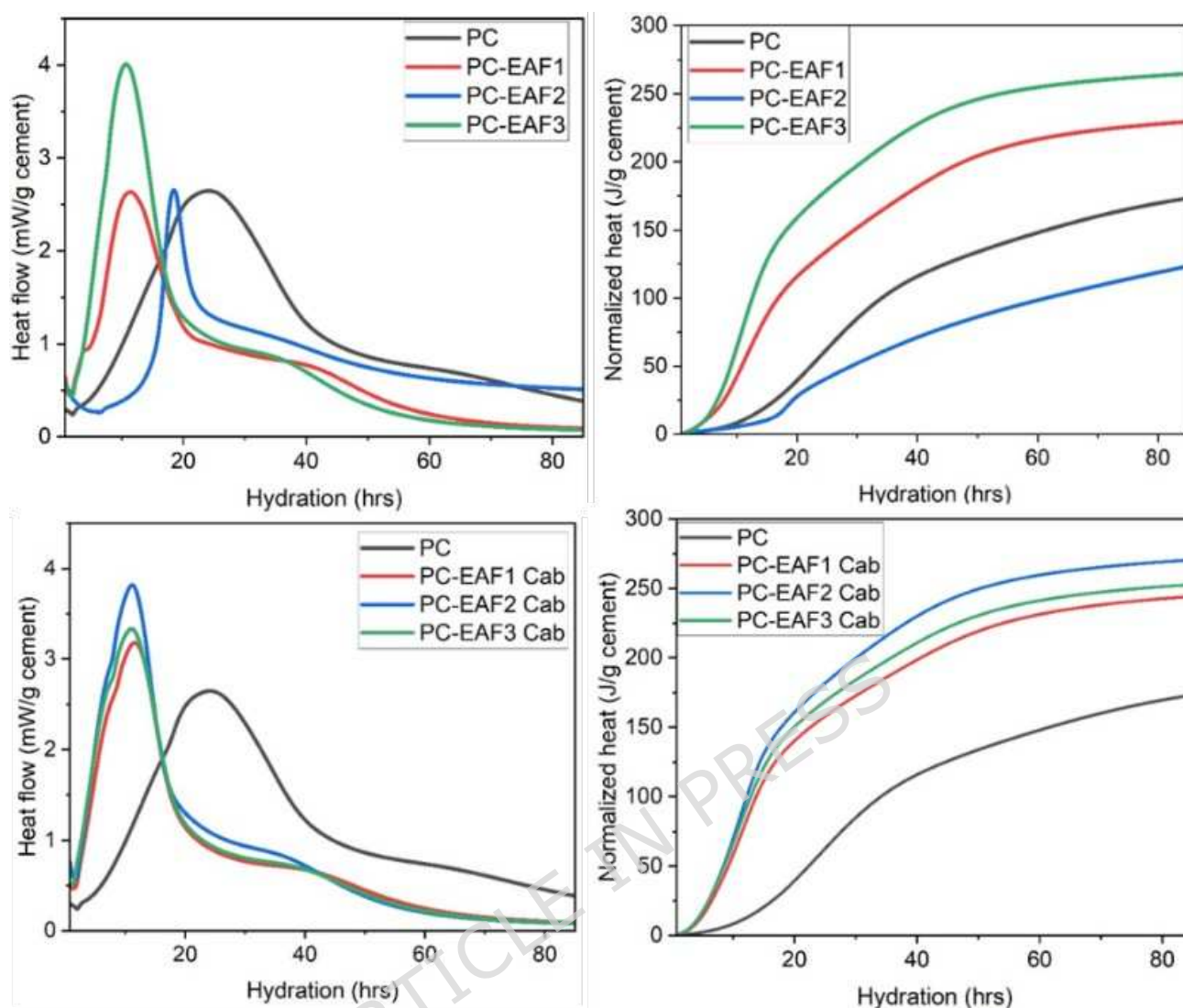


Figure 5: Heat flow and heat release of blended cement with carbonated and non-carbonated EAF slags.

When incorporating Gly and TIPA ligands into the PC system (Fig. 6), both accelerated the main hydration kinetics peaks (8-10hr) but exhibited distinct reaction kinetics. Gly-modified PC showed moderate peak intensity ( $\sim 3.3$  mW/g), while TIPA-modified PC demonstrated the highest peak intensity ( $\sim 6.7$  mW/g) during early hydration. Pure PC displayed delayed and broader peaks with lower heat flow intensity centred around 36 hr. In blended cement systems with non-carbonated and carbonated EAF slags, Gly and TIPA ligands exhibited distinct heat flow patterns like PC ligand-modified systems. Gly-modified (Fig.7a), non-carbonated EAF blends showed uniform hydration processes with similar peak intensities around 16hr. However, PC-EAF1 Gly illustrated lower exothermic peaks around 16hr and a more pronounced peak at 40hr of hydration. In contrast to Gly, TIPA incorporation significantly altered hydration (Fig. 7b), producing higher peak intensities comparable to TIPA-modified PC, with varying peak timing among EAF sources. PC-EAF1 TIPA exhibited an early sharp peak followed by secondary hydration, correlating with strength results (Fig.

4c and 4d). Higher TIPA-induced heat flow corresponded to acceleration of setting times, while lower intensities indicated extended setting times (Fig. 4a, and 4b). Carbonated systems with Gly ligands demonstrated delayed hydration kinetics (Fig. 7c), characterised by delayed main exothermic peaks, reduced peak heat flow intensities ( $\sim 3$  mW/g), and uniform behaviour across all three EAF slag sources compared to non-carbonated counterparts. TIPA-modified carbonated samples maintained high peak intensities with synchronized hydration timing (Fig. 7d), suggesting that the effect of direct carbonation of EAF slag before incorporation in PC influences the mechanism of ligand and extending the reaction kinetics in blended cement systems. Both Gly and TIPA-modified carbonated systems showed delayed main hydration peaks compared to PC-Gly and PC-TIPA, corresponding to extended final setting times.

As a general observation from the results, one exothermic peak and one exothermic shoulder were found. For instance, in both non-carbonated and carbonated EAF blends, the intensity of the first peak reflects the dissolution of various mineral phases of the composite material upon water contact. While Aft precipitation is known to occur during this early period in PC systems [41], no direct experimental evidence such as in situ XRD or microscopy was obtained in this study to confirm Aft formation at this stage; this remains consistent with the literature but should not be inferred directly from the calorimetric data alone [42]. However, the intensity of the first peak also represents the hydration heat peak attributed to the dissolution of  $C_3A$  and the initial dissolution of  $C_2S$ ,  $C_4AF$ , and  $C_2F$ , which is reflected by the precipitation of CH, CASH, and Aft. Furthermore, the exothermic shoulder arises from the continued dissolution of  $C_2S$ ,  $C_4AF$ ,  $C_2F$ ,  $CaCO_3$ , and  $MgCO_3$ , with continued precipitation of C-S-H and AFm [22], [43]. It can be assumed that the dissolution of phases like  $C_3A$ ,  $C_{12}A_7$ , and part of  $C_4AF$ , aided by TIPA, is responsible for the high additional intensity peak observed in PC-EAF1 TIPA, along with the rapid interaction with gypsum to form the Aft phase [37]. Furthermore, this Aft phase could be responsible for the delay in the hydration of the EAF blend due to the encapsulation of Aft at the surface of EAF1 blended particles, leading to a reduction in the hydration of mineral phases within particles, manifested as delayed hydration peaks and lower hydration peaks after 11hr [22]. In both non-carbonated and carbonated EAF blends, TIPA shows a high main sharp exothermic peak and a reduction in the area of the second exothermic peak around 40 hr, while Gly shows a low main exothermic peak and a higher area of the second exothermic peak at 40hr.

Total heat release patterns (Fig. 8) revealed variations in reaction kinetics among blended ligand systems with and without carbonation. In ligand-PC systems, PC-TIPA showed significantly higher heat release than PC-Gly and pure PC, correlating with early-age strength. However, blended cement systems with non-carbonated and carbonated slag demonstrated higher heat release than PC-Gly, while PC-TIPA exhibited higher early-age compressive strength, suggesting no direct correlation between heat release and compressive strength, as noted by Lothenbach et al. [44].

Variations in strength primarily correlate with microstructural development, pore structure densification, and reaction product formation. TIPA ligands may enhance product formation due to higher heat release compared to Gly ligands. The diverse heat flow and heat release patterns observed in calorimetry curves indicate varying reactivity and accessibility across different EAF slags, resulting in distinct interactions with ligands. Similarly, carbonated EAF slag reactivity and ligand interactions appear to follow comparable patterns regarding reactive phase accessibility and reactivity. However, the dissolution properties are beneficial to the compressive strength development of the EAF blended cementitious materials.

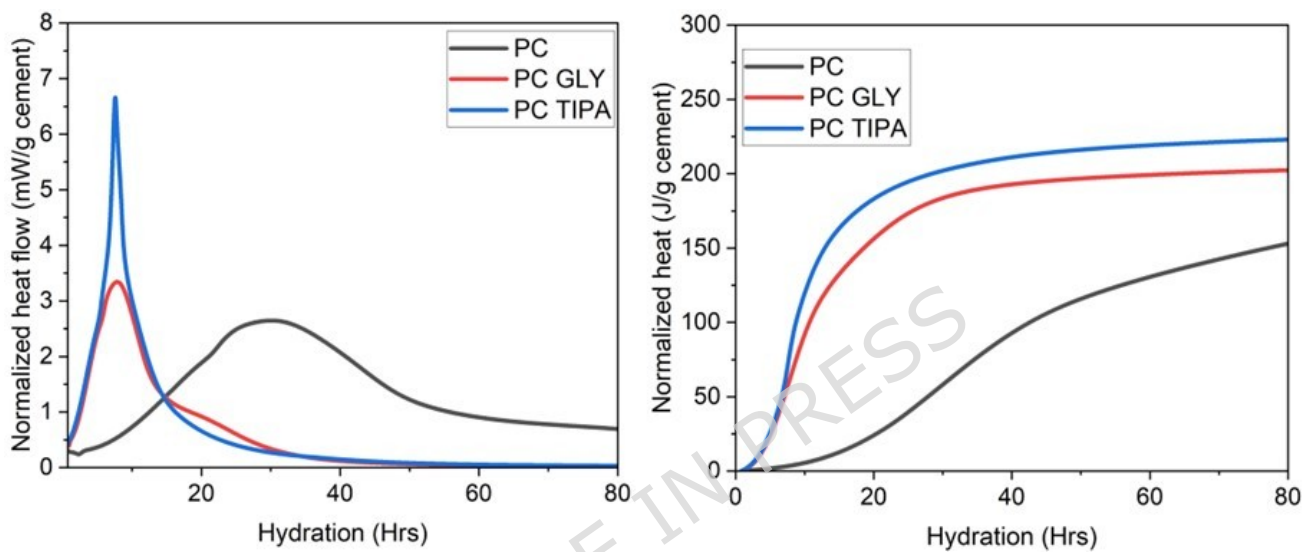


Figure 6: Influence of Gly and TIPA on the reaction kinetics of PC

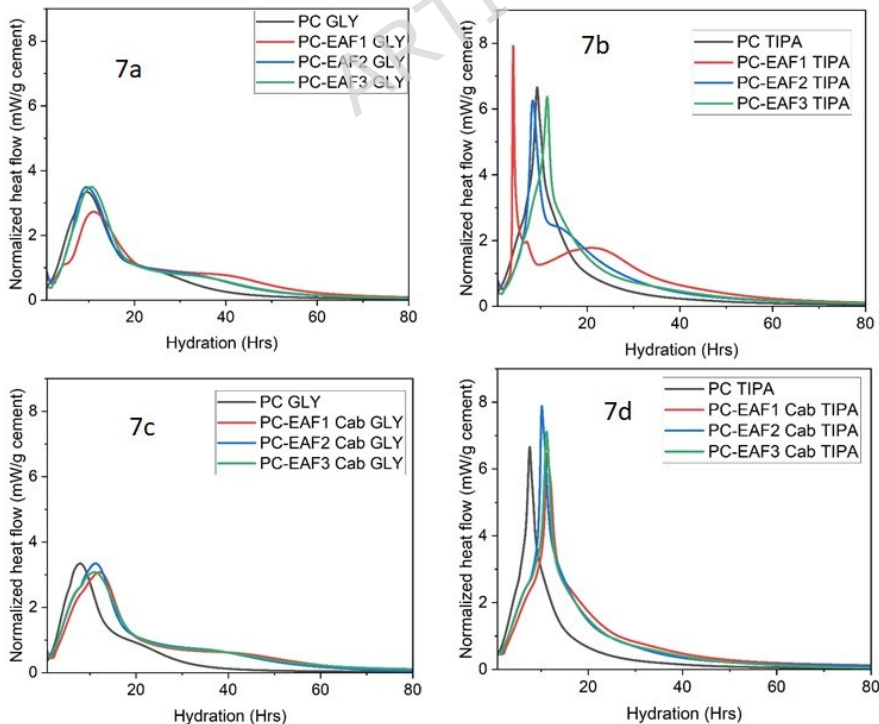


Figure 7: Heat flow of non-carbonated and carbonated EAF blend in PC (a: Gly non-carbonated EAF blend, b: Tipa non-carbonated EAF blend; c: Gly carbonated EAF blend; d: Tipa carbonated EAF blend).

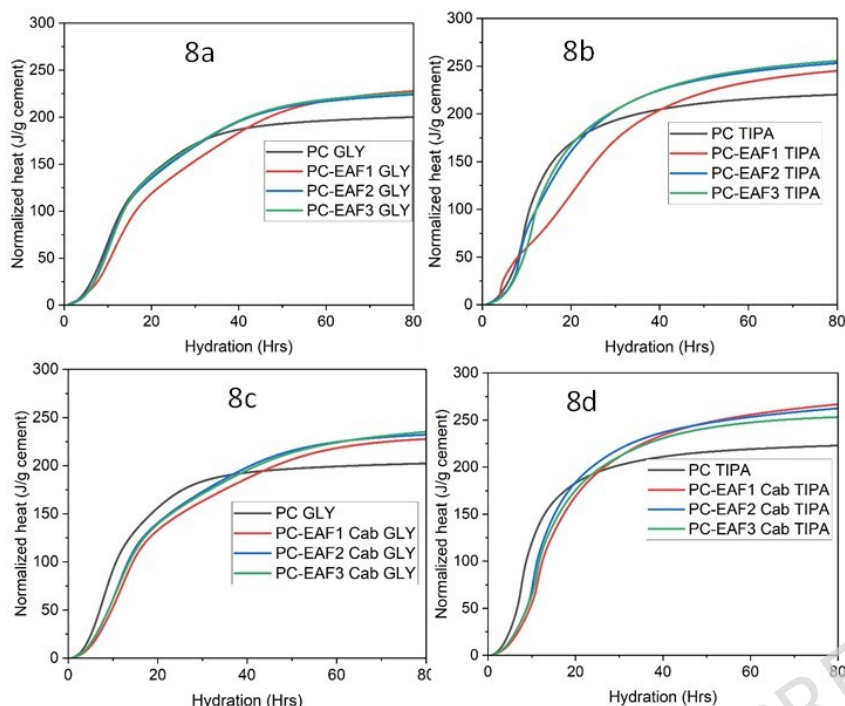


Figure 8: Cumulative heat of non-carbonated and carbonated EAF blend in PC (a: Gly non-carbonated EAF blend, b: Tipa non-carbonated EAF blend; c: Gly carbonated EAF blend; d: Tipa carbonated EAF blend).

### 3.3. Impedance spectroscopy

Electrical conductivity measurements over time (0.5 hr to 28 days) for various cement compositions, including control systems and their carbonated/non-carbonated variants are illustrated in Fig. 9. Pure PC systems exhibited higher initial electrical conductivity (1.1-1.15 S/m) from 0.5 to 3 hr, attributable to elevated ionic concentrations in the pore solution during the dissolution stage compared to EAF slag blends where the crystalline minerals of the slag are responsible for the delay in chemical activity, reducing the concentration of ions in the electrolytic pore solution. On the other hand, carbonated EAF blends showed lower initial conductivity (0.8-0.9 S/m) than non-carbonated blends, indicating there are fewer ions released from carbonated minerals compared to  $C_2S$ .

During the first 3 hr, samples demonstrated minor conductivity decrease which indicate only minor changes in ionic conductivity during this time. However, between 3 and 6 hr PC sample showed clear drop in conductivity, indicating formation of reaction products, which is in line with setting time and calorimetry results. Conversely, samples containing carbonated and non-carbonated EAF slag showed higher conductivity at 6 hr compared to control PC, which matches with the slower hardening

of these samples and illustrating higher ion mobility at this time. Varying conductivity patterns among carbonated EAF slag blended samples indicate differences in the pore solution chemistry. The differences in the electrical responses are small for instance, at 6h the conductivity in PC-EAF1-Cab and PC-EAF2-Cab are 0.76264 and 0.78352 S/m , but should be relevant as based on our previous study [23] the average error between three similar measurement was around 0.015 S/cm during the first 1 day. Between 3-6hr, all samples exhibited decreasing conductivity, marking the nucleation phase characterized by paste densification and reduced pore connectivity. This stage involves ettringite formation, CH crystallization, and C-S-H precipitation, corresponding to initial and final setting times (Fig. 4a) [26]. It would be important to note that the formation of cementitious phases reduced pore volume, restricting ionic mobility and decreasing conductivity [23].

The 6-24 hr period showed significant conductivity decrease across all systems, indicating intensive hydration with extensive reaction product formation. This led to denser microstructure and increased bulk resistance. Carbonated and non-carbonated samples maintained higher conductivity values, suggesting less dense microstructure and different pore solution chemistry, with sustained ionic mobility compared to PC. This pattern aligns with findings from Nwaubani et al., [45] who demonstrated that partial PC substitution with EAF slag results in slower initial reaction product formation, followed by enhanced hydration product development at later ages. Similarly, Muhood et al. [10] confirmed that EAF slag reactivity in cement predominantly manifests at later stages. From 24 hr to 28 days, all systems exhibited stabilized conductivity (approximately 0.05-0.1 S/m) with minimal variations, indicating the development of a stable, compact microstructure characterized by reduced pore connectivity.

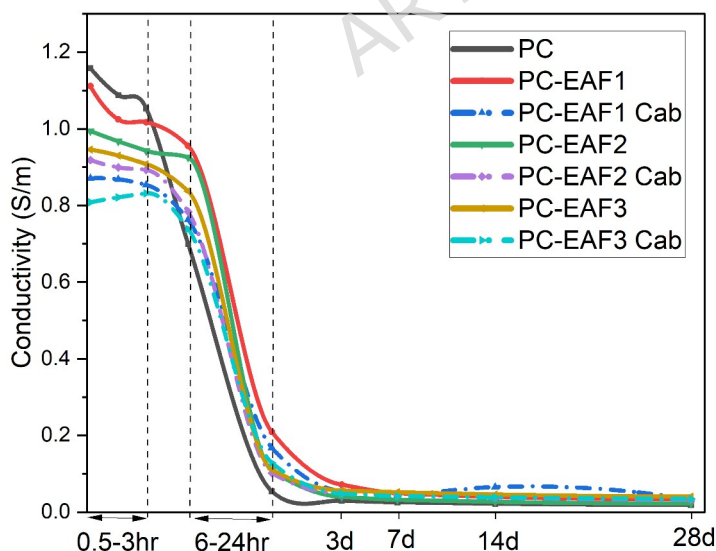


Figure 9: Electrical conductivity of PC and blended PC with non-carbonated and carbonated EAF slags.

The conductivity measurements with Gly and TIPA ligands revealed the modification of electrical conductivity patterns by altering the hydration mechanism. In PC system, both ligands exhibited initial conductivity values of 1.15-1.2 S/m, as illustrated in Fig. 10a and 10b, with TIPA showing higher values during early hydration (0.5-3hr). During the first 3 hr, similar steadily decreasing conductivity trends were observed with and without ligands in PC systems. This suggests that while organic ligands increased dissolution of ions as indicated by higher conductivity, these ions participated in product phase formation.

In EAF slag blends (PC-EAF1, PC-EAF2, PC-EAF3), Gly-containing samples displayed higher initial conductivity (0.95-1.07 S/m Fig.10a) compared to TIPA-containing blends (0.9-1.0 S/m Fig.10b). This indicates that Gly ligand may promote higher dissolution in non-carbonated EAF slag system during the first 3h. In blended cement systems with carbonated EAF slag, both ligands showed a progressive increase in conductivity during the first 3h, exceeding values observed in ligand-free blends. While ligand presence did not prevent conductivity decrease during 3-6 hr, the reduction pattern differed from ligand-free systems, indicating an influence of ligands on ion mobility and hydration processes. A pronounced conductivity decrease occurred between 6-24 hr in all systems, indicating substantial hydration product formation. Conductivity measurements from 24 hr to 28 days showed a continuous decrease, with higher conductivity detected in both carbonated and non-carbonated samples containing either ligand compared to pure PC systems, suggesting enhanced ionic mobility in blended systems. The PC substitution with carbonated and non-carbonated EAF-EAF3 maintained the highest long-term conductivity with both ligands, which appears contradictory to strength values. This discrepancy may be explained by the fact that compressive strength primarily depends on porosity, while conductivity reflects pore connectivity. Furthermore, the low reactivity of EAF3 as illustrated on the R3 test may potentially be the cause of this observation. TIPA-containing samples consistently demonstrated higher conductivity values compared to Gly-containing samples in later stages, particularly in carbonated systems, indicating TIPA's effectiveness in maintaining ionic mobility within the pore network during microstructural development. This may be due to TIPA's ability to influence reactivity of iron-rich phases (RO phase) and also  $C_2S$  as observed in earlier studies [23] thus maintaining higher conductivity.

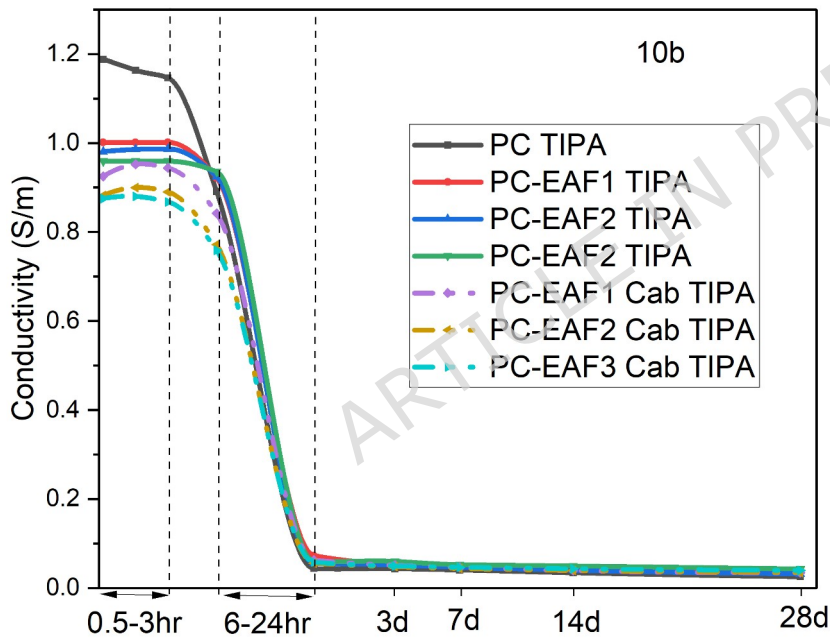
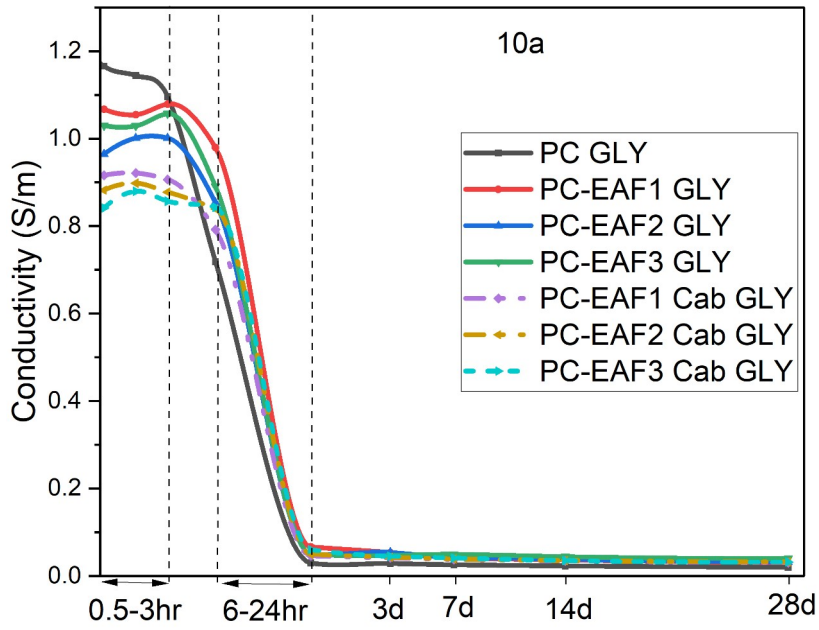


Figure 10: Electrical conductivity of (10a) Gly-modified blended PC with non-carbonated and carbonated EAF slag; (10b) Tipa-modified blended PC with non-carbonated and carbonated EAF slag).

### 3.4. Phase assemblage analysis

DTG was employed to analyze the hydration products of the samples after 28 days curing. Samples were chosen based on strength results and trends in isothermal calorimetry as reported in Fig. 3b, 4 and 7. As seen in Fig. 11, all samples revealed three main thermal weight loss intervals: 60-200 °C, 400-460 °C, and 600-750 °C. The first range of weight loss at 60-200 °C is characteristic of the

loss of C-S-H, AFm, and AFt. The peak at 400-460°C is consistent with the dehydroxylation of CH, and the final peaks at 600-750°C is assigned to the decarbonation of CaCO<sub>3</sub>. As illustrated in Fig. 11a, after 28 days of hydration, the thermal weight loss of PC-Gly and PC-TIPA at 60-200°C was higher than that of PC, suggesting that PC Gly and PC-TIPA generate more C-S-H and AFt, which is probable to contribute to the strength. It can also be observed that in the presence of TIPA, in the PC samples, the formation of AFm can be observed which will furthermore improve the strength. Triethanolamine, which is in the same family as TIPA has been shown to increase the dissolution of alumina and iron-rich phases to form AFms or hydrogarnets [22], which could be the case in this system. When blending PC with EAF1 slag, carbonated and non-carbonated (Fig. 11b), without the ligands, PC-EAF1 and PC-EAF1 Cab show higher thermal weight loss of CSH and AFt compared to PC control. However, in the presence of TIPA or Gly (Fig. 11c and Fig. 11d), the decomposition peak assigned to C-S-H and AFt increases and is comparable with PC Gly and PC-TIPA.

The amount of CH in the range of 400-460°C can reflect the degree of cement hydration. The thermal weight loss of PC is more pronounced than when organic ligands are included in PC. Nevertheless, when blending PC with carbonated and non-carbonated EAF1 slags, the proportion of CH increases but not more than in a pure PC system because supplementary cementitious material will consume CH to form C-S-H, moreover, higher CH is formed due to higher dissolved alite (C<sub>3</sub>S) contained in PC compared with substituted PC. With the addition of TIPA, lower weight loss of CH is observed than when glycine is added, illustrating their different influence on the hydration process. TIPA can form complexes with dissolved ions from C<sub>4</sub>AF and thus increasing its dissolution, which will provide more Ca in pore solution to form CH, and Al and Fe to form AFt and AFm.

In the 600-750°C range, the thermal weight loss peak intensity of all samples with ligands were lower than that of PC. The higher CaCO<sub>3</sub> content in the PC reference paste is attributed to its greater clinker content relative to EAF slag blended systems. Hydration of C<sub>3</sub>S and C<sub>2</sub>S produces portlandite (CH), the primary hydration product susceptible to atmospheric CO<sub>2</sub> uptake and conversion to CaCO<sub>3</sub> [46]. The 20 wt.% clinker displacement by EAF slag reduces CH yield proportionally, resulting in lower CaCO<sub>3</sub> content in blended systems, consistent with the established dependence of CO<sub>2</sub> binding capacity on total available CaO [42]. Additionally, it appears that the addition of ligands in the blended PC with carbonated and non-carbonated EAF slag strengthens the peak at 60-200°C while weakening the peaks at 400-460°C and 600-750°C. This suggests that TIPA and Gly enhance the consumption of CH and CaCO<sub>3</sub>, leading to the formation of more C-S-H and AFt phases.

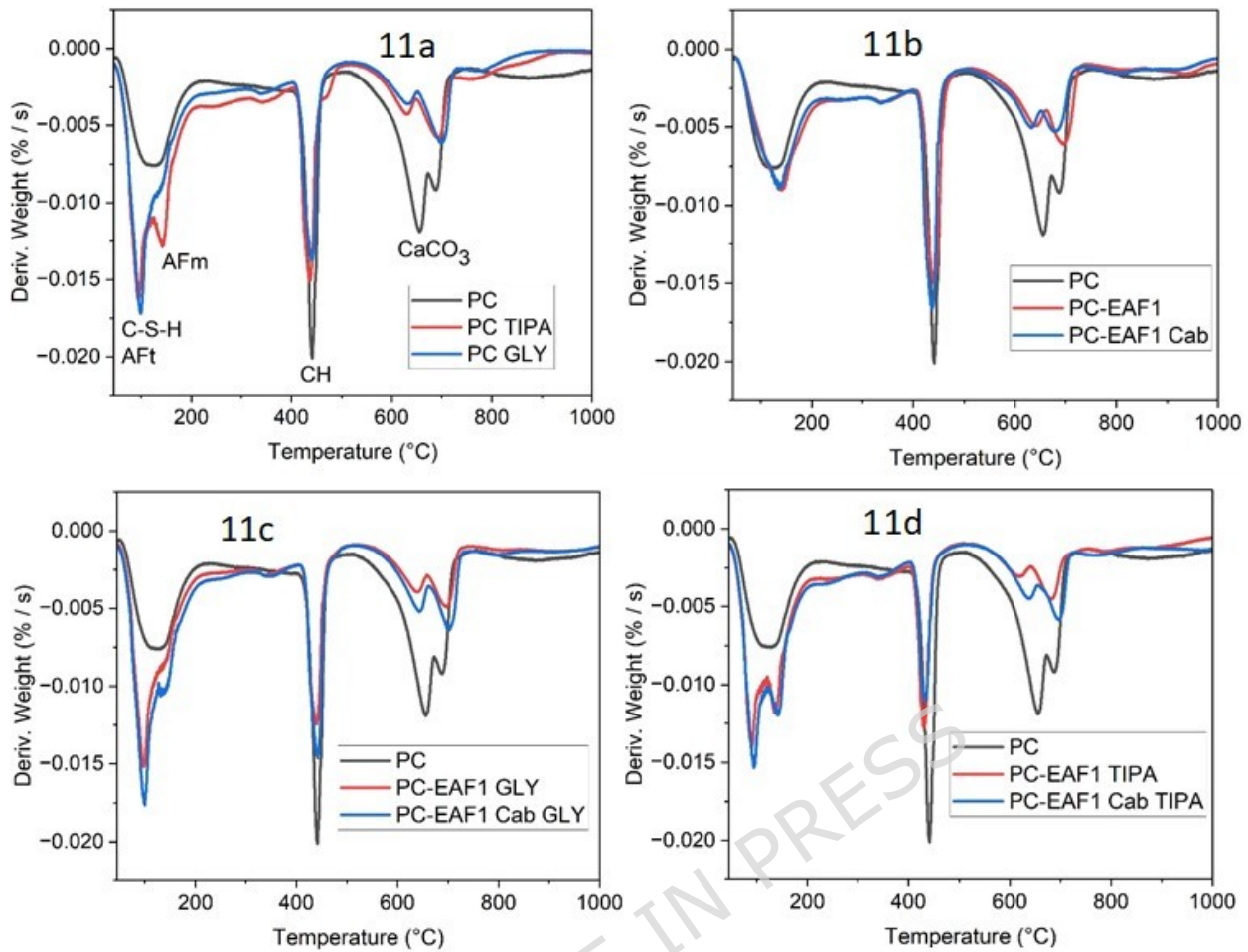


Figure 11: Thermogravimetry data; 11a: PC modified system with TIPA and Gly ligands, 11b: blended PC with non-carbonated and carbonated EAF1 slag, 11c: Gly-modified blended PC with non-carbonated and carbonated EAF1 slag, 11d: TIPA-modified blended PC with non-carbonated and carbonated EAF1 slag.

X-ray diffractograms of 28 days hydrated samples revealed notable changes in the signal intensities, as shown in Fig.12. It was observed that the peak intensities of  $C_3S$ ,  $C_2S$  and  $CaCO_3$  slightly decreased when TIPA and Gly were added to PC. However, these peak intensity reductions were more pronounced in the non-carbonated EAF blend and the carbonated EAF blend, although when ligands are included in the blended cement system, there seems to be a higher peak intensity for  $C_2S$  and  $C_3S$ . Interestingly, the blended PC-EAF1 and PC-EAF1 cab samples diffractogram shows complete disappearance of ettringite (AFt) peaks, which were present in all other samples at  $10^\circ 2\theta$ , revealing that AFt has been transformed to carboaluminates (AFm) on reaction with  $CaCO_3$  [47]. Nevertheless, the CH peak intensity sequentially decreases with the inclusion of TIPA and Gly in the paste. However, blending PC with EAF1, both carbonated and non-carbonated, unexpectedly increased the CH peak intensity, which correlates with the thermogravimetry results shown in Fig. 11. In the blended system with ligands, the CH peak intensity decreases more with TIPA and less with Gly. These observations suggest that TIPA and Gly potentially enhance the hydration process

in both pure PC and blended PC-EAF1 carbonated composite cement, with TIPA showing a more pronounced effect on promoting dissolution, aligning with the strength results in Fig. 4.

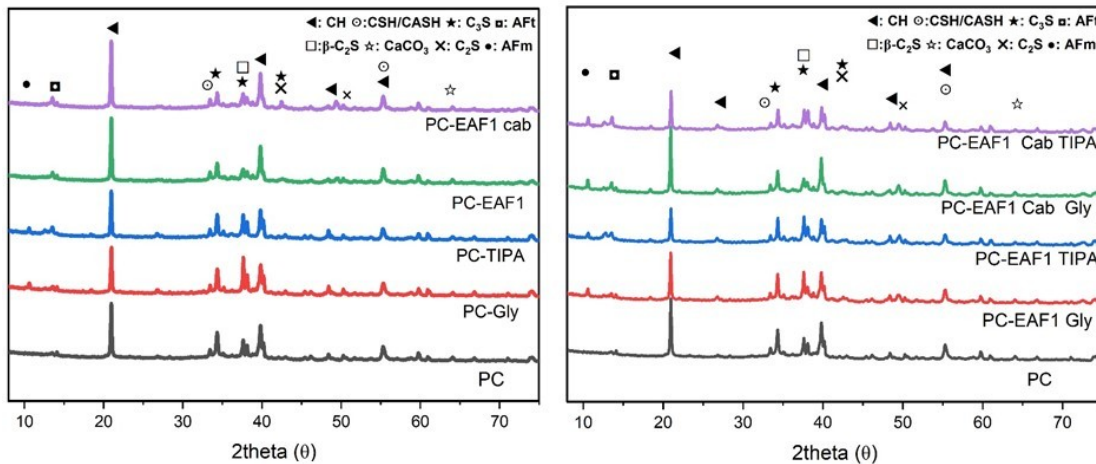


Figure 12: X-ray diffractograms of the samples after 28 days of curing.

#### 4. Discussion

##### EAF slag, TIPA and Gly ligands influence the hydration process of blended PC samples

The addition of carbonated and non-carbonated EAF slag in PC alters the mechanical properties and degree of hydration of PC. A decrease in electrical conductivity during the first three hours of hydration is observed as demonstrated in Fig. 9 which is likely related to mobility of ions in fresh pastes. The release of Ca and Mg ions from EAF slag modifies the hydration kinetics of clinker phases; specifically, it appears to hinder the hydration of  $C_2S$ ,  $C_3S$ , and  $C_4AF$  while promoting  $C_3A$  reactivity, which subsequently influences the nucleation and growth of CH and C-S-H [48],[49], [50]. This observation confirms that non-carbonated and carbonated EAF slag blends exert substantial dilution and electrical insulating effects during paste microstructure development, causing electrical conductivity reduction. Also a prolonged setting time with more effect in the carbonated system with ageing (from 6 hours to 24 hours) during hydration was observed. This align with the work of Luckman et al. [51] who reported that the reaction kinetics of steel slag starts between 2 and 7 days and show a measurable increase between 7 and 28 days. This later-stage reaction led to a progressive increase of compressive strength in EAF slag blends, with more effect in carbonated EAF blend. Comparative analysis indicates that the addition of non-carbonated EAF slag to the cementitious matrix leads to a less pronounced reduction in electrical conductivity compared to carbonated EAF slag. The carbonation of  $C_2S$  to  $CaCO_3$  and its volumetric stability during carbonation [39] modify their dissolution dynamics during the hydration process with less charge-

transfer ion resistance in electrical conductivity, ultimately producing a consistent peak heat flow across different EAF slag types, while a different trend is noted in non-carbonated EAF blend.

The addition of organic ligands TIPA and Gly modifies the hydration reaction across various cementitious systems. The compressive strength of PC was only 20 MPa at 1 day; while adding ligands, the compressive strength increased to 30 MPa with Gly ligand and 36 MPa with TIPA ligand. In contrast, for non-carbonated EAF slag blends, ligand addition resulted in a reduction in strength compared to the ligand-free system, although TIPA showed higher strength than Gly. Both ligands exhibited higher strength and pronounced effects in carbonated EAF blends. Based on the hydration heat, TIPA-containing samples exhibit higher heat flow, attributed to its capacity to promote the dissolution of key phases such as  $C_4AF$  and  $C_3A$  and accelerate the consumption of gypsum to form AFt and AFm phase enhancing strength gain. The higher conductivity observed in the PC system during the first 24 hours is consistent with literature reports indicating that TIPA increases the concentration of Ca, Al, Fe, ions in the pore solution through its known complexation capability [52]–[54]. This ion enrichment of the pore solution supports the observation where higher ionic mobility at early age correlates with accelerated hydration product formation [54].

However, when blending PC with carbonated/non-carbonated EAF slag, conductivity becomes less directly correlated with strength and potentially indicating the influence of other factors such as complexation ability and possible lower mobility of complexed ions. The molecular structural differences between TIPA and Gly significantly influence their interaction with ions in PC and carbonated/non-carbonated EAF slag systems, leading to distinct dispersion and hydration behaviors. Research by multiple investigators has demonstrated that TIPA present superior complexation capacity, conferred by its multiple hydroxyl groups forming stable coordination complex with metal cations [52], [54]. For instance, it could be that while TIPA can promote dissolution of the mineral phases of the blended system, but it would have lower mobility due to the formation of ion pairs and complexes that reduce the charge transport capability despite the presence of ions in pore solution system. For example, Wang et al., [52] reported that complexation of TIPA on the surface of steel slag which promoted dissolution. Furthermore, TIPA ligand can be adsorbed onto the surface of EAF slag and PC cement particles due to the hydrogen bonding from TIPA ligands, potentially creating a repulsive force due to the steric hindrance of the bulky isopropanol group [23]. Conversely, Gly, with its simplified molecular structure and greater hydrophilicity due to its amino and carboxyl functional groups, facilitates water interaction but provides less delay on the hydration. Showing that higher hydrophilic does not mean promoting cement hydration, revealing that other factors like dissolution, interaction with metal ions, and phases formation need to be taken into account. Furthermore, the repulsive force generated in the Gly system is an electrostatic interaction, which may not be effective in achieving enhanced connectivity at a later stage through the repulsion effect and low steric hindrance [55].

In blended PC systems with carbonated EAF slag, both TIPA and Gly delayed early hydration between EAF slag and gypsum, as shown by a shift in the main calorimetric peak and reduced conductivity. The formation of carbonate compounds during carbonation seem to modify the dissolution mechanism and ligands interaction regardless the type of EAF slag. Ligands could facilitate improved diffusion of reactants, thereby promoting long-term precipitation product formation. Furthermore, TIPA ability to complex Fe ions will further enhance Fe ion dissolution and promote the reaction forming iron-containing phases ( $\text{Ca}_4\text{Fe}_{(x)}\text{Al}_{(2-x)}\text{O}_6\text{CO}_3 \cdot 12\text{H}_2\text{O}$ ) [21], [22].

Also, in general, the quantity of reactants such as  $\text{C}_3\text{S}$ ,  $\text{C}_2\text{S}$ ,  $\text{C}_4\text{AF}$ , and  $\text{CaCO}_3$  decreases during the hydration process to form reaction products and ligands influencing these reactions. The decrease of reactants and growth of reaction products changes the pore connectivity that leads to conductivity modification. Correspondingly, blended PC with TIPA generates more CSH, AFt, and AFm, which contribute to strength enhancement. Overall, the addition of ligands in blended PC seem to reduce CH content by promoting its conversion to CSH.

#### **Relationship between heat flow, setting time, and electrical conductivity.**

The relationship between hydration heat flow and electrical conductivity measurement of pure PC and blended PC with carbonated and non-carbonated EAF1 slag, shows a complex dependency on the solid-liquid phase transformation during the 72 hr of hydration (Fig. 13). It is generally assumed that conductivity at early age predominantly reflects the bulk solution ionic mobility during the dissolution stage [23]. However, it is a more complex structure than simple ions mobility. The initial setting time generally denotes the initial formation of the solid precipitation products within the paste, and the point in which the paste loses its plasticity. This will decrease electrical conductivity as it will be affected by both pore solution resistivity and connectivity as well as the tortuosity of the pore structure. For instance, in this study, this occurs between 3-3.5 hr, this transformation of the solid-liquid interphase during the induction stage and the acceleration stage becomes the key factor in determining the mobility of ions in the pore structure based on the porification through the formation of the electrical double layers on solid, newly-formed surfaces. At this stage ion mobility is reduced, having impact on conductivity measurement. After this stage, the pore structure and its evolution become the dominant factor influencing conductivity, corresponding to the moment when the majority of the pores shrink, creating tortuous channels. This stage usually marks the end of the setting time, around 5-6 hr after mixing, when the development of surface charges on the precipitated phase is observed, the absorption of ions at the solid-liquid interface, the mobilization of ions modified the pore structure and the formation of gel layers with distinct interfacial properties. These phenomena, therefore indicate a continued reduction in electrical conductivity. Despite the modifications in the solid-liquid phase and the formation of the hardened matrix, the chemical reactions forming the product phases were observed on the calorimetry curve with the appearance of exothermic peaks.

The different observations between conductivity and the heat flow of hydration show that the intensity of the heat flow peak does not influence the material's electrical conductivity. This is because these are internal chemical reactions that cannot be detected by conductivity, as they do not involve a physical transformation. This demonstrates the complementarity between calorimetry and impedance spectroscopy, as well as with the setting time.

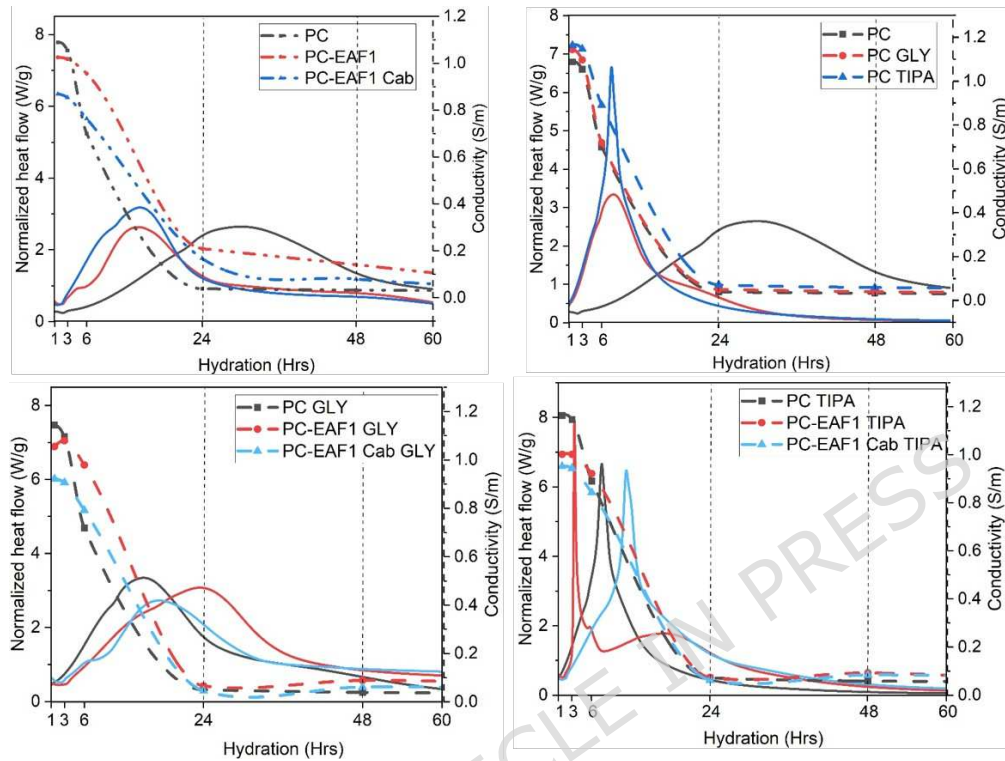


Figure 13: Correlation between electrical conductivity and heat flow.

Beyond confirming trends observed by calorimetry and setting time measurements, ACIS provided unique and complementary insights into the hydration of EAF slag blended systems that could not be obtained from other techniques alone. From the first 30 min of hydration, ACIS continuously distinguished carbonated from non-carbonated EAF slag blends through systematically lower initial conductivity in carbonated systems (0.8–0.9 S/m versus 0.95–1.07 S/m for non-carbonated blends), directly reflecting initial pore solution chemistry rather than any developed microstructure. It is acknowledged that fresh paste does not yet possess a microstructure in the conventional sense; at this early stage, conductivity reflects exclusively the liquid stage of the paste and ionic composition of the pore solution arising from immediate phase dissolution upon water contact. The 3–6 hr conductivity inflection point identified the nucleation phase across all systems, during the acceleration period, the intensity of the heat flow peak is not directly correlated with the rate of decrease in electrical conductivity. The progressive growth of reaction product phases including C-S-H, CH, and AFt leads to a continuous decrease in conductivity as the paste loses plasticity and the pore network becomes progressively less connected, regardless of the magnitude of the

concurrent exothermic reaction [23], [26], [27]. As illustrated in Fig. 13, this effect is most visually pronounced in the pure PC system, where the main heat flow peak occurs sharply between 3–8 hr while conductivity decreases monotonically from the onset of hydration. In EAF slag blended systems, the accelerated main heat flow peak reflecting the pozzolanic reaction of slag phases coincides with a period where conductivity has already reached lower values, consistent with earlier pore network densification occurring independently of the exothermic reaction rate. In all systems, conductivity decrease is therefore governed by the physical densification of the paste microstructure rather than by the rate of heat release, confirming that ACIS and calorimetry capture complementary but mechanistically distinct aspects of the hydration process. It is further acknowledged that electrical conductivity in fresh and hardening cement paste is governed not only by ionic mobility but also by the moisture state of the mixture containing different EAF slag. The free water available for ion transport is the primary conductive medium, and any factor reducing its availability such as physical water adsorption onto particle surfaces will reduce measured conductivity independently of changes in hydration reaction progress. For instance, the particle size distributions of the EAF slags such as EAF1 ( $d_{50} = 4.50 \mu\text{m}$ ) and EAF3 after milling ( $d_{50} = 5.20 \mu\text{m}$ ) result in higher specific surface areas relative to PC ( $d_{50} = 9.73 \mu\text{m}$ ). This increased surface area may promote physical adsorption of water onto slag particle surfaces, reducing the volume of free water available for ionic conduction and contributing to the lower initial conductivity observed in EAF slag blended systems independently of differences in ion-releasing capacity. This surface-area-driven water trapping effect should therefore be considered as a contributing factor alongside pore solution chemistry when interpreting absolute conductivity differences between PC and EAF slag blended systems. Finally, ACIS differentiated the ligand-specific effects of TIPA and Gly on ion mobility: Gly maintained pore connectivity (0–3 hr) in non-carbonated systems, while TIPA sustained at later stages (24 hr to 28 days) in carbonated systems, a reversal in behaviour that was not detectable from calorimetric data alone. Collectively, these observations demonstrate that ACIS does not simply confirm what calorimetry shows, but provides continuous growth of reaction product, and pore-network-sensitive information that meaningfully advances the understanding of how carbonation state and ligand type govern hydration kinetics in the EAF slag blended cement system.

The 28-day compressive strength achieved by PC-EAF1 Cab-TIPA (64 MPa) demonstrates that a 20 wt.% PC substitution with carbonated EAF slag combined with TIPA can meet structural concrete strength requirements, reducing the clinker factor while valorising EAF slag waste. TIPA, already widely used as a grinding aid in industrial cement production, showed additional capacity to accelerate  $C_4AF$  and  $C_3A$  dissolution and sustain ionic mobility in carbonated systems supporting its direct applicability at standard dosages (250 ppm) without significant retardation risk. Gly, offering more moderate strength enhancement but greater hydration uniformity across different EAF slag sources, is particularly relevant where slag compositional variability is unavoidable in industrial

settings. Pre-carbonation of EAF slag combined with organic ligand addition therefore represents a viable dual strategy for improving the reactivity of a low-grade industrial byproduct while reducing the carbon footprint of cement-based materials

## 5. Conclusion

In this study, electrical conductivity was used to evaluate the influence of 20 wt.% replacement of carbonated/non-carbonated EAF slag on the hydration process of Portland Cement (PC) and to reflect the influence of organic ligands triisopropanolamine (TIPA) and glycine (Gly). The findings revealed a complex relationship between heat flow of hydration and electrical conductivity, significantly influenced by solid-liquid phase transformations during hydration. Initial microstructure formation impacts conductivity due to changes in pore solution resistivity and connectivity, influenced by the type of phase product formed during hydration. The study also highlighted the complementary relationship between isothermal calorimetry and electrical conductivity in early-age liquid-solid transition. Additionally, conductivity depends not only on ion mobility in the pore structure but also on pore connectivity and the growth of later phase products.

Carbonated and non-carbonated EAF slag exhibited low reaction kinetics but enhanced 28-day compressive strength, especially with direct carbonation pretreatment. The inclusion of EAF slags led to decreased electrical conductivity due to changes in mass transfer and low reaction kinetics. The addition of TIPA and Gly ligands decreased early-stage strength but improved later-stage strength, attributed to the presence of amorphous silicate in carbonated EAF slag and ligand interactions during hydration. The peak intensity of  $C_2S$ ,  $C_3S$ , and  $CaCO_3$  decreased, while  $AFt$  and  $AFm$  increased, and  $CH$  decreased in the blended EAF cement composite, indicating enhanced hydration by TIPA and Gly. However, more research is still needed to understand better the factors that contribute on the overall electrical conductivity in cementitious binders.

## Credit authorship contribution statement

**Julson Aymard Tchio:** Conceptualisation, Investigation, Data curation, Formal analysis, Methodology, Validation, Formal analysis, Visualisation, Writing – original draft, Writing – review & editing. **Elijah Adesanya:** Conceptualisation, Resources, Data curation, Supervision, Visualisation, Writing – review & editing. **Brant Walkley:** Data curation, Supervision, Visualisation, Writing – review & editing. **Rafal Sliz:** Data curation, Validation, Writing – review & editing. **Juho Henrik Yliniemi:** Conceptualisation, Data curation, Resources, Methodology, Supervision, Visualisation, Writing – review & editing, Funding acquisition, Project administration.

## Declaration of Competing Interest.

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

The datasets generated and/or analysed during the current study are available in the Fairedata repository, through the links: <https://doi.org/10.23729/fd-e0a0c62e-180d-373a-aea1-ccc28266a857>

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