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Synergistic enhancement of grinding efficiency, hydration kinetics, and mechanical properties of Portland cement by ethaline

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Abstract

Deep eutectic solvents (DES) are emerging as green solvents explored extensively across diverse applications. DES form through hydrogen bonding between a hydrogen bond donor and acceptor, and since Portland cement hydration products are similarly governed by hydrogen bonding, DES incorporation is expected to influence hydration kinetics. This study investigates, for the first time, the effect of a green additive ethaline DES formed by choline chloride-ethylene glycol (1:2 M ratio) on the grinding efficiency, hydration behaviour, fresh properties, and mechanical performance of Ordinary Portland Cement (OPC-53). FTIR confirmed formation of a hydrogen-bonded eutectic structure, while density and rheological measurements showed a temperature-dependent decrease in density and viscosity; Arrhenius analysis exhibited excellent linearity, indicating thermally activated flow behaviour. DES improved grinding efficiency by reducing particle agglomeration, yielding higher Blaine fineness at equivalent milling energy on doses ranging 0.2-2.0 wt.%. Mortar flowability increased from 215 ± 3 to 234 ± 3 mm, while initial and final setting times were moderately extended, reflecting controlled hydration and suitable for mechanistic construction. Cumulative heat of hydration decreased by 2-11%, lowering the risk of thermal cracking. Optimum dosages (1.0-1.5 wt.%) raised 28-day compressive strength from 56 to 62 MPa and improved tensile strength, whereas excessive addition (2 wt.%) reduced strength due to over-retardation. FTIR and ESEM revealed modified hydration products and a denser microstructure, confirming that hydrogen-bond-mediated DES–cement interactions regulate hydration and refine microstructure. These findings position ethaline DES as a multifunctional, eco-friendly grinding aid and performance-enhancing admixture for Portland cement.

Keywords: Deep eutectic solvent; Ethaline; Ordinary Portland cement; Sustainable admixture; Grinding aid; Hydration; Mechanical properties.

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1. Introduction

Portland cement is a hydraulic binder that exhibits high strength and controlled setting characteristics, making it the most widely used construction material worldwide [1]. Nevertheless, its considerably adverse impact on environment has driven extensive research toward the development of more sustainable and environmentally benign cementitious formulations, such as lime calcined clay cement (LC³) and calcium sulfoaluminate (CSA) cement, which are still in nascent stage [2,3]. The principal constituent of Portland cement is clinker, which is manufactured through an energy-intensive pyro-processing route [4]. The clinker production leads to a substantial fraction of global anthropogenic CO₂ emissions [5]. Consequently, considerable research efforts are directed toward developing sustainable, ultra-high-performance cementitious systems that reduce clinker consumption while simultaneously improving mechanical performance, durability, and service life [6]. One of the most employed strategies for lowering the clinker factor is the incorporation of supplementary cementitious materials (SCMs), including fly ash, agricultural ashes, blast furnace slag, calcined clays, silica-rich industrial by-products, and other pozzolanic materials [7-9]. These SCMs partially replace clinker, thereby reducing both energy consumption and carbon emissions while often enhancing the long-term performance of cementitious materials [10-11]. In addition to SCMs, a wide range of functional additives have been incorporated into Portland cement formulations to tailor desirable properties. Some of these are polymers, hydrogels, organic compounds, and nanostructured materials that function as superplasticizers, air-entraining agents, viscosity modifiers, adsorbents, corrosion inhibitors, self-healing carriers, and reinforcing nanomaterials [12-18]. Such additives enable improved workability, higher mechanical strength, enhanced durability, reduced permeability, superior resistance to aggressive environments, and suitability for advanced applications such as ultra-high performance concrete and mechanized construction. Another important strategy to reduce energy consumption is the use of grinding aids during clinker and mineral grinding. In general, grinding aids are expected to reduce particle agglomeration, enhance milling efficiency, decrease specific energy consumption, and improve cement fineness, thereby lowering manufacturing costs and environmental impacts [19-20]. Despite their numerous advantages, both SCMs and functional additives can significantly influence the hydration behaviour of Portland cement [21-23]. These SCMs and additives alter hydration kinetics, nucleation and growth of hydration products, and the phase assemblage of calcium silicate hydrate (C-S-H), portlandite (Ca(OH)₂, C-H), ettringite, and other hydration products [13, 24]. These changes directly affect the microstructure, setting

characteristics, strength development, durability, and long-term performance of cementitious materials.

Triethanolamine (TEA) is the most widely used organic additive as grinding aid due to its ability to improve clinker grindability, accelerate aluminate hydration, and enhance early-age strength [25]. TEA acts by chelating Ca^{2+} , Al^{3+} , and Fe^{3+} ions released during clinker dissolution, thereby modifying the ionic composition of the pore solution and regulating hydration reactions [25]. In situ transmission electron microscopy and molecular simulations further reveal that TEA functions as a transient kinetic regulator by competing with silicate and aluminate species for Ca^{2+} coordination, thereby altering nucleation pathways without becoming structurally incorporated into C-(A)-S-H [26]. The addition of only 0.02 wt.% TEA decreases the fractions of low- and high-density C-S-H while increasing ultra-high-density (UHD) C-S-H by approximately 70% [27]. This enhancement results from the formation of lamellar $\text{Ca}(\text{OH})_2$ with a larger specific surface area, promoting stronger interfacial bonding with C-S-H gel and producing a denser, mechanically robust cement matrix [27]. TEA has also been found to accelerate pozzolanic reaction of fly ash leading to reduced C-H and increased C-S-H [28]. Recently, ionic liquids have attracted attention as multifunctional admixtures. Zinc- and copper-based ionic liquids have been reported to improve workability, regulate setting behavior, enhance mechanical strength and durability, and impart antimicrobial properties through uniform metal-ion distribution within the cement matrix [29-30]. In addition, organic complexing agents such as ethylenediaminetetraacetic acid (EDTA) and glutamic acid (GLTA) have been explored to promote calcium dissolution during carbonation curing, thereby accelerating CO_2 sequestration [31].

Unlike ionic liquid, deep eutectic solvents (DESs) are liquid mixtures typically formed by combining a hydrogen-bond acceptor (HBA) and a hydrogen-bond donor (HBD) in an appropriate molar ratio [32]. Strong hydrogen bonding interactions between the components, forming network, produce a pronounced melting-point depression and a highly tuneable liquid phase [32,33]. Variation of the HBA/HBD combination and molar ratio enables control over properties such as viscosity, polarity, hydrogen-bonding ability, hydrophobicity/hydrophilicity and solvation behaviour, making DESs attractive multifunctional additives for materials processing and green alternatives to inorganic acids [34,35]. The molecular characteristics of DESs may be particularly relevant to the coupled processes governing Portland cement hydration. Cement hydration involves particle wetting and dispersion, clinker dissolution, ionic

transport, nucleation, and growth of hydration products. The formation of initial hydration products creates new reactive surfaces that promote subsequent nucleation and growth, resulting in a self-reinforcing progression of hydration that can be conceptually related to the Matthew effect [36]. DES may modulate this process through its ability to alter particle–particle and particle–liquid interactions, improve dispersion, facilitate dissolution, and modify the local hydrogen-bonding and solvation environment. Thus, DES has the potential to function not merely as a processing aid but as a multifunctional additive capable of influencing successive stages of cement hydration. However, DESs have not been studied till now for their possible applications in the Portland cement as the additives.

In the present work, an ethaline DES has been studied for the first time as a multifunctional additive for Portland cement, serving both grinding aid as well as a modifier of cement hydration and mechanical performance. The ethaline DES, composed of choline chloride and ethylene glycol at a 1:2 M ratio, was synthesized using a hot-plate magnetic stirrer at 65-70 °C under continuous stirring for 30 min. The choice of this DES has been based on its hydrophilicity and low viscosity. The choline chloride has the antimicrobial property [37], and ethylene glycol is known for its efficacy as the grinding aid [13]. The physicochemical properties of this DES, including dynamic viscosity and density at different temperatures, have been evaluated, while its chemical structure is characterized by Fourier-transform infrared spectroscopy (FTIR). The characterized DES was subsequently employed as a grinding aid during the milling of Portland cement (OPC-53). The effects of DES incorporation on Blaine fineness, initial and final setting times, compressive and tensile strengths, heat of hydration, and fluidity are systematically evaluated in accordance with relevant Bureau of Indian Standards (BIS) protocols. The experimental results are used to determine the influence of DES on the grinding efficiency, hydration behaviour, and mechanical performance of OPC-53, with the observed macroscopic properties are correlated with microstructural features obtained by environmental scanning electron microscopy (ESEM).

2. Experimental

2.1. Materials and Instruments

All chemicals utilized were of analytical grade and were used without further additional purification. Deionized water from a Milli-Q system was employed in every experiment. Choline chloride and Ethylene glycol were procured from Sigma-Aldrich (Steinheim, Switzerland). Ordinary Portland Cement (OPC-53 grade) was obtained from a local supplier in

India. Standard quartz sand meeting the Bureau of Indian Standards (BIS) specifications (IS 650:1966) was used for preparing mortar. This sand consisted of three equal portions, such as grade I (1–2 mm), grade II (500 μm – 1 mm), and grade III (90 – 500 μm). Dynamic viscosity was measured by Rheometer (MCR 702 from Anton Paar). Density was measured using a pycnometer. Fourier-transform infrared (FTIR) spectroscopy was conducted using a Perkin Elmer spectrometer fitted with an attenuated total reflectance (ATR) accessory. Spectra were acquired in the range of 400–4000 cm^{-1} with a resolution of 4 cm^{-1} , averaging 16 scans per sample. An environmental scanning electron microscope (ESEM) of model FEI QUANTA 200 was utilized to examine the surface features of hardened mortar specimens. Grinding was done by using a ball mill of capacity 1 kg. Blaine fineness was measured using a variable flow type air permeability apparatus (Blaine type) (IS 5516). Setting times of cement pastes were determined with a Vicat apparatus (IS: 5513-1976). Mortar flowability was evaluated using a flow table (IS: 5512-1983). Compressive strength testing was carried out with a 2000 kN capacity motorized hydraulic compression testing machine. The heat of hydration was determined using an isothermal calorimeter (IS: 11262-1985).

2.2. Synthesize of DES

The DES was prepared using choline chloride (ChCl) as the hydrogen-bond acceptor (HBA) and ethylene glycol (EG) as the hydrogen-bond donor (HBD) at a 1:2 M ratio. This ratio of HBA and HBD were selected based on the literature [38]. The measured amounts of choline chloride (10.0 g) and ethylene glycol (8.89 g), based on the intended molar composition, were placed into a glass beaker. The combined mix was heated to 65–70 $^{\circ}\text{C}$ with constant stirring for 30 min using a hot plate magnetic stirrer, resulting in a uniform, clear, transparent, and viscous fluid. No visible phase separation or crystallization was observed in the final DES, confirming the successful creation of a stable eutectic mixture. After cooling to room temperature, the prepared DES was kept in a tightly closed container for subsequent uses.

2.3. Characterizations of DES

The density and dynamic viscosity of the synthesized deep eutectic solvent (DES) were measured at different temp., such 15, 30, 40, 50, 60, and 70 $^{\circ}\text{C}$. A pycnometer was employed for density determinations, whereas a rheometer was used to measure dynamic viscosity. Prior to each test, the DES was allowed to reach thermal equilibrium at the targeted temp. to ensure thermal stability. Each measurement was repeated for three times.

The chemical interactions and functional groups present in choline chloride, ethylene glycol, and the synthesized DES were analyzed using FTIR spectroscopy. Samples were applied directly onto the ATR crystal with no preliminary preparation. The characteristic peaks in FTIR spectra were used to identify functional groups and for confirming hydrogen-bond interactions between choline chloride and ethylene glycol.

2.4. Application of DES in OPC

The energy consumption determined by ball mill grinding at different running times of OPC was done without DES as the control, the other four with DES as a grinding aid with different amounts of DES 0.5, 1.0, 1.5, & 2.0 % in 100 g of 28 days hydrated OPC mortar. The ball mill used for grinding had specifications: voltage 230 V, current 10 amp. resulting power 2.3 kW. The energy consumption was obtained from relation: power x time of grinding. The Blaine was determined for analyzing grinding ability as per protocol of Bureau of Indian Standards [39].

The setting time was determined for one without DES as the control, other five with different amounts of DES added by partially replacing equivalent amounts of OPC. The constituents of these six specimens were: control: cement (400 g) + distilled water (98 g), with 0.2 wt.% DES: cement (399.2 g) + DES (0.8 g) + distilled water (98 g), with 0.5 wt.% DES: cement (398 g) + DES (2 g) + distilled water (98 g), with 1 wt.% DES: cement (396 g) + DES (4 g) + distilled water (98 g), with 1.5 wt.% DES: cement (394 g) + DES (6 g) + distilled water (98 g), and with 2.0 wt.% DES: cement (392 g) + DES (8 g) + distilled water (98 g). The water requirement was set at 0.85 times the normal consistency (28.8 %) of the cement weight. Sample preparation was carried out under laboratory conditions of temperature 27 ± 2 °C and relative humidity of 65 ± 5 % as per standard protocol [40]. Gauging was completed within 3-5 min from the time of water addition. The paste was placed in a Vicat mould resting on a non-porous plate, transferred immediately to a humidity chamber at 27 ± 2 °C and $\geq 90\%$ relative humidity, and tested at regular intervals. Initial setting time (IST) was recorded when the Vicat needle (1 mm diameter) failed to penetrate to within 5 ± 0.5 mm of the mould base. Final setting time (FST) was recorded when the annular attachment made an impression on the surface but did not penetrate.

Compressive strength was determined on 70.6 mm cube specimens following BIS protocol [41]. Mortar mixes contained OPC (200 g), standard sand (600 g), and water (81.6 g) calculated as (normal consistency/4 + 3) wt.% of the combined cement and sand mass. With DES incorporated mortars 0, 0.2, 0.5, 1.0, 1.5 and 2.0 wt.% DES replacing OPC, with sand and water

kept constant. Mortar was cast in 70.6 mm cube moulds in three layers, compacted on a vibrating table at $12,000 \pm 400$ vibrations min^{-1} for 2 min to eliminate entrapped air, then stored in a humidity chamber (27 ± 2 °C, $\geq 90\%$ RH) for 24 h. After demoulding, specimens were water-cured and tested at 3, 7, 28, and 60 d on three specimens per mix using a 2000 kN motorized hydraulic compression testing machine.

Tensile strength was evaluated following IS 4031 standard procedure [41]. The control mortar consisted of OPC (40 g), standard sand (120 g), and distilled water (16.3 g) to find with compressive strength. The DES-modified mixes substituted 0.2, 0.5, 1.0, 1.5 and 2.0 wt.% of OPC with DES (0.08, 0.20, 0.40, 0.6 and 0.80 g, respectively), maintaining constant sand and water contents. Mortar was cast in briquette moulds in three layers with adequate tamping and vibration, stored under moist conditions for 24 h, demoulded, and water-cured for 3, 7, 28, and 60 d. Tensile strength was determined in triplicate by applying a uniaxial load at a controlled rate until fracture, and results are reported as mean $\pm$ standard deviation.

The heat of hydration was determined by an acid-dissolution isothermal calorimetric method following standard protocol [42]. Cement pastes for calorimetric analysis were prepared using OPC (60 g) and distilled water (24 g; w/c = 0.40). DES-modified systems incorporated 0.2, 0.5, 1.0, 1.5 and 2.0 wt.% DES (0.12, 0.30, 0.60, 0.90 and 1.20 g, respectively) as partial OPC replacement, with water held constant at 24 g. Pastes were hand-mixed for 4 min, transferred to sealed plastic containers, and stored vertically at 27 ± 2 °C. The calorimeter heat capacity was standardized using the reference dissolution of zinc oxide in a 2 N HNO_3/HF acid mixture. The heat of solution of both anhydrous and hydrated cement specimens (at 3, 7, 28, and 60 days) was determined from the temperature rise upon acid dissolution. The heat of hydration was computed as the difference between these two values and expressed in J g^{-1} of cement [42].

Mortar fluidity was evaluated using a flow table test in accordance with IS 4031 (Part 7) [43]. The control mortar was prepared with OPC (420 g), standard sand (1440 g), and distilled water (230 g). For DES-modified mortars, OPC was partially replaced with 0.2, 0.5, 1.0, 1.5 and 2.0 wt.% DES, with sand and water kept constant. All constituents were mixed for 4 min, and fresh mortar was placed in a standard truncated-cone mould centred on the flow table, levelled, and the mould lifted. The table was then raised and dropped 25 times through 12.5 mm in 15 s. The spread diameter was measured at four equidistant points and averaged. Flow percentage was calculated as [43]:

$$\text{Flow (\%)} = [(D - D_o) / D_o] \times 100$$

where D is the average spread diameter after the flow test (mm) and D_0 is the original base diameter of the mould (mm).

3. Results and discussion

The chemical structure of ethaline formed by choline chloride (ChCl) and ethylene glycol (EG) in 1:2 M proportions is illustrated in **Fig.1**. To understand the physicochemical properties of ethaline, the different characterization techniques were used as described below.

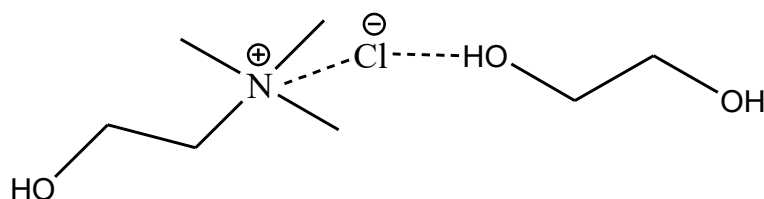


Fig. 1. Chemical structure of ethaline DES formed by choline chloride and ethylene glycol. The dash line represents hydrogen bonding.

3.1. Characterizations of ethaline

FTIR spectroscopy was used to understand the nature of bonding between ChCl and EG and to confirm the formation of the ethaline. The spectra of the individual components and ethaline are given in **Fig. S1** (Appendix-A). The broad O-H stretching region at 3200–3400 cm^{-1} showed the most noticeable changes upon ethaline formation. For pure EG and ChCl, the O-H stretching bands occur at 3295 and 3226 cm^{-1} , respectively, whereas the ethaline exhibited a single broad band at 3337 cm^{-1} . This shift to higher wavenumber relative to both parent components indicated a net weakening of the average hydrogen-bond strength in the mixture, consistent with disruption of the strong, ordered EG–EG and ChCl ionic hydrogen-bond networks present in the pure components. The single, broadened band in the DES further suggested that the resulting O-H environment was more heterogeneous, reflecting the formation of new, comparatively weaker O–H(EG)⋯Cl[−] interactions in place of the stronger self-associated networks of the individual components. This behaviour is consistent with reports for ethaline, where similar changes in the O-H stretching region have been attributed to weakening of the original EG–EG hydrogen-bond interactions along with formation of new hydrogen bonds between EG and ChCl [44]. Additional evidence was obtained from the C–O stretching region at 1000–1090 cm^{-1} . The distinct band at 1035 cm^{-1} in pure EG became broader and less intense in the ethaline, suggesting perturbation of the local EG environment and

disruption of the ordered intermolecular interactions present in the pure liquid. In contrast, the O-H bending vibration showed only minor changes, from 1650 cm^{-1} for EG and 1640 cm^{-1} for ChCl to 1644 cm^{-1} in the ethaline, indicating comparatively limited sensitivity of this mode to DES formation. Similarly, the C-N⁺ stretching band of ChCl changed only slightly, from 952 to 954 cm^{-1} , suggesting that the choline cation remained largely unaffected while the chloride anion participated more directly in hydrogen bonding with EG. Thus, the noticeable changes in the O-H stretching and C-O stretching regions, together with the comparatively small changes in the O-H bending and C-N⁺ vibrations, demonstrated that DES formation involved a reorganisation of the hydrogen-bond network, dominated by weakening of the original EG-EG and ChCl self-association and the establishment of new, distributed EG $\cdots$ Cl⁻ hydrogen bonds.

The rheological behaviour of ethaline was investigated to evaluate its flow characteristics and suitability for application as a multifunctional additive in Portland cement. It is seen from **Fig. 2a** that the variation of torque with shear rate for ethaline DES increased linearly with increasing shear rate over the measured range (0–1000 s^{-1}). The linear torque–shear-rate relationships indicated that ethaline has essentially Newtonian flow behaviour. The different slopes of the torque–shear-rate profiles at different temperatures indicate that the flow resistance of ethaline is temperature dependent. The corresponding shear stress as a function of shear rate is shown in **Fig. 2b**. The shear stress increased linearly with increasing shear rate at all investigated temperatures (15–70 °C), further confirming the Newtonian behaviour. The approximately linear shear stress–shear-rate relationships indicate that the rheological response of ethaline remains predictable over the investigated range, without appreciable shear-thinning or shear-thickening behaviour. The temperature-dependent differences among the flow curves further demonstrate that the resistance to flow changes with temperature. This predictable rheological behaviour would be advantageous for its use in Portland cement because it facilitates reproducible dosing, mixing, and dispersion of the DES within the cement matrix. In particular, the absence of significant non-Newtonian behaviour suggests that the flow characteristics of ethaline remain relatively stable under the range of shear conditions relevant to processing.

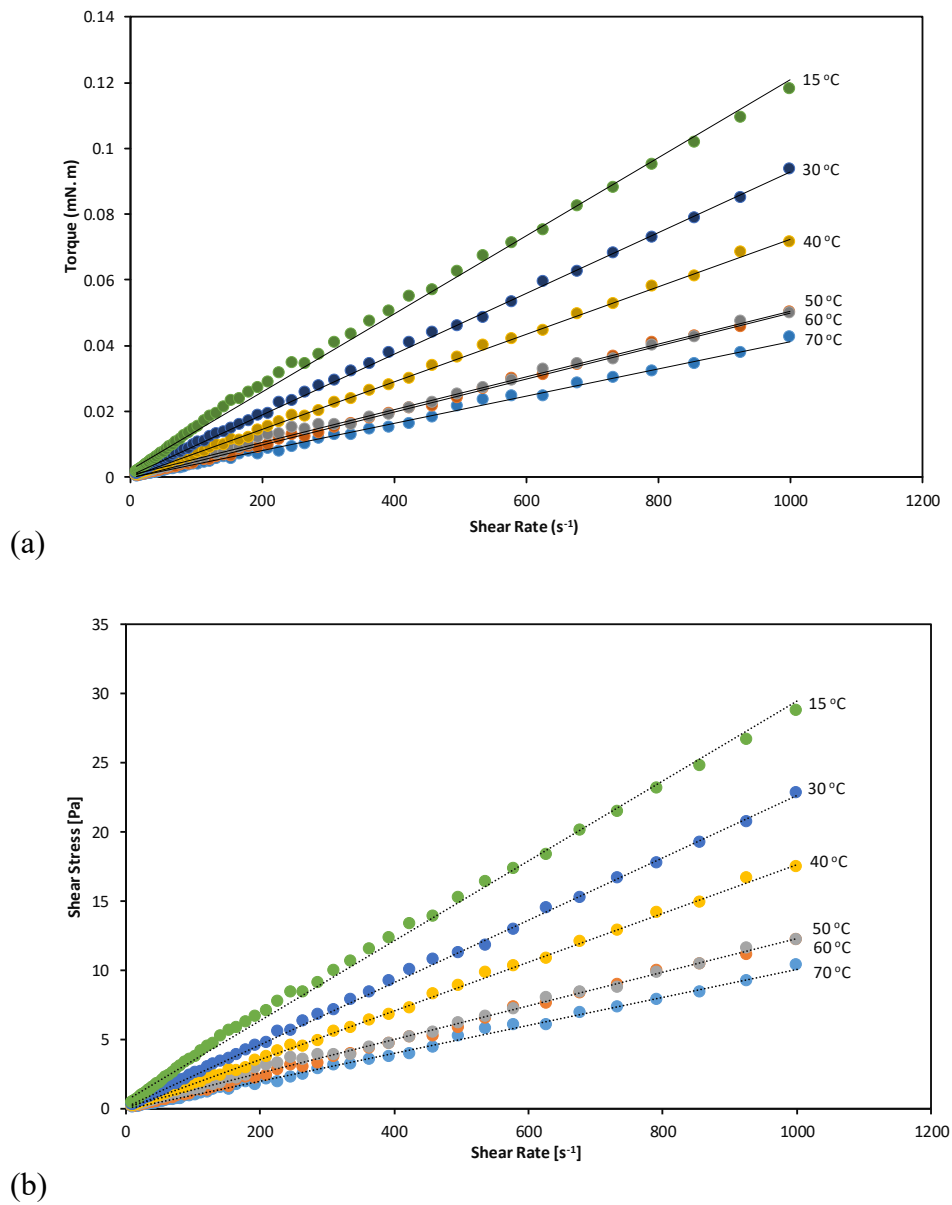
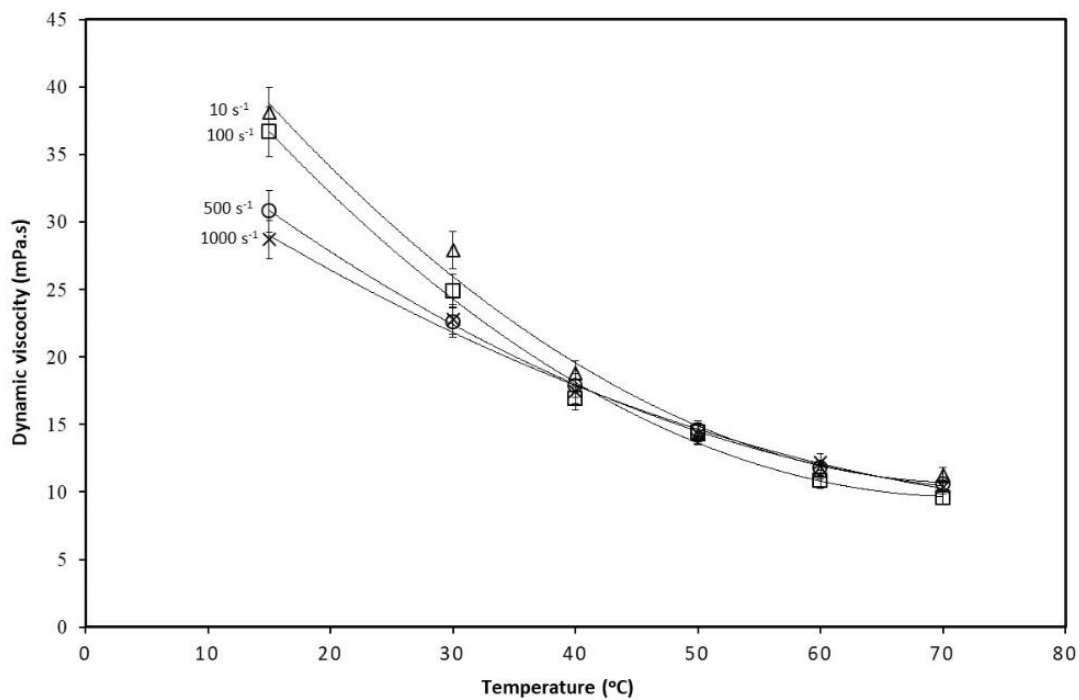


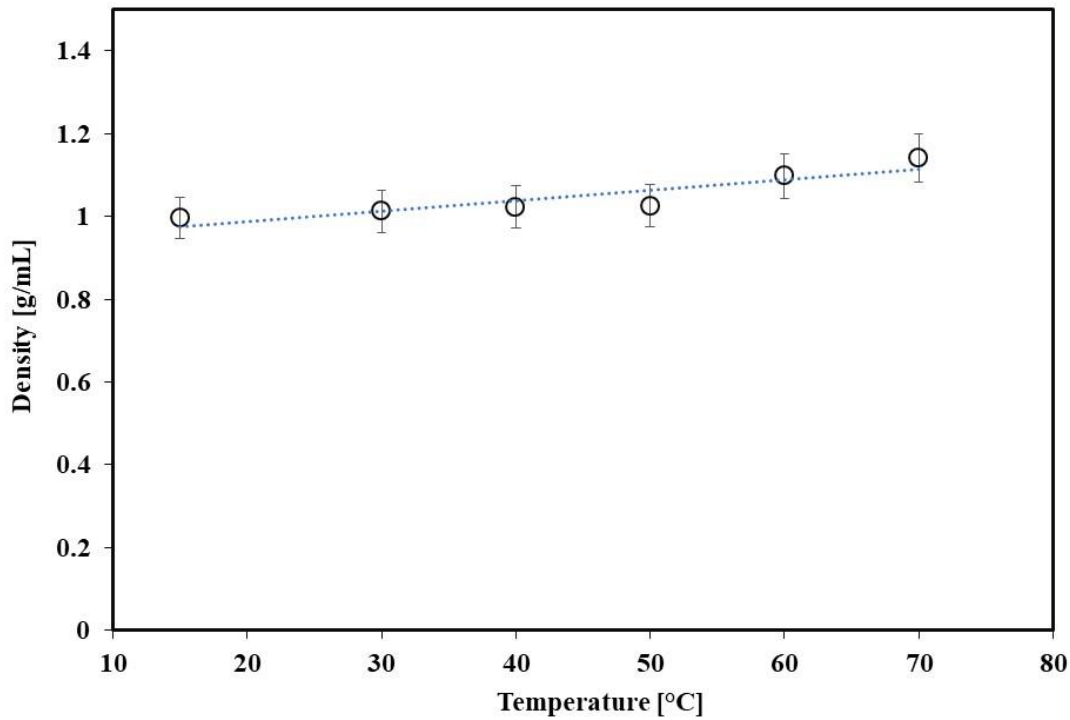
Fig. 2. Variations of torque as a function of shear rate (a) and shear stress as a function of shear rate (b) for ethaline DES (ChCl-EG, 1:2 M ratio) measured at temperatures ranging from 15 to 70 °C.

The variations of dynamic viscosity of ethaline at varying temperature and at different shear rates were studied, and data are given **Fig. 3a**. It is seen from curves given in **Fig. 3a** that the apparent viscosity decreased systematically with increasing temperature at all studied shear rates, suggesting the strong temperature dependence of the flow resistance of ethaline. Similar trend was observed by Gabriele et al. in variation of viscosity with temperature without shear [45]. The decrease was more evident at lower shear rates, where the viscosity values are comparatively higher, whereas the apparent viscosity values at the different shear rates become

progressively closer at elevated temperatures. At temperatures above approximately 60 °C, the apparent viscosity curves converged to a relatively narrow range, indicating that the influence of shear rate on the measured viscosity becomes less pronounced at higher temperatures. This temperature-dependent reduction in apparent dynamic viscosity was consistent with increased molecular mobility and reduced resistance to flow at elevated temperatures. The observed behaviour is particularly relevant to the use of ethaline as a cement additive, since its lower apparent viscosity at elevated temperatures can facilitate its distribution and mixing within the cementitious system. The variation in density of ethaline with temperature is shown in **Fig.3b**. The density was found to vary systematically with temperature over the range studied. The measured ethaline density showed an increase with temperature under the experimental conditions used in the present work, which is contrary to the observed decrease in liquid density of triethylene glycol with increasing temperature [45]. This behaviour suggested that the temperature dependence of ethaline density was influenced by changes in its molecular organization and intermolecular interactions.



(a)



(b)

Fig. 3. Variations of apparent dynamic viscosity measured at different shear rates (a) and density (b) as a function of temperature of ethaline.

Arrhenius plots of the natural logarithm of apparent dynamic viscosity ($\ln \eta$) versus reciprocal temperature, expressed as $1000/T$ (K^{-1}), for ethaline DES at shear rates of (a) 10, (b) 100, (c) 500, and (d) 1000 s^{-1} over the temperature range of 15-70 °C are shown in **Fig. S2** (Appendix A). The reasonably good linear correlations were obtained at all shear rates, with $R^2 > 0.97$, indicating that the temperature dependence of apparent viscosity followed Arrhenius behaviour. This thermally activated flow is consistent with the hydrogen-bonded structure of ethaline, in which increasing temperature facilitates molecular mobility by weakening intermolecular interactions. The apparent activation energy for viscous flow (E_a) was calculated using the Arrhenius relationship [45]:

$$\eta = A \exp^{(E_a/RT)}$$

in logarithmic form,

$$\ln \eta = \ln A + E_a/RT$$

where η is the apparent dynamic viscosity (mPa·s), A is the pre-exponential factor, E_a is the apparent activation energy for viscous flow (J mol^{-1}), R is the universal gas constant ($8.314 \text{ J mol}^{-1} \text{ K}^{-1}$), and T is the absolute temperature (K). Because the plots were constructed using $1000/T$ rather than $1/T$, E_a was calculated from the slope using following relation:

$$E_a = \text{slope} \times 1000 \times R$$

The calculated E_a values were 19.98, 20.77, 16.55, and 15.70 kJ mol^{-1} at 10, 100, 500, and 1000 s^{-1} , respectively. Thus, E_a decreased progressively with increasing shear rate, with a reduction of 5.07 kJ mol^{-1} (approximately 24%) between 100 and 1000 s^{-1} . The E_a values reported in literature without shear are 27.20 and 28.12 kJ mol^{-1} for the same DES [45,46]. The viscosity with shear is measure of a fluid resists internal deformation when a sliding force is applied. The shear-dependent decrease in the apparent activation energy observed in the present work suggests that applied shear partially disrupted the hydrogen-bonding network within the DES, thereby reducing intermolecular resistance and facilitating molecular rearrangement during flow. The combined temperature- and shear-dependent behaviour indicated that ethaline exhibits weak shear-thinning characteristics and thermally activated viscous flow. The reduction in E_a at higher shear rates is particularly relevant to the application of ethaline as a cement-grinding additive. Under the high-shear conditions prevailing in cement mills, particularly at shear rates above approximately 500 s^{-1} , the DES is expected to flow and distribute more readily than would be predicted from its low-shear viscosity. This behaviour is expected to facilitate rapid spreading and dispersion over cement particle surfaces, leading to more uniform particle coating during milling. Consequently, the shear-dependent rheological response of ethaline may contribute to reduced interparticle adhesion and agglomeration, enhancing grinding efficiency and resulting to lower energy input.

3.2. Characterizations of ethaline in Portland cement

The compatibility between the ethaline and hydrated products, such as C-S-H, in the DES-modified OPC mortar was investigated by comparing the FTIR spectra of the synthesized DES shown in **Fig.S1c**, OPC mortar given in **Fig.S3a** and DES-modified OPC mortar containing 2.0 wt.% DES shown in **Fig. S3b**. The mortar samples were prepared and cured for 28 d as detailed in the experimental section. FTIR spectrum of the OPC mortar shown in **Fig. S3a** exhibits characteristic bands associated with cement hydration and carbonation products, including the Si-O stretching vibration of the silicate network of C-S-H at 1084 cm^{-1} , carbonate

bands at 1410 and 874 cm^{-1} attributed to carbonation and/or calcite formation, and a low-wavenumber band at 515 cm^{-1} associated with Si-O bending and Ca-O vibrations. These bands are assigned based on literature [47,48]. The absence of a distinct O-H stretching band in the OPC mortar spectrum, together with the strong carbonate bands at 1410 and 874 cm^{-1} , suggested substantial carbonation of the analyzed mortar surface, which may mask the broad O-H contribution from hydration products. The FTIR spectrum of the DES-modified cement mortar shown in **Fig. S3b** provided evidence for the presence of DES-derived functional groups while retaining the characteristic spectral features of the cementitious matrix. A broad and intense band centered at 3337 cm^{-1} was assigned to O-H stretching vibration in the DES, which was observed at 3339 cm^{-1} in pure DES. The band at 2947 cm^{-1} corresponded to aliphatic C-H stretching vibrations. The band at 1477 cm^{-1} assigned to a combination of CH_3 asymmetric deformation of ChCl and CH_2 scissoring vibrations of EG. The bands at 1039 and 952 cm^{-1} are associated with C-N^+ stretching vibrations of the choline quaternary-ammonium moiety. The characteristic bands of the hydrated and carbonated OPC matrix are also retained in the DES-modified mortar. The bands at 1416 and 872 cm^{-1} corresponded to carbonate vibrations. The strong band at 1083 cm^{-1} is mainly attributed to Si-O stretching vibrations of the polymerized C-S-H gel. This region contained overlapping contributions from C-O vibrations associated with EG, making the individual DES and C-S-H contributions difficult to resolve. In the lower fingerprint region, the band at 515 cm^{-1} is assigned mainly to Si-O and Ca-O vibrations originating from the cementitious matrix and/or silica-containing sand phases. FTIR spectrum showed that the characteristic functional groups of the ChCl-EG DES are retained within the mortar specimen after 28 d of curing, while the characteristic bands of C-S-H and carbonate phases were remained unaltered.

The surface morphology and microstructural features of OPC mortar (control) and OPC mortar with 0.5 and 2 wt.% DES specimens cured for 28 d were studied using environmental scanning electron microscopy (ESEM). The ESEM images thus obtained are given in **Fig. 4**. The ESEM images revealed noticeable differences in the microstructure of the control and ethaline-modified OPC-53 mortars. The control mortar exhibited a relatively heterogeneous and loosely packed morphology, with irregular agglomerates, interconnected pores, and distinct microcracks given in **Fig.4a**. At 0.5 wt.% ethaline, the microstructure appears comparatively denser and more compact, with reduced visible pore spaces and improved packing of hydration products. It is seen from ESEM image of 0.5 wt.% ethaline-modified mortar given in **Fig.4b** that the surface appears comparatively more uniform, with closely distributed particulate

features and fewer prominent large voids in the matrix. At 2 wt.% ethaline, ESEM images in **Fig.4c** show a more developed and closely packed microstructure with abundant fine crystalline/hydration products and fewer large voids. The ESEM images of 2 wt.% ethaline-modified mortar reveals a highly particulate morphology, with numerous fine angular and irregular features distributed across the observed surface. The ESEM images demonstrated the denser surface morphology, improved particle arrangement, and reduction in visible void structure and cracks with increasing ethaline content.

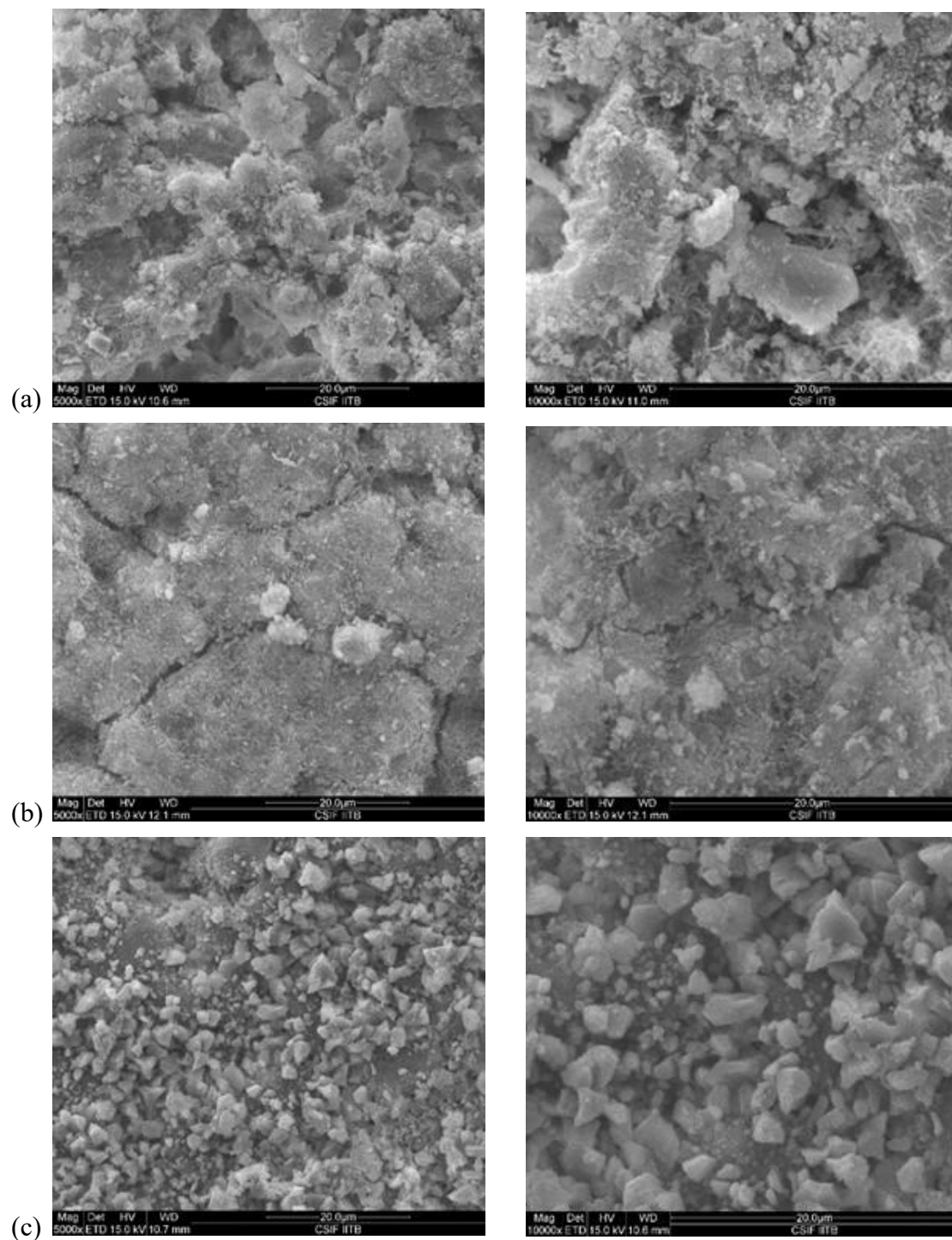


Fig. 4. The ESEM images two different magnifications of specimens of control OPC-53 mortar (a), OPC-53 mortar with 0.5 wt.% ethaline (b), and OPC-53 mortar with 2 wt.% ethaline.

It is known that the considerable heat is generated during hydration of Portland cement, which causes thermal stress induced cracks resulting to loss of durability of cementitious structures [49]. Therefore, the effect of ethaline addition on the heat of hydration of OPC-53 mortar was studied, and data thus obtained are given in **Table 1**. It is seen from Table 1 that there was a systematic reduction in heat evolution with increasing ethaline dosage at all curing ages. The control mortar exhibited heat evolution of 292 ± 10 , 329 ± 11 , 334 ± 14 , and 339 ± 16 kJ/kg at 3, 7, 28, and 60 d, respectively. On addition and increasing ethaline content from 0.2 to 2.0 wt.%, the heat of hydration systematically decreased. For example, the heat decreased from 292 kJ/kg for the control to 265 kJ/kg at 2.0 wt.% ethaline at 3 d curing, while at 28 d it decreased from 334 to 299 kJ/kg. At 60 d, the corresponding values were 339 and 301 kJ/kg, respectively. The systematic decrease in heat of hydration suggested that ethaline modified the hydration kinetics of OPC-53, resulting to a lower rate or of heat evaluation by hydration reactions. The effect is particularly evident at 1.0–2.0 wt.% ethaline, indicating a stronger influence on the hydration process at higher additive concentrations. The lower heat evolution at 3 and 7 d is consistent with a moderated early hydration process, while the persistence of lower values at 28 and 60 d indicates that the influence of ethaline extends beyond the initial hydration period. The ESEM observations discussed above provided complementary morphological evidence for effect of lower heat of hydration. Compared with the control mortar, the ethaline-modified specimens exhibited noticeable differences in ESEM images, such as denser surface morphology, improved particle arrangement, and reduction in visible void structure and cracks with increasing ethaline content.

Table 1. Comparison of heat of hydration of specimens of control OPC-53 mortar and OPC-53 mortar with varying amount ethaline cured for different time.

Ethaline (wt.%)	Heat of hydration (kJ/kg)			
	3 d	7 d	28 d	60 d
0 (Control)	292±10	329±11	334±14	339±16
0.2	289±11	322±10	329±12	330±15
0.5	282±9	316±12	319±13	325±14
1.0	273±10	308±9	314±12	309±13

1.5	269±8	301±7	305±13	304±12
2.0	265±8	292±8	299±13	301±12

3.3. Characterizations of cementitious properties

The efficacy of ethaline as an additive to OPC was evaluated by partially replacing OPC with 0.2, 0.5, 1.0, 1.5, and 2.0 wt.% ethaline, and the resulting cementitious properties were compared with the control. The variations in initial setting time (IST) and final setting time (FST) with ethaline content are given in **Table 2**. A systematic increase in both IST and FST was observed with increasing ethaline dose. The control cement exhibited an IST and FST of 170 and 205 min, respectively, whereas the corresponding values increased to 182, 187, 192, 194, and 197 min for IST and 221, 228, 236, 241, and 245 min for FST at 0.2, 0.5, 1.0, 1.5, and 2.0 wt.% doses of ethaline, respectively. The observed longer duration of setting time is consistent with the lower heat evolution observed during the early stages of hydration, indicating a retardation of the hydration process in the presence of ethaline. The ionic and hydrogen-bonding constituents of ethaline, particularly Cl^- from ChCl and the hydroxyl groups of EG, could have interacted with the interface of hydrating clinker phases and the aqueous phase. These interactions could have temporarily formed the covering layer on reactive clinker phases, particularly C_3S and C_3A , thereby reducing the dissolution rate and hence release of Ca^{2+} and other ionic species into the pore solution. Consequently, the nucleation and growth of early hydration products, particularly C-S-H, might have delayed, resulting in a longer induction period and slower hardening. This interpretation is consistent with the reduction in heat of hydration observed for ethaline-modified OPC. It is important to note that the retardation was moderate and systematic rather than excessive. The controlled extension of setting time can provide a longer period for mixing, transportation, placement, and compaction of cementitious materials.

Flow values, representing the fluidity and workability of the OPC paste, were measured at different ethaline doses. As can be seen from data given in **Table 2**, the flow value increased systematically with increasing ethaline doses. The control mixture exhibited a flow value of 215 mm, which increased to 223, 225, 229, 230, and 234 mm at 0.2, 0.5, 1.0, 1.5, and 2.0 wt.% ethaline, respectively. This systematic increase indicated that ethaline improved the workability and flowability of the cement paste. The enhanced flow may be attributed to the ability of ethaline to modify interparticle interactions and reduce particle agglomeration,

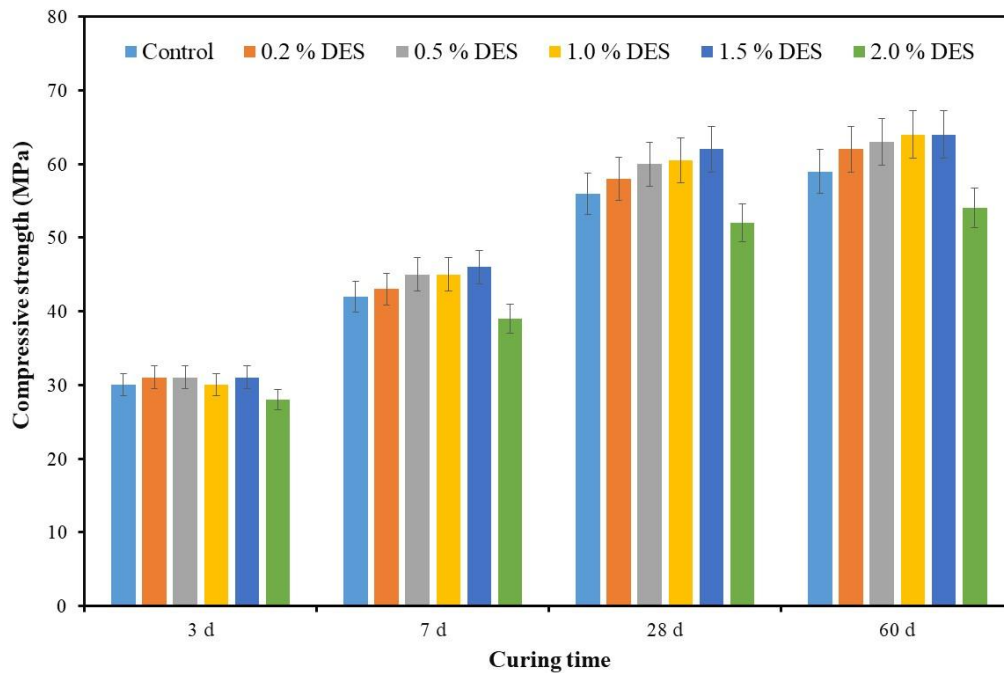
thereby promoting better dispersion of cement particles. In addition, the relatively low viscosity of ethaline may facilitate particle movement within the paste.

Table 2. Comparison of initial setting time (IST) and final setting time (FST) of specimens of control OPC-53 mortar and OPC-53 mortar with varying amount of ethaline.

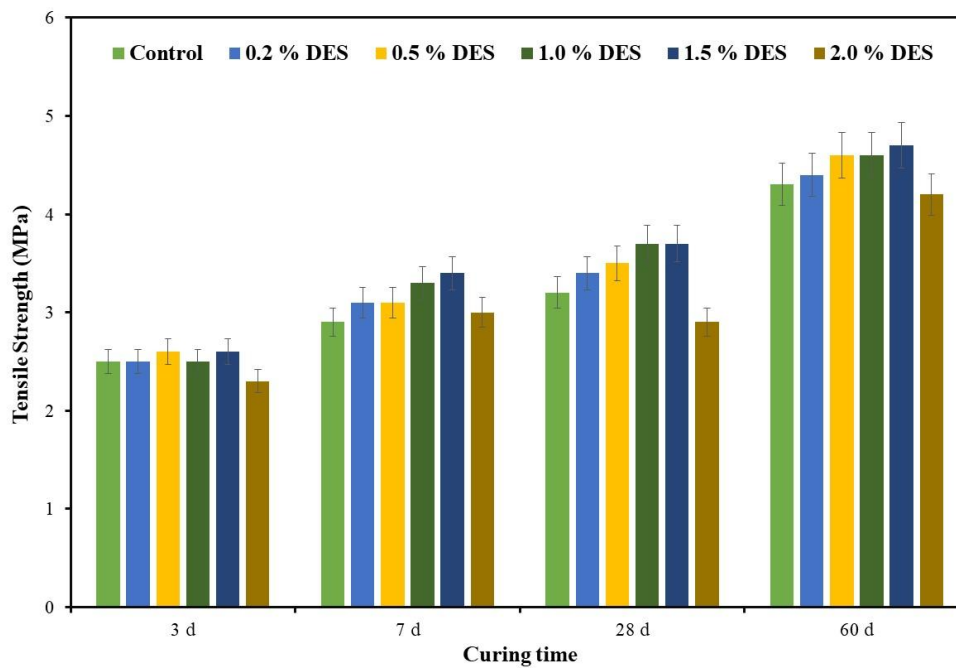
Ethaline (wt.%)	Setting Time (min)		Flow (mm)
	IST	FST	
0 (control)	170 ± 10	205 ± 12	215 ± 3
0.2	182 ± 10	221 ± 12	223 ± 3
0.5	187 ± 10	228 ± 12	225 ± 3
1.0	192 ± 10	236 ± 12	229 ± 3
1.5	194 ± 10	241 ± 12	230 ± 3
2.0	197 ± 10	245 ± 12	234 ± 3

The compressive and tensile strength data obtained at different doses of ethaline at different curing times are plotted in **Fig. 5**. It is seen from Fig.5 that there was a consistent enhancement effect of ethaline addition to compressive strength and tensile strength of OPC-53 mortar, although the extent of improvement depended on the ethaline dosage and curing age. The control mortar exhibited the lowest strength development, whereas incorporation of ethaline significantly improved both compressive and tensile strength up to an optimum dosage of 1.5 wt.%. At this dosage, the strength enhancement was distinctly noticeable, indicating that ethaline promotes a more favourable cementitious microstructure. Further increasing the dosage to 2.0 wt.% does not provide a comparable additional benefit, suggesting that the effect of ethaline reaches an optimum at 1.5 wt.%. The compressive strength appeared to reach a plateau at 28 d, indicating that the major contribution of ethaline to compressive load-bearing capacity is largely realized by this age. In contrast, the tensile strength continues to increase with curing age, particularly for the 1.5 wt.% dose of ethaline. This difference may be associated with continued development and refinement of the hydrated cement matrix, which can progressively improve resistance to crack initiation and propagation even after the compressive strength has approached saturation. Thus, tensile strength seems to suggest ongoing microstructural development at later ages. The better performance at 1.5 wt.% ethaline is consistent with the ESEM observations, where the optimum mixture exhibits a comparatively compact and well-integrated cementitious matrix with fewer visible large pores/voids and a

more continuous morphology. These observations provide morphological support for the improved mechanical performance, although ESEM alone cannot establish the specific hydration mechanism responsible. The results therefore suggest that an appropriate ethaline concentration promotes a more compact cementitious structure, whereas increasing the dosage beyond 1.5 wt.% does not produce further proportional improvement.



(a)



(b)

Fig.5. Variations of compressive strength (a) and tensile strength (b) of specimens of control OPC-53 mortar and OPC-53 mortar with varying amount of ethaline.

3.4. Grinding performance of ethaline

The ball mill used for grinding studies was operated at 230 V and 10 A, corresponding to a power input of 2.30 kW, and the energy consumption was calculated from $E = P \times t$. The effect of ethaline (DES) addition on the grinding efficiency of OPC-53 cement in terms of Blaine fineness at different grinding times and energy inputs are given in **Table 3**. At an energy consumption of 1.53 kWh, the Blaine fineness increased from 217 m²/kg for the control to 218, 218, 220, and 221 m²/kg with 0.5, 1.0, 1.5, and 2.0 wt.% DES, respectively. With increasing grinding energy, the improvement became more pronounced. At 2.68 kWh, the Blaine value increased from 223 m²/kg for the control to 226, 228, 230, and 231 m²/kg with increasing DES content from 0.5 to 2.0 wt.%. At the highest energy input of 3.45 kWh, the corresponding values were 231, 235, 235, 237, and 240 m²/kg, respectively. The increase in Blaine fineness at comparable energy inputs indicated that ethaline facilitates particle-size reduction during grinding. Thus, ethaline consistently enhanced cement fineness at a given energy input, with the highest Blaine values obtained at 2.0 wt.% DES. This improvement can be attributed to adsorption of DES on freshly generated cement surfaces, which may reduce interparticle attraction and limit re-agglomeration. The resulting better dispersion allows the grinding media to interact more effectively with individual cement particles, increasing the fraction of mechanical energy utilized for particle fracture and generation of new surfaces. Consequently, higher cement fineness can be achieved at the same energy input. Notably, although 2.0 wt.% DES provided the highest grinding fineness, the optimum dosage for subsequent mechanical performance may differ, indicating that grinding efficiency and overall cement performance should be considered separately.

Table 3. The variation of Blain as function of energy consumption with varying doses of ethaline.

Grinding time (h)	Energy (kWh)	Blaine of OPC-53 with different doses of ethaline (m ² /kg)				
		Control	0.5 wt. %	1.0 wt. %	1.5 wt. %	2.0 wt. %
0.67	1.53	217±4	218±5	218±6	220±5	221±4
0.83	1.92	220±5	220±6	221±5	223±6	224±6
1.0	2.30	220±4	221±5	223±4	223±4	225±3

1.17	2.68	223±3	226±3	228±5	230±5	231±5
1.33	3.07	228±4	230±4	232±3	234±3	235±5
1.50	3.45	231±6	235±5	235±4	237±5	240±2

4. Conclusions

The present study demonstrates, for the first time, the potential of a deep eutectic solvent (DES) based on choline chloride and ethylene glycol (1:2 M ratio) as a multifunctional additive for Portland cement, with beneficial effects extending from grinding efficiency to hydration behaviour and mechanical performance. This DES, known as ethaline, was readily synthesized by mild heating and stirring, and its formation was confirmed by FTIR analysis, which showed characteristic changes in the O–H stretching region associated with hydrogen-bond formation between the hydrogen-bond acceptor and donor components. The temperature-dependent rheological and density measurements further demonstrated this DES has Newtonian behaviour and the tuneable physicochemical properties, supporting its suitability for cement-processing applications. Incorporation of DES during cement grinding produced a clear improvement in grinding efficiency. Increasing DES dosage progressively enhanced the fineness of the ground cement, with 2.0 wt.% DES producing the highest Blaine fineness. This improvement is attributed to the ability of the DES to reduce particle-particle adhesion and agglomeration during milling, thereby facilitating more effective size reduction and dispersion of cement particles. The enhanced fineness provides a larger reactive surface area and consequently has implications for subsequent cement hydration. The DES-treated cement also exhibited improved flowability, indicating that the additive can simultaneously influence particle dispersion and fresh-state behaviour. The hydration results showed that DES incorporation modified the heat-evolution profile and hydration kinetics of Portland cement. The changes in hydration behaviour are likely associated with interactions between the DES components, water, and the hydrating cement phases. The extensive hydrogen-bonding network characteristic of DES may facilitate redistribution of water and ionic species at the cement–water interface, thereby influencing dissolution of clinker phases and subsequent formation of hydration products. The resulting changes in hydration were reflected in the development of the cement microstructure observed by ESEM, suggesting a more interconnected and compact hydration-product matrix at optimum DES doses. The mechanical-strength results further

confirmed that DES addition can improve cement performance. However, the optimum dosage for mechanical properties did not necessarily correspond to the dosage producing maximum grinding fineness. This distinction is particularly important because excessive DES addition may alter the hydration environment and fresh-state characteristics even when further improvements in fineness are achieved. Thus, the results indicate that grinding efficiency and cement performance should be optimized independently but considered as interconnected processes. The experimental results obtained in the present work establish choline chloride–ethylene glycol DES as a promising multifunctional cement additive capable of improving grindability, particle dispersion, hydration behaviour, workability, and mechanical performance.

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Declarations

Ethics approval and consent to participate: Not applicable. This study did not involve human participants, animal subjects, or any procedures requiring ethical approval from an institutional review board. The research did not involve human participants.

Consent for publication: Not applicable. The manuscript contains no individual person's data in any form.

Competing interests: The authors declare no competing interests.

Appendix A. Supplementary data

Supplementary data to this article can be found online at:

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