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From waste to resource recycled lime sludge: Sustainable low clinker cementitious binder, a comprehensive study on hydration, strength of concrete

G Murali ^{a,b,*}, Leong Sing Wong ^{a,b}, V.R Ramkumar ^c, Sallal R. Abid ^d, S Karthik ^e

^a Department of Civil Engineering, College of Engineering, Universiti Tenaga Nasional, Jalan IKRAM-UNITEN, 43000, Kajang, Selangor, Malaysia

^b Institute of Energy Infrastructure, Universiti Tenaga Nasional, Jalan IKRAM-UNITEN, 43000, Kajang, Selangor, Malaysia

^c Institute of Wood Science & Technology (IWST), Indian Council of Forestry Research & Education [ICFRE], India

^d Department of Civil Engineering, Wasit University, Kut, 52003, Iraq

^e School of Civil Engineering, SASTRA Deemed University, Thanjavur, 613401, India

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ABSTRACT

This research focuses on recycling lime sludge as a calcium carbonate (CaCO_3) source in producing a low-clinker cementitious binder and other supplementary cementitious materials. The study comprises three main phases. In the first phase, raw lime sludge collected from the textile industry was analyzed for pH, metal content, anions, and microstructure characterization. The second phase involved recycling and blending lime sludge with other materials to create a low-clinker binder. Various physical properties of the blended cement, consistency, setting time, fineness, and soundness, were examined. The third phase concentrated on understanding these cementitious system's hydration mechanisms, strength development, and microstructure evolution. Notably, the study found that incorporating recycled lime sludge into the binder components resulted in desirable physical and chemical properties. The 15% recycled lime sludge blend with 30% calcined clay promotes the ideal hydration reactions necessary to enhance concrete performance. Moreover, it demonstrated the positive influence of recycled lime sludge on cement properties and hydration products. The specimen, consisting of 45% calcined clay, demonstrated the highest compressive strength at 28 days, followed by the C2 specimen, comprising 30% calcined clay and 15% recycled lime sludge. The strength improvement is primarily attributed to the formation of additional hydrated calcium silicate, a key product of hydration that precipitates within the aqueous voids, contributing to developing a more compact structure.

1. Introduction

The increasing global population has driven the demand for infrastructure, leading to a significant increase in cement usage, second only to water [1]. However, this surge in cement production has resulted in a substantial rise in carbon dioxide (CO_2) emissions, and the cement sector has emerged as the second most significant source of anthropogenic CO_2 emissions [2], accounting for approximately 8% of global emissions [3]. The primary source of these emissions (about 95%) is the calcination of limestone to produce cement clinker [4]. Hence, it is crucial for the cement sector to urgently address its energy consumption and take measures to mitigate

* Corresponding author. Department of Civil Engineering, College of Engineering, Universiti Tenaga Nasional, Jalan IKRAM-UNITEN, 43000, Kajang, Selangor, Malaysia.

E-mail addresses: murali@uniten.edu.my, murali_220984@yahoo.co.in (G. Murali).

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CO₂ emissions during the production process. To tackle this environmental challenge, there is an increasing demand for innovative and sustainable cementitious binders that can substitute a portion of traditional cement clinker with waste materials sourced from diverse industries. Some viable and economically feasible waste materials include bottom ash, fly ash (FA), ground granulated blast furnace slag, dredged sediments, mine wastes and other industrial byproducts [5]. This shift toward alternative materials aims to significantly reduce CO₂ emissions from cement manufacture, but the development of feasible technologies is urgently required [6,7].

Common waste disposal methods in India include composting, ocean dumping, and landfilling. Cordeiro et al. (2016) [8] found that sugarcane bagasse ash can be incorporated into cement and lime mortar, exhibiting a pozzolanic reaction and filler effect. Maheswaran et al. [9] and Modolo et al. [10] studied lime sludge (LS) as a substitute for cement in concrete and mortars. They concluded that substituting up to 10% of cement with LS is suitable, as further increases led to a substantial decrease in compressive strength. Studies by Kumar et al. [11] and Brás et al. [12] explored using FA and LS as cement substitutes, consistently finding that LS negatively impacted compressive strength. Hiroyuki [13] analyzed textile dyeing factory sludge obtained from Tirupur, India, for physico-chemical characteristics and heavy metals. Solidification/stabilization with Portland cement was studied as a treatment technique, with results showing decreased mechanical characteristics as the percentage of sludge in construction increased. In summary, research has indicated that LS can serve as a partial substitute for cement, being effective for up to 10% replacement in concrete and mortar; its excessive use reduces compressive strength. Further research is essential to explore its broader applications in the cement and construction industries.

Research suggests CaCO₃ (calcium carbonate) significantly enhances low-clinker cement production [14]. Carboaluminate production in limestone-Portland cement blends consumes C₃A and continues even with aluminosilicate-based supplementary cementitious materials (SCM) [15]. Carboxyaluminates have been observed in metakaolin-limestone and FA-limestone mixtures [16,17]. Replacing 1/3rd of cementitious paste volumes with limestone fines improves concrete strength and durability but may require more superplasticizer [18].

Portland limestone cement with up to 35% limestone prevents ettringite conversion and promotes carboaluminate hydrates [19]. Higher limestone concentrations reduce setting time and consistency [20]. Limestone is often used to enhance aluminate-rich SCMs [21]. Sahu et al. [22] examined a two-mortar type with four LS binders. Binder 1 consisted of 70% FA and 30% LS, while Binder 2 had the same components but with the addition of 1% gypsum. The inclusion of gypsum improved compressive strength by accelerating the pozzolanic reaction. Another study [23] explored replacing 5%, 10%, and 15% of cement with LS, 15% FA and 5% metakaolin in a quaternary cementitious mixture. This resulted in improvements of 12%, 13%, and 9% compressive strength of the cementitious material. Ramkumar et al. [24] noticed a reduction in compressive strength when substituting LS at 10% or higher levels in the quaternary system. Limited studies have examined cement mortar and concrete's mechanical properties and durability that

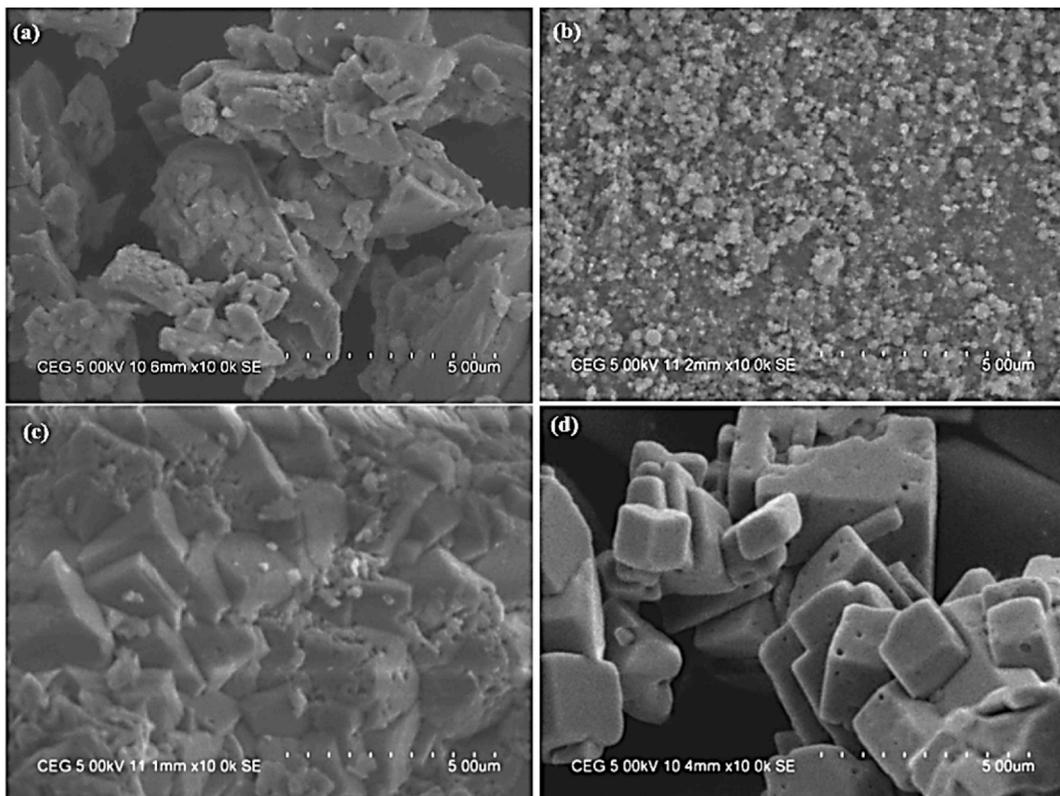


Fig. 1. SEM images (a) CC, (b) silica fume, (c) LS and (d) RLS ([26] License number: 5701191019805).

incorporate recycled LS with different SCM combinations.

This study holds significant potential for understanding the unique properties of LS and facilitating its effective use as a cementitious binder. The article investigates the potential of substituting clinker with SCM materials like LS (as CaCO_3), calcined clay (CC), and silica fume (SF) in cement production. Initially, a physicochemical process was employed to convert LS into CaCO_3 . Both raw and recycled lime sludge (RLS) were studied for their physical and chemical properties to assess the treatment's effectiveness. Subsequently, blended cement was produced by inter-grinding clinker, gypsum, SF and CC. The resulting blended cement underwent analysis for setting time, consistency, compressive strength, and hydration products. This research presents a novel and practical approach to utilizing LS in large quantities to produce CaCO_3 . It could also benefit nearby cement factories by providing a source of CaCO_3 from lime waste generated by the paper and textile industries.

2. Experimental study

2.1. Materials

- The clinker samples employed in this research were obtained from Ultratech Cements Pvt Ltd's Reddipalayam Plant, with Fig. 1a displaying the SEM image of the raw clinker.
- Zone II Ennore sand meeting the requirements of IS: 383–2016 [25] was used as the fine aggregate had an absorbency in the water of 0.6, a specific gravity of 2.67, a fineness modulus of 2.90, and a bulk density of 1811 kg/m^3 .
- The production of CC involved the static calcination of kaolinitic clay (with a presumed kaolinite content of around 60%) in a muffle furnace at 800°C , followed by a soaking time of 30 min.
- Calcined clay, gypsum, and SF were obtained from the Astra chemicals, and the SEM image is presented in Fig. 1b. The chemical properties of other raw materials are also presented in Table 1.
- Sludge samples were systematically collected from three distinct Common Effluent Treatment Plants (CETP), chosen based on the scale of their operations within the textile industry. The LS sample was taken after the effluent's hardness had been neutralized with calcium hydroxide and sodium carbonate. LS collected in this process is bagged and maintained in the CETP's facilities. For the sake of the subsequent analytical research, the obtained samples of LS were sun-dried to reduce the moisture content, then crushed, then wholly mixed, and last sieved through a size of $425 \mu\text{m}$ – ASTM 40 (Mesh 40). The cone and quartering procedure collected just 5 kg from each 500-kg sample for chemical analysis. The SEM image of LS and RLS is shown in Fig. 1 c and d, respectively. LS has been named recycled and raw LS (mixed sludge from all CETP).

2.2. Characterization of lime sludge

Many studies have focused on analyzing the chemical composition of textile sludge, primarily identifying metals, anions, or metal oxides using techniques like X-ray fluorescence spectrometry (XRF), X-ray diffraction (XRD) ion chromatography, atomic absorption spectrometry, and inductively coupled plasma-optical emission spectrometry. However, it is challenging to definitively determine the presence of metal oxides in textile sludge, and estimating the content of metals and anions may not comprehensively characterize the sludge's composition. Some scientists have explored the incineration of textile sludge to transform it into oxides suitable for manufacturing products such as tiles, paving blocks, bricks and even cement substitution [13,24,27]. The current method for handling textile sludge is time-consuming, inefficient, environmentally harmful, and economically unviable. It obscures the sludge's true chemical composition and poses contamination risks. Factors like sludge composition, bonding, treatment methods, and sampling techniques impact sludge properties. Poor sampling and analysis strategies have led to inaccurate scientific evaluations. This research addresses these issues by selecting a suitable sample collection technique based on literature insights. It also intends to scientifically analyze textile sludge's complete composition using analytical tools like XRF, XRD, and Thermogravimetric analysis (TGA).

2.2.1. The pH of samples

Samples were suspended in de-ionized water and given a good stir for 30 min before their pH was measured. Afterwards, the solution was filtered using grade 1 Whatman filter paper once the suspended particles had settled. The samples were subjected to drying in a hot air oven at 105°C until they reached a consistent weight, which allowed the moisture content to be determined using the standard weight loss method in conformity with ASTM D 2216–98 [28] and it is expressed in Eq (1).

$$\text{Moisture content (\%)} = \frac{\text{Weight loss (g)}}{\text{Sample weight (g)}} \times 100 \quad (1)$$

Table 1
Properties (Physical and chemical).

Constituents	CaO (%)	SiO ₂ (%)	Al ₂ O ₃ (%)	Fe ₂ O ₃ (%)	MgO (%)	Na ₂ O (%)	LOI (%)	IR (%)	SO ₃ (%)	Specific gravity
Clinker	63.85	21.49	5.40	4.25	1.12	0.11	0.70	0.53	1.02	–
LS	51.53	0.43	–	–	3.64	–	42.46	0.65	0.25	2.4
CC	0.06	54.96	39.75	4.16	0.02	0.18	0.24	–	0.1	–
Gypsum	32.12	1.06	0.08	0.20	0.60	0.01	20.15	–	45.62	2.3
SF	3.12	87.67	2.25	2.48	0.64	0.58	1.01	–	–	2.2

IR: Insoluble residue.

2.2.2. Metals and anions

Metals may be present in textile sludge because they are frequently added as dyes, pigments, mordants, and other auxiliary chemicals during the textile-processing stage. The metal content of LS was determined by subjecting powdered sludge to aqua regia (1:3 of HNO₃ and HCl) and heating it on a hot plate until practically dry. The contents were allowed to cool to ambient temperature before being dissolved in deionized water and filtered by Whatman filter paper of grade 42. The filtrate was prepared in a 100 ml amount. Inductively coupled plasma and an optical emission spectrometer were used to evaluate this filtrate for the existence of different metals in the textile sludge. Anions present in the textile sludge were estimated using a sample created using material equivalent to 10 times the weight of the dry, powdered textile sludge. This solution was mixed for 10 min using a magnetic stirrer and it was filtered through Whatman filter paper, grade 42 (USEPA 300.0). The filtrate obtained was analyzed for chloride and sulfate ions using Mohr volumetric titration against silver nitrate and turbidimetric method (USEPA 9038).

2.2.3. Identification of compounds using FTIR

The functional groups of the compound in the textile sludge have been studied using the Fourier transform–infrared spectroscopy (FTIR) method using JASCO 4700. The FTIR spectrometer in the wavenumber range of 4000–400 cm⁻¹ at a spectral resolution of 4 cm⁻¹, accumulating 40 scans. In this regard, potassium bromide (KBr) and all the samples were ground well to avoid scattering of infrared rays and dried overnight at 105 °C. The pellet was prepared by the solid dispersion of mixing of KBr and sample using a hydraulic press with a force of 5 kg/cm². The measured thickness of the pellet was roughly 0.5 mm and the diameter was 13 mm.

2.2.4. Identification of compounds using XRD

The dried and powdered sludge samples were subjected to XRD analysis to detect the compounds present in them. The XRD analysis was conducted using Bruker X-ray diffractometer operated at 40 kV and 30 mA with Cu K–alpha radiation in the 5–80° 2θ range. The peak positions in the obtained diffraction patterns of the sludge samples were matched manually. To confirm the presence of Ca(OH)₂ in the textile sludge samples were heated up to 620 °C for 3 h using the furnace to remove hydroxide content from the sludge altogether. The resulting residues of the samples were subjected to XRD analysis and the difference in the pattern from the original samples was compared.

2.2.5. Estimation of compounds using volumetric method

At the end of softening of effluents, calcium carbonate is precipitated due to calcium hydroxide and sodium carbonate addition to the effluents. In order to estimate the calcium carbonate amount existing in the sludge sample, the complexometric ethylenediaminetetraacetic acid (EDTA) titration was carried out at a pH 12 and using murexide as an indicator.

2.2.6. Estimation of compounds thermogravimetric method

The samples from the sludge were taken for the TGA method to estimate the compounds present in the textile sludge. The thermogram was obtained from TA Q50 thermogravimetric analyzer in the room temperature range of 800 °C at a heating rate of 10 °C/min under a nitrogen atmosphere and the change in weight of the sample as a function of temperature was calculated.

$$\text{CaCO}_3(\%) = \frac{\text{Weight loss } (\%)}{\text{Molecular weight of CO}_2} \times \text{Molecular weight of CaCO}_3 \quad (2)$$

$$\text{Ca(OH)}_2(\%) = \frac{\text{Weight loss } (\%)}{\text{Molecular weight of H}_2\text{O}} \times \text{Molecular weight of Ca(OH)}_2 \quad (3)$$

The Ca(OH)₂ amount present in the textile LS sample was estimated using the carbonation method. For this study, the sludge sample with Ca(OH)₂ in its composition was purged with carbon dioxide gas and completely converted into CaCO₃. The resulting sample was subjected to TG analysis, and the changes in the weight loss were considered and calculated as the Ca(OH)₂ content present

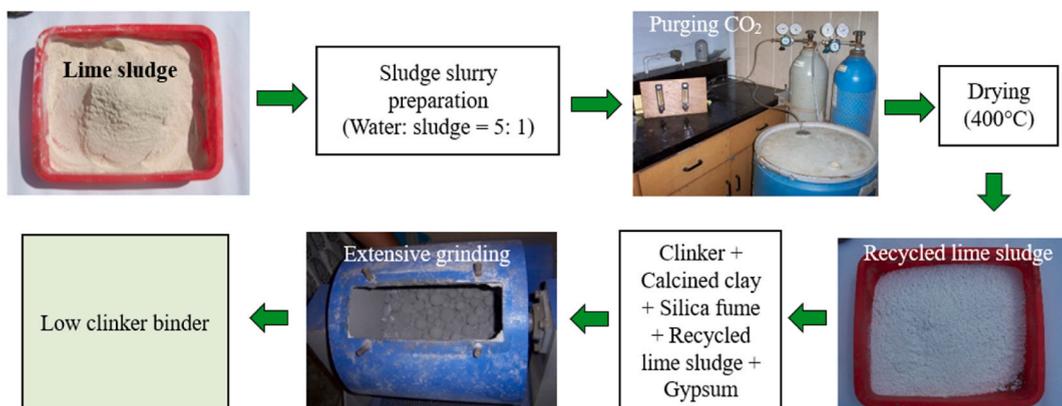


Fig. 2. Manufacturing process of sustainable low-clinker cementitious binders (Reproduced from Refs. [26,30] License number: 5701120429289).

in that particular sludge sample.

2.3. Treatment lime sludge

Previous research studies indicate that the creation of hydration products like CSH, CAH and other hydration products are hindered by contaminants like $\text{Ca}(\text{OH})_2$ found in raw LS [9,13]. Treating the raw LS before blending it into the clinker and other SCM materials is necessary and its procedure is shown in Fig. 2. LS collected from different CETPs must be mixed thoroughly to make a homogenous mix using machine mixing. Homogenous ground mixed sludge is mixed with water to make up slurry 5:1 (Sludge: Water). Subsequently, the slurry preparation was made that provides an effective medium of CO_2 mixing with CaO, which can be ensured. Also, the formation of CaCO_3 can be facilitated in water with minimum residual time and pressure of 2 Pa [29]. After a residual period of 2 h, the slurry is subjected to filtration and then dried under the sun. Subsequently, the RLS is kept in a furnace for drying. This was done to remove CaOH and other free and bound water during drying up to 400°C , ensuring the effective grinding of RLS during ball milling. RLS was checked for chemical analysis to check its purity.

2.4. Preparation of low clinker binder and mixing combination

Producing hydration products like CSH and CAH is challenging due to impurities, such as $\text{Ca}(\text{OH})_2$, present in the raw LS [9,13]. It must be recycled before adding the raw LS to the clinker and other SCM components. The primary step involves combining finely ground mixed sludge with water to create a slurry with a ratio of 5 parts sludge to 1 part water. The slurry preparation was conducted to provide a medium that effectively facilitates mixing CO_2 with CaO, ensuring its efficacy. Additionally, the precipitation of CaCO_3 can be enhanced in aqueous environments characterized by short residence times and a minimum pressure of 1 Pa. Following a period of 2 h, the slurry undergoes filtration and subsequent sun-drying. Once the LS has been processed, it is stored in a dryer until completely dry. The subsequent stage involved the elimination of CaOH and other forms of unbound and bound water by a drying process conducted at temperatures up to 400°C . This procedure is crucial to efficiently grinding the RLS. Effective grinding is essential in the production of blended cement, and this process can be improved by removing free water from raw materials. Lab-scale ball milling is used to finely grind all the blended raw ingredients, reducing the severity of the impact of the steel balls and producing finer cement particles. The raw materials underwent a grinding process utilizing a ratio of 1:7 between the raw materials and steel balls. To achieve optimal grinding efficiency in mass manufacturing, it is recommended to use a precise ratio. In this experiment, we varied the proportions of the raw components to create three distinct mixtures. The clinker percentage varied from 50% to 95%, CC from 15 to 45%, SF from 15 to 45%, LS of 15%, and gypsum from 2.5 to 5%. The cement mixtures were labelled C to C6, and their detailed mixing combination is demonstrated in Table 2.

2.5. Physico-mechanical studies

IS:4031:1988, Part 4,5 [31] was used to analyze cementitious binders for their setting time and consistency. Vicat's apparatus was utilized to quantify the level of consistency. The soundness test was conducted using the Le Chatelier method in accordance with the IS:4031:1988, Part 3 [32]. The Blaines fineness was performed as per the recommendation of IS:4031:1988, Part 2 [33]. Compression tests were conducted on 50 mm cubical specimens as per the IS: 4031:1988, Part 6 [34]. After the compression tests, samples were collected from the failed specimens for crushing before the microstructure analysis.

2.6. Specimens preparation

The water-to-binder (w/b) ratio 0.4 was used to prepare all mixtures. The binder and fine aggregate are combined and blended during the mixing process for 1 min. The mixing process continued for 5 min after the inclusion of the coarse material. The SP-mixed water was added gradually to achieve a uniform mixture. The fresh mixture was used to prepare the 100 mm cubical specimen. The mixing combination adopted in this study is demonstrated in Table 3.

3. Discussion of results

3.1. pH and moisture content

The pH and moisture content of the raw and recycled LS samples are demonstrated in Table 4. The pH value ranged from 9.38 to 9.72, indicating the marginal difference between the three samples. This may be due to the similar treatment process involved in all the CETPs. The observed moisture content was high in CETP 1 sample (10.61%) and low in CETP 2 (4.09%). The difference in moisture content was due to variables in drying time. However, to have a homogeneous mix, all the collected LS were mixed to yield raw LS.

Table 2
Mixing proportions of cement mortar specimens.

Mix No	Clinker (%)	CC (%)	LS (%)	SF (%)	Gypsum (%)
C	95	–	–	–	5
C1	65	15	15	–	5
C2	50	30	15	–	2.5
C3	50	–	15	15	5
C4	50	–	15	30	2.5
C5	50	45	–	–	5
C6	50	–	–	45	5

Table 3
Mixing proportions of concrete.

Mix Id	Fine aggregate (Kg/m ³)	Coarse aggregate (Kg/m ³)	w/b ratio	SP (%)
C	801	1112	0.4	0.6
C2	801	1112	0.4	0.8
C5	801	1112	0.4	0.8

Table. 4
pH value and moisture content of collected samples.

Properties/sludge	CETP1	CETP2	CETP3	LS	RLS
pH value	9.72	9.33	9.38	9.31	9.42
Moisture content (%)	10.61	4.09	5.48	6.54	2.16

After CO₂ purging treatment, the RLS was also analyzed for pH and moisture content.

3.2. Results of metals and anions

The amount of metals and anions present in the RLS samples is demonstrated in Table 5. The possibility exists for synthesizing compounds of both anions, namely chloride and sulfate, with all the metals in the sludge samples as their composition. However, due to their solubility in water, the dissolved content ran with water during the plant's treatment process. Among these compounds, chlorides of copper (I), chromium (III) and, nickel (II) and sulphates of barium (II) and strontium (II) are insoluble in water and they may present in the composition of the sludge. This result also showed that the metallic elements and anions present in the sludge were quantitatively very low and the contribution of these elements in the composition of LS is almost negligible (less than 1% of mass). According to the classification of the Ministry of Environment and Forest Notification (MoEF) (India), Hazardous Waste Management Rule 2008, this small amount of contamination of both metals and anions present in the LS will never pose any threat to the environment during the disposal, storage and in the utilization. The results suggest that heavy metals and other impurities in RLS are less than 1% by mass. Hence, using RLS in cement and concrete may be strongly recommended as a disposal method and alternative resource/forcement-based industries for producing blended cement using RLS.

3.3. Results of FTIR

Fig. 3a displays the FTIR spectra obtained from various samples of RLS. The band obtained in the range of 713–721 cm⁻¹ and 1796 and 1797 cm⁻¹ was due to the symmetric vibrations of the CO₃²⁻ group, and the band obtained in the range of 872–876 cm⁻¹ and 1429 to 1446 cm⁻¹ were due to the asymmetric vibrations of CO₃²⁻ group. A strong, sharp band occurring in the range of 3694–3696 cm⁻¹ was the specific characteristic of the O–H bond; this may be due to moisture or water molecules in the sample. This finding substantiates the hypothesis that the LS sample contains CO₃²⁻ and HO⁻ groups. From Fig. 3b, it was observed that the band in the range of 3647–3700 cm⁻¹ belongs to the symmetric stretching of the OH⁻ group, and this indicates that the presence of water molecules, which may come from Ca(OH)₂ is confirmed. The unique finding of this study indicates that LS contains CO₃²⁻ and OH⁻ in the system along with CaO. The presence of OH⁻ group in the form of Ca(OH)₂ hinders the hydration process and reduces the strength of cement, as studied by researchers [9,13].

3.4. XRD analysis of CETP

The XRD pattern of the raw LS is shown in Fig. 4. It can be seen that the phases in the sludge were calcium carbonate (CaCO₃). It was evidenced by the 2θ values of 23.09°, 29.44°, 35.95°, 39.40°, 43.17°, 47.53° and 48.50° for LS collected from CETP 1. Similarly, the values were 23.07°, 29.44°, 35.97°, 39.42°, 43.17°, 47.57° and 48.54° for LS collected from CETP 2 and 23.15°, 29.48°, 36.48°, 39.48°, 43.23°, 47.65° and 48.60° for LS collected from CETP 3. These values were matched with JCPDS Card No: 83–0577 and the 2θ value of 18.57° and 47.06° present in the sludge collected from Arulpuram CETP confirmed that the presence of calcium hydroxide (Ca(OH)₂) at JCPDS Card No: 72–0156 for sludge collected from CETP 1. This calcium hydroxide pattern presents only in LS from CETP 1, indicating that the lime (Ca(OH)₂) added during the effluent in the treatment process was not reacted completely due to its less solubility in water. The peaks identified at other 2θ angles were almost negligible and rejected the possibility of other compounds identified earlier by the ICP – OES method. After the ignition of all the samples up to 620 °C, it was found that the missing peaks at 2θ values of 18.57° and 47.06° in the sludge collected at CETP 1 were shown in Fig. 4. The remaining peaks were precisely matched with the patterns of the other sludge. This study confirms that the textile sludge sample from CETP 1 contains both Ca(OH)₂ and CaCO₃ in its

Table 5
Minor constituents present in textile LS (mg/kg).

Metals	Na	K	Fe	Mn	Ni	Zn	Ba	Sr	Li	As	Cu	Cr	Al	Cd	Cl ⁻	SO ₄ ²⁻
CETP 1	2210	668	110	2	BDL	0.2	BDL	0.6	4	0.9	130	190	220	20	11,982	4704
CETP 2	2230	395	160	168	19	142	96	29	9	0.4	130	190	220	20	5140	1872
CETP 3	1960	774	210	10	7	91	66	18	65	1.5	130	190	220	20	2694	5280

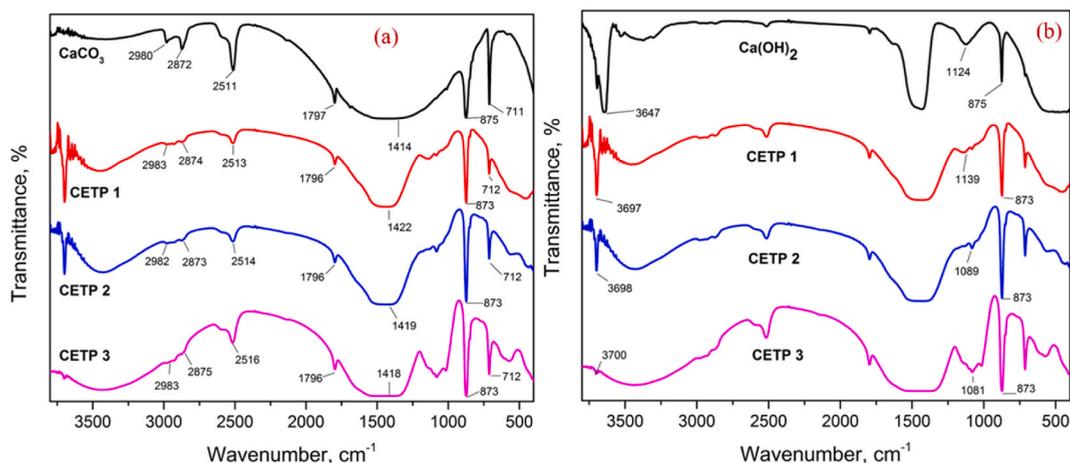


Fig. 3. FT-IR spectra for textile LS samples (a) compared with CaCO_3 and (b) compared with Ca(OH)_2 .

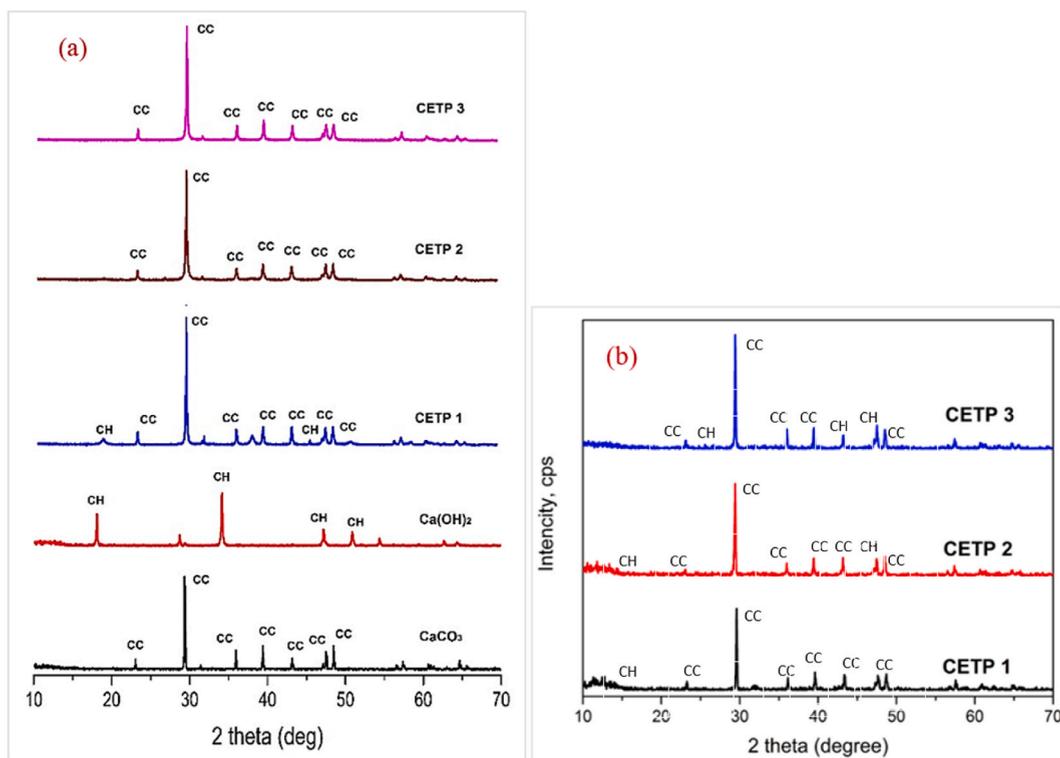


Fig. 4. XRD patterns for textile LS samples (a) before heating (b) heating up to 620 °C.

composition. The complexometric titration result reveals that the total calcium in the textile LS samples of CETP 1, CETP 2 and CETP 3 were 26.06, 34.70 and 29.64 % (as Ca), respectively. Most commonly, researchers have reported that the percentage of all the calcium content obtained was converted to an equivalent of calcium carbonate. However, calcium carbonate is not required to account for all calcium in the sample. The lime softening reactions clearly explained that the precipitate has calcium carbonate as a significant quantity. The XRD and FTIR explained that there were possibilities for calcium hydroxide and calcium carbonate in the textile LS. This is due to the poor solubility of calcium hydroxide, which was added during the softening of effluents.

3.5. TGA analysis of CETP

Fig. 5 displays the thermogram for the sludge, together with the calcium carbonate and calcium hydroxide. Calcium carbonate was confirmed to be the primary ingredient in the textile LS by a large amount of weight loss seen between 620 and 795 °C, which was caused by the emission of CO_2 gas. The thermogram of the sludge sample showed a curve between 300 and 400 °C, which was

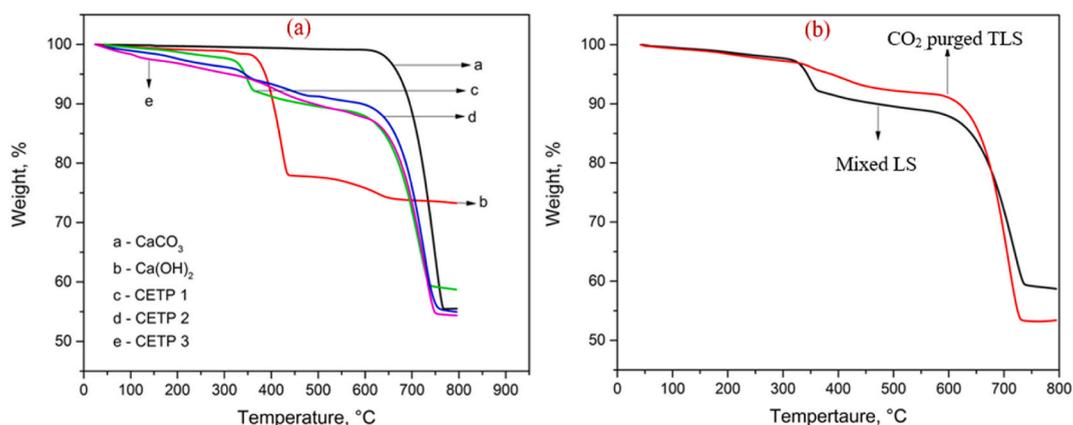


Fig. 5. (a) Thermogram for LS and (b) Comparative thermogram of sludge from CETP 1 and its CO₂ purged.

attributed to the dehydration of calcium hydroxide, the same as was seen for pure Ca(OH)₂. This result verifies the presence of Ca(OH)₂ in LS obtained solely from CETP 1. As XRD did previously, TGA reveals that calcium hydroxide was indeed positively found at CETP 1. The increase in CaCO₃ between 321 and 368 °C could be attributed to factors other than the existence of calcium hydroxide, as it is not explicitly stated that the only source of CaCO₃ is the presence of calcium hydroxide.

The weight loss between 606 and 795 °C is approximately 31.0%, as the thermogram produced from the CETP 1 sludge sample shows. After purging CO₂ gas through that sludge sample, the percentage rises to 42.50%, with a corresponding weight loss differential of 1.50%. This measures the actual Ca(OH)₂ concentration in the sludge sample. The amounts of Ca(OH)₂ and CaCO₃ were determined using the provided formula from the weight loss measurements. The quantity of CaO and Ca(OH)₂ present in the sludge were nearly 55.50 % and 1.50%, respectively, in the CETP's as demonstrated in Table 6.

4. Properties of cement

4.1. Consistency

The standard consistency of cement prepared with different combinations of blended cement from gypsum, RLS, SF and CC is presented in Table 7. C2, C4 and C6 samples exhibited the highest standard consistency value (32%), while others had a marginal difference ranging from 29 to 31%. When SF, CC, and RLS were included in the paste, the water content needed to attain a normal consistency was increased marginally. The higher consistency was recorded in three samples (C2, C4 and C6). The C2 sample contains 15% RLS with 30% CC exhibiting a 32% consistency. The C4 sample showed the same behavior, containing 15% RLS and 30% SF. Conversely, adding 45% SF without RLS showed the same consistency (32%).

4.2. Setting time

The initial and final setting times of blended samples are shown in Table 7. It is observed that the highest initial and final setting time lengthened by the C2 samples was about 158 and 284 min, respectively. This is due to the combined effect of CC (30%), LS (15%) and lower clinker content (50%). Adding CC to the sample (C5) up to 45% shortened the initial setting time of 141 min. The C4 sample containing a 30% SF and 15% LS resulted in 155 and 262 min of initial and final setting time, respectively. Increasing the SF content to 45% in the C6 sample reduced the setting time (131 and 255 min) compared to the C4 sample. The effects of SF on the time it takes cement paste to set were examined by Rao [35]. Initial setting time was shown to reduce with increasing SF concentration. The decreased ingredients barely impacted the cement paste setting time. Nevertheless, large SF contents drastically reduced the initial setting time. SF does not appear to affect the final setting time. SF's pozzolanic effect appears most prominent in the first few hours after hydration [35]. On the other hand, the constant LS of 15% in C1–C4 samples displayed a marginal difference in setting time between these samples. Therefore, the effect of RLS in setting time is minimal, irrespective percentage of CC and SF added to the samples.

A small quantity of RLS (5%) intergrinding with clinker comprising 5% of gypsum reduced initial and final setting time [36]. The

Table 6
TGA results.

Sample Name	Temperature range (°C)	Weight loss (%)	CaO (%)	Ca(OH)
CETP 1	321–368	–	–	–
	606–795	31.00	42.10	13.00
CETP 2	321–478	–	–	–
	640–795	29.50	41.00	14.50
CETP 3	624–795	28.50	39.00	15.50
	627–795	42.50	55.50	1.50
RLS – CO ₂				

Table 7
Physical properties of cement.

Properties	C	C1	C2	C3	C4	C5	C6
Standard consistence (%)	29	30	32	30	32	31	32
Initial setting time (minutes)	133	144	158	141	155	141	131
Final setting time (minutes)	260	275	284	280	262	276	255
Soundness (mm)	5	4	3	3	2	2	2

clinker type had a greater impact on the result of the LS inclusion than blended cement's fineness. Tsivilis et al. [37] conducted a study in which they manufactured limestone cement by incorporating limestone inclusions, ranging from 5% to 35%, into a mixture of clinker and 5% gypsum. Findings indicated that the setting time was not drastically diverse from the control without limestone. Application of LS powder as a substitute for pozzolanic cement was studied by Heikal et al. [38]. According to the research, limestone powder shortened the initial and final setting times and decreased the total porosity. Nevertheless, it was noted that the limestone increased the levels of free lime and combined water. The minimum first setting time required by the ASTM C109-16 standard [34] is 45 min. Nevertheless, in practical applications, it is typically preferable to have an initial setting time of not less than 90 min. A highly lengthy final setting period is typically avoided due to the potential for high formwork costs.

4.3. Soundness

Soundness test was performed to characterize cement paste samples that do not manifest cracks, collapse, or other imperfections arising from a large-volume transformation. The expansion results of the Le-Chatelier experiment are graphed with the corresponding samples depicted in Table 7. The high soundness value was recorded from the C sample (5 mm) containing no CC, SF, and LS. However, adding CC and LS decreased the soundness value to 3 and 4 mm. Additionally, adding CC and SF independently without LS made the recorded soundness value 2 mm. The noted behavior can be attributed to the active involvement of SCM and RLS in the hydration process, resulting in additional CSH and other hydration products from CaOH within the cement system. It is clear from Table 7 that the SF and LS combination displayed a lower soundness value, while the behavior is the same for the independent addition of SF and CC into the samples. As per the Indian standards [39,40], the recommended value of a soundness test was 10 mm. In this study, all samples exhibited soundness values less than 6 mm. Since no eruptive reaction product was found, the logical conclusion is that the CC, LS, and SF were highly non-expandable.

4.4. Particle size distribution

It is well recognized that the component's particle size distribution influences the qualities of cementitious materials because it affects the water demand, void volume, and rheology. It is crucial to acknowledge that if RLS is interground with clinker, the Blaine fineness of the resulting binder is typically greater compared to pure Portland cement. This is ascribed to the comparatively softer nature of RLS, which facilitates easier grinding. Adding CC up to 30% with 15% RLS to the C1 and C2 samples resulted in 315 and 396 m^2/kg Blaine's fineness, which results higher than the control sample (C) (Fig. 6 a). Adding a SF of 15 and 30%, and 15% RLS increased Blaine's fineness to 417 and 512 m^2/kg for the C3 and C4 samples, respectively. The highest value of Blaine's fineness was recorded from the C6 sample, which contains 45% SF. It is clear from the above discussion that the fineness of the binder was detected in the samples comprising SF.

By increasing the mass percentage of RLS added to the clinker from 0% to 15%, Tsivilis et al. [41] demonstrated the necessary increase in specific surface area (from 260 to 366 m^2/g) to achieve the same setting characteristics and strength gain. Schmidt et al. [42] presented that the surface area needed to achieve the same voids content, packing density and consumption of water for RLS cement increased by a similar amount. The particle size distribution of the seven binders used is depicted in Fig. 6 b. Based on the recommendations and findings of other researchers, intergrinding was used for effective output. Our research indicates that the D_{50}

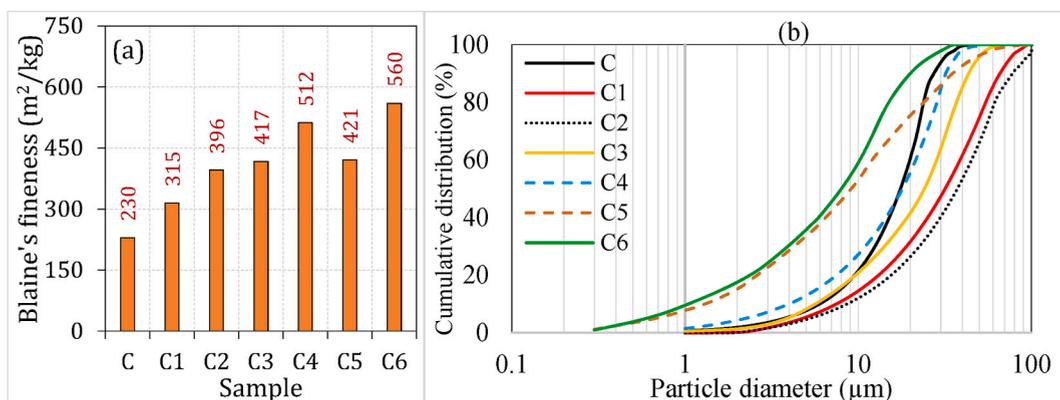


Fig. 6. (a) Fineness and (b) particle size distribution.

particle size ranges from 7 to 15 μm was recorded for all binders. Finely grinding RLS led to reduced water need, decreased bleeding, enhanced workability, and increased strength due to particle gradation within the cement [42]. Adding 15%–20% RLS can improve packing density; the effect is less noticeable in finer cement. RLS is typically simpler to grind than clinker; consequently, it makes up the bulk of the smaller particles when combined during the grinding process, expanding the particle size distribution. The RLS must be finely ground before use, as coarser powder could lead to more significant gaps and more air pockets in powder particles. This phenomenon can lead to heightened water absorption and diminished strength [43]. Bentz [44] points out environmental and financial gains from including RLS in cement grinding. However, it is further postulated that limestone grinding may have additional advantages if it is done separately to attain a relatively larger particle size than intergrinding. The limestone is ground to a less fine consistency, which requires a comparably lower amount of energy, and is then combined with Portland cement of a suitable size.

4.5. Compressive strength

The compressive strength of specimens at the age of 5 different days is depicted in Fig. 7. Combining CaCO_3 as recycled LS with pozzolanic compounds and conventional organics enhances their strength significantly. Nevertheless, the increase in strength observed in lime-based binders is characterized by a gradual progression and necessitates a longer duration. The compressive strength values ranged from 29.1 to 36.3 MPa for 3 days, from 44.20 to 50.6 MPa for 7 days, from 55.3 to 62.8 MPa for 28 days, from 63.20 to 69.10 for 56 days, and from 65.4 to 71.2 MPa for 90 days. From Fig. 7, the C specimen's compressive strength at 28 days was 55.30 MPa. Adding CC and RLS to the C1 and C2 samples exhibited an 8.68 and 13.02% improvement compared to the C specimen. A similar trend was observed at 56 (4.27 and 8.07%) and 90 days (6.27 and 8.87%). This is due to adding 15% RLS and up to 30% CC. However, adding 45% of CC into the specimen (C5) resulted in a 13.56% greater compressive strength than the C specimen. The strength development primarily depends on the formation of hydrated calcium silicate, the main hydration product. This compound precipitates within the aqueous voids, leading to the creation of a denser structure. The initial formation of hydrates, which have significant binding forces, can be responsible for this phenomenon or the subsequent transformation of these hydrates into other hydration products with weaker binding forces [38].

On the other hand, adding SF together with RLS exhibited a positive result in compressive strength. For instance, the C3 and C4 specimens exhibited a 4.70 and 11.03% enhancement in compressive strength compared to the C specimen at 28 days. An enhancement was observed by about 2.53 and 6.65% for 56 days and 3.06 and 6.42% for 90 days. The C6 specimen containing 45% of SF exhibited a 12.48, 7.28, and 7.19% increase in strength for the 28, 56, and 90 days, respectively. A silica-rich hydrated layer forms from the rapid interaction between SF and lime. Because of its inherent instability, the layer above rapidly transforms into C–S–H gel. The elimination of free lime during the curing period typically takes place between 7 and 28 days due to the remarkable reactivity demonstrated by SF particles. The crystallinity of the resulting C–S–H gel is dramatically increased during this reaction [45].

4.6. XRD analysis of cement mortar sample

Fig. 8 depicts the X-ray diffraction (XRD) analysis performed on the three different types of binders used in the concrete composition. The C2 and C5 samples comprising LS and CC exhibited notable ettringite peaks throughout the hydration process and the B1 sample. Aluminate phases originate in the CC and react with sulfate ions in the control binder and $\text{Ca}(\text{OH})_2$ to generate additional ettringite; other carboaluminate hydrates are responsible for the observed phenomena. The formation of carboaluminates, namely hemicarboaluminate (Hc) and monocarboaluminate, was observed after a curing duration of 28 days in samples C2 and C5. This process is facilitated by substituting sulfate ions with carbonate ions, which are introduced by incorporating LS and CC. The synthesis of carboaluminates phases involves the chemical interaction of LS and aluminate hydrates, while adding CC can supply aluminate phases. As the curing time increased, incorporating CaCO_3 as a supplementary material into the carboaluminate phases reduced the peak intensity of calcium hydroxide (CH) at $29.5^\circ 2\theta$. This finding demonstrates that aluminates and silicates in LS might form supplementary hydration products, enhancing the hydration process. Moreover, it is crucial to recognize that the continuous blended cement hydration process in C sample resulted in an increased generation of $\text{Ca}(\text{OH})_2$, as indicated by the amplified peak intensity detected at $18.1^\circ 2\theta$. Also, it is worth mentioning that the peaks associated with Mc in samples C2 and C5 displayed a greater intensity

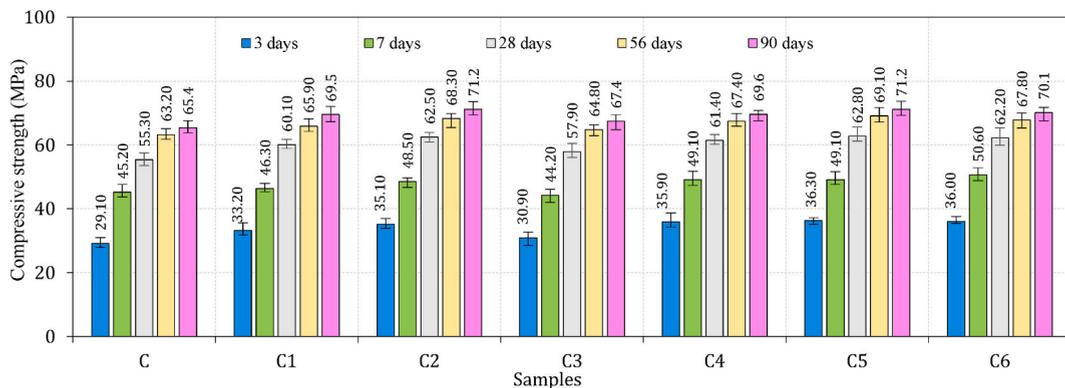


Fig. 7. Compressive strength of specimens.

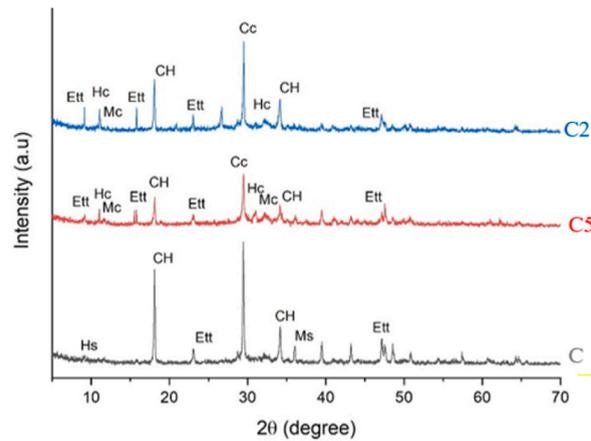


Fig. 8. XRD analysis for the best mixtures (a) C (b) C2 and (c) C5.

when compared to the C sample. The observed phenomenon can be attributed to the creation of Mc by incorporating CO_3 into monosulfate [46].

4.7. TGA analysis of cement mortar sample

Fig. 9 presents the results of Differential Thermal Analysis (DTA) and TGA, including data on the weight loss and differential weight loss of the selected samples. The initial mass reductions characterized by endothermic peaks occurring between temperatures of approximately $80\text{--}200\text{ }^\circ\text{C}$ indicate the removal of structurally bound water within the C-(A)-S-H phase and other hydrates in all examined specimens. The thermal decomposition of the hydrated substance calcium hydroxide ($\text{Ca}(\text{OH})_2$) was found in all samples at temperatures ranging from $400\text{--}450\text{ }^\circ\text{C}$. The thermal breakdown of calcium carbonate (CaCO_3) within the $600\text{--}700\text{ }^\circ\text{C}$ temperature has been thoroughly characterized in C5. This is attributed to the higher concentration of unreacted CO_3 originating from LS in comparison to C2. Moreover, the utilization of TGA and DTA techniques confirms the existence of ettringite, carboaluminates, monosulfate, calcium hydroxide ($\text{Ca}(\text{OH})_2$), and many other hydrates within the blended binders B2 and C5. The observed breakdown peaks align with the XRD findings, indicating that they are caused by the reaction between the material and aluminates and CO_3 originating from SCM.

4.8. SEM analysis of cement mortar sample

The SEM images of collected samples are depicted in Fig. 10. After a duration of 28 days, the sample exhibited the existence of C(A)SH, which was discerned through the observation of aggregated, needle-shaped formations that appeared to be covered with delicate layers of metal (Fig. 10). The binder system of C2 and C5 samples may have included aluminum derived from CC, which might potentially explain the observed morphological disparity between the three samples. The introduction of aluminum from CC into the blended binder system is the reason for the morphological distinctions between C and C2, leading to the formation of larger and more extended chain structures in C(A)SH, as shown in Fig. 10b. The inclusion of LS and CC in the mixture led to the initiation of additional hydration reactions. As a consequence, the pore size of the matrix decreased and its density increased after 28 days compared to the C sample [46,47]. The C5 samples exhibited minimal quantities of carboaluminates characterized by a morphology resembling plates

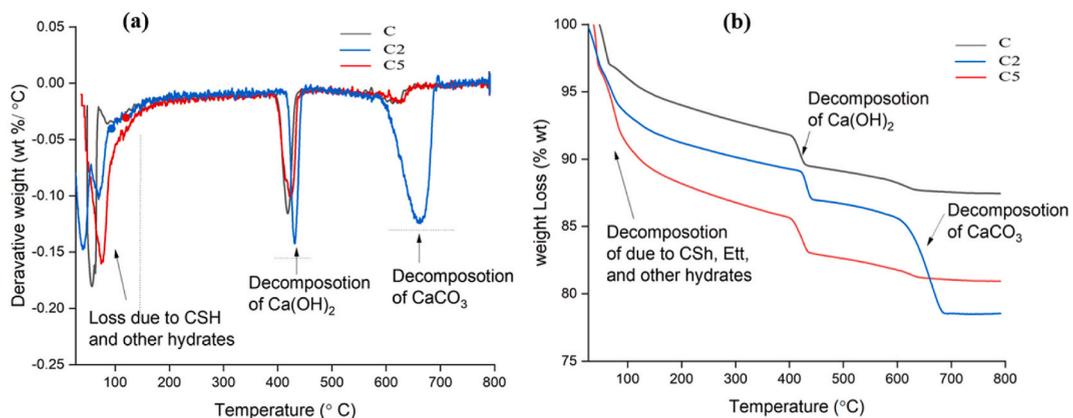


Fig. 9. TGA analysis for the best mixtures (a) C (b) C2 and (c) C5.

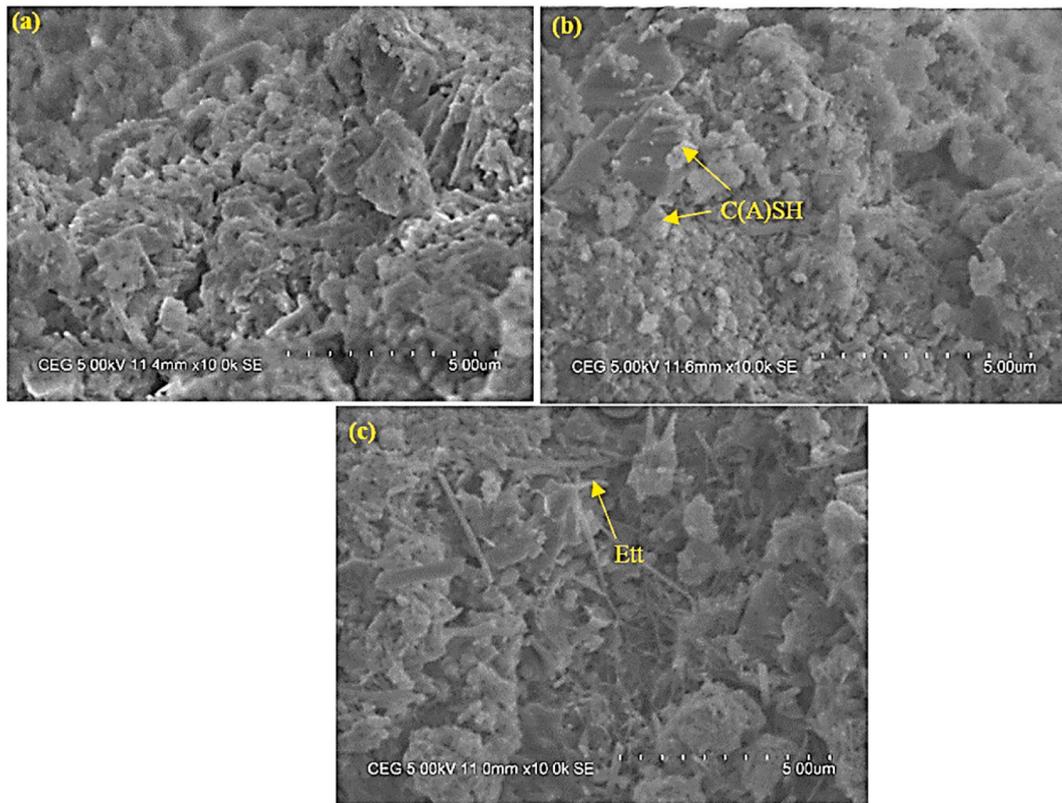


Fig. 10. SEM analysis of samples (a) C, (b) C2 and (c) C5.

(Fig. 10c). The diminished crystallization pressure and increased density of ettringite and carboaluminates (Hc and Mc) are accountable for their lessened efficacy in mitigating volume alterations and higher packing efficiency in the C2 and C5 compared to the C specimens.

5. Discussions on mechanical properties of concrete

5.1. Compressive strength of concrete

The compressive strength of specimens containing three different binders is revealed in Fig. 11. The recorded compressive strength of the C specimen was 48.91 MPa. Adding 30% of CC and 15% of LS to the C2 specimen improved the strength up to 54.80 MPa with a percentage improvement of about 12.05% compared to the C specimen. Incorporating CC and LS in blended cement leads to the consumption of portlandite and subsequently facilitates the synthesis of pozzolanic (C-A-S-H). Furthermore, differing dissolution rates between LS and CC introduce a heightened level of intricacy in comprehending the impact of the pozzolanic reaction, particularly regarding the properties of C-A-S-H. The properties of the C-S-H are influenced by its composition, leading to changes in stiffness, distribution characteristics of the hydrates, packing density and morphology. The effectiveness of the pozzolanic C-A-S-H in decreasing the capillary porosity, reducing permeability and increasing compressive strength is a notable advantage in the blended cement system. Increased strength and faster reaction times depended on particle size and surface area. Adding LS increases the C-S-H gel in the blended cement concrete mix, accounting for the increased compressive strength observed in the C2 specimen. Increasing the LS to 30% and reducing a CC to 15% in C5 specimens exhibited less strength compared to the C2 specimen. For instance, the noted strength decrement for the C5 specimen was 8.77% compared to the C2 specimen. This behavior is due to the 30% content of LS in concrete. Xu et al. [48] reported that using LS less than 15% exhibited a better performance in cement properties. Kumar and Prasad [49] reported that the 10% LS addition into the concrete is optimum for excellent compressive strength.

6. Conclusions

This research aimed to understand better the hydration process and the strength of low-clinker cementitious binders, which included LS as a source for CaCO_3 , CC, and SF in blended cement systems. The LS was characterized and recycled before being combined with other binder ingredients and concrete's compressions behavior. Based on the research, several significant conclusions were derived as follows.

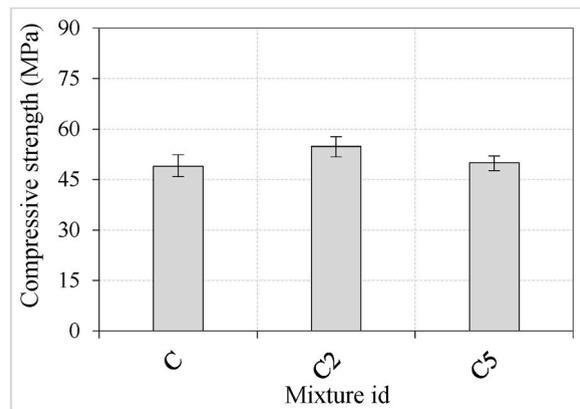


Fig. 11. Strength of concrete under compression.

- i. Quantitatively, the LS contained only trace amounts of metallic elements and anions, and their contributions to LS's overall composition are minimal (less than 1% by mass). The LS contains CO_3^{2-} and OH^- in the system, along with CaO . The hydration process is slowed, and cement strength is diminished when an OH^- group, such as $\text{Ca}(\text{OH})_2$, is present. However, the formation of hydration products like C-S-H , Ettringite, C-A-S-H and other hydration products can be activated in the blended cement system by treating the LS, which could enhance its performance. In purging CO_2 gas through the sludge sample, the observed percentage increases to 55.50%, accompanied by a weight loss differential of 1.50%, determining the exact amount of $\text{Ca}(\text{OH})_2$ in LS. RLS contained 51.10–55.50% CaO and 1.50 % $\text{Ca}(\text{OH})_2$, respectively.
- ii. The pH values ranged between 9.38 and 9.72, suggesting a marginal difference among the three samples, potentially attributed to the similar treatment processes employed in all CETPs. Notably, the moisture content was elevated in CETP 1 sample (10.61%) and comparatively lower in CETP 2 (4.09%). The variance in moisture content can be attributed to differences in drying time. Nonetheless, in order to achieve a uniform mixture, all collected LS samples were combined to produce raw LS.
- iii. The highest compressive strength at 28 days was exhibited by the C5 specimen, comprising 45% CC, followed by the C2 specimen, comprising 30% CC and 15% RLS. The enhancement of strength primarily relies on forming additional hydrated calcium silicate as the primary outcome of hydration, which precipitates within the aqueous voids to create a more compact structure. Also, adding LS promotes the formation of additional carbo aluminate hydrate in the system through physical and chemical effects. This phenomenon can be explained by forming hydration crystals from the previously formed hydrates with powerful binding forces.
- iv. Adding 15% RLS and 30% CC in C2 binder-based concrete specimens exhibited the highest compressive strength of 54.80 MPa compared to C and C5 specimens. Increasing the LS content to 30% in the C5 specimen, a decreasing trend of compressive strength was observed.
- v. Introducing aluminum from CC into the blended binder system is responsible for the morphological distinctions between C and C2. This process leads to larger and more extended chain structures in $(\text{C}(\text{A})\text{SH})$. The incorporation of LS and CC in the mixture prompted additional hydration reactions. Consequently, after 28 days, the matrix exhibited a reduction in pore size and increased density compared to the control (C) sample.

CRediT authorship contribution statement

G Murali: Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Leong Sing Wong:** Writing – review & editing, Writing – original draft, Validation, Formal analysis. **V.R Ramkumar:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Sallal R. Abid:** Writing – review & editing, Writing – original draft, Investigation, Formal analysis, Conceptualization. **S Karthik:** Writing – review & editing, Writing – original draft, Validation, Investigation, Formal analysis.

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

Data will be made available on request.

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