Recent Progress in Newer Cementitious Binders as an Alternative to Portland Cement: Need for the 21st Century



Rajesh Kumar

Abstract This article reviews about different kind of newer alternative cements to ordinary Portland cement (OPC), which can reduce energy requirements and greenhouse gas emission. Some of these novel binders can be produced using low-grade industrial waste materials and fuels. The practical feasibility of these alternative binders can only be justified after comprehensive investigation of different physicomechanical, microstructural, and durability attributes. In the presented study, nine prominent alternative cementitious binder systems, i.e., calcium sulfoaluminatebelite (CSAB) cement, alkali-activated cement (AAC), reactive belite-rich Portland cement (RBPC), magnesium oxides-based (MgO) cement, Belite-ve'elimite-Ferrite (BYF) cement, carbonatable calcium silicate cements (CCSC), limestone calcined clay cement (LC³), calcium sulfoaluminate (CSA) cement and calcium hydro silicatebased cement (Celitement), are discussed along with the raw materials required for synthesis, phases formed, required sintering temperature, etc. Along with the above, their current position has been depicted as compared to conventional OPC binder system. It was inferred that all the newer cementitious binders can be developed using industrial wastes such as-low grade limestone/clay, fly ash, and slags with attainment of the desirable physico-mechanical and durability properties along with cost and energy reduction by 25–55%. Development of the above alternative binder also leads to the reduction of greenhouse gases (GHGs) by 15-50%.

Keywords Calcium sulfoaluminate–belite cement · Reactive belite-rich Portland cement · Geopolymer · Calcium sulfoaluminate cement / Low carbon cement

Department of Civil Engineering, Indian Institute of Technology Delhi, Delhi 110 016, India

R. Kumar (🖂)

Organic Building Materials (OBM) Group, CSIR—Central Building Research Institute, Roorkee, Uttarakhand 247 667, India

e-mail: rajeshkumar@cbri.res.in

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1 Introduction

Globally, concrete is the second most widely used man-made materials after water. The major causes of wider adoptability of concrete are superior strength, long-term durability, and fire resistance at lower cost as compared to another similar kind of binder materials. For the production of concrete, major resources are natural aggregates, i.e., sand, stone, and water. Nevertheless, to bind fine and coarse aggregates as a heterogeneous mix, an essential ingredient is OPC, which reacts in the presence of water. Worldwide, about two billion tons (BT) per year of cement is being produced by cement industries. However, the manufacturing process of OPC emits a very large amount of carbon dioxide (CO_2) which covers almost 7% of the total greenhouse gas emission.

Generally, OPC is the most widely used binder material. One ton of OPC clinker is produced by taking 1.7 T of limestone, sand, clay, and iron slag. But, the increasing cost and scarcity of pure mineral materials have signaled the cement industries to explore newer cementitious binders. It is now being emphasized to use different kinds of cements based on their effect on workability, physico-mechanical, and durability properties. The major chemical compositions of OPC are CaO, Al_2O_3 , Fe_2O_3 , and SiO_2 , which are replaced with newer chemical oxides, and thus form new cementitious phases (apart from C_3S , C_2S , C_3A , and C_4AF) during newer cement production.

2 New Generation Binders

i. Calcium Sulfoaluminate-Belite (CSAB) Cement

CSAB cement is produced by sintering fly ash, gypsum, and limestone at temperature of about 1200–1250 °C which is less than the conventional sintering temperature for OPC (1450 °C). After sintering, CSAB cement makes three phases, i.e., Ye'elimite phase $(C_4A_3\check{S})$ (35–70%), dicalcium silicate $(\beta$ -C₂S) (< 30%) phase, and "ferrite" phase (10-30%). Ye'elimite phase contributes to early age strength development (in place of Alite), while β -C₂S phase provides long-term strength. As compared to OPC, the developed CSAB cement saves energy up to 25% with reduction of limestone quantity and CO₂ by 60 and 20%, respectively. CSAB clinker is more friable and softer than that of OPC clinker, which decreases the grinding energy. Due to the above advantages, CSAB cements are being produced in China for more than 35 years. Now a days, the industrial wastes such as Al-rich sludge, aluminum anodization sludge, bottom ash, Class C, and F-fly ash, coal combustion residuals, desulfurization gypsum, flue gas desulfurization sludge, fluidized bed combustion (CFBC) ash, and high-alumina fly ash were being utilized as primary raw materials to synthesis the CSAB cement. Limestone, gypsum (7%), low calcium fly ash, and phosphogypsum were used as raw material for CSAB cement [1]. These materials were heated at 1250 °C for 30 min, and after sintering, it was found that mainly two major phases



Fig. 1 CS of mortar cube specimens [3]

were formed, i.e., C_2S and C_4A_3S phases. Adolfsson et al. [2] sintered limestone, GGBS, basic oxygen/electric arc furnace slag, argon oxygen decarburisation, and ladle slag, at 1200 °C for approximately 30 min. After sintering; clinker phases, i.e., sulphoaluminate, C_4A_3S , etc., were formed. Jewell et al. [3] used limestone, bauxite, fluidized bed combustion (CFBC) ash and fly ash. These raw materials were heated at a temperature of 1250 °C. It was noticed that for CSAB cement, at w/c ratio of 0.48, compressive strength (CS) at 56 days was 40.1 MPa (Fig. 1), while, for OPC, CS at 56 days was 42.6 MPa at w/c ratio of 0.43. Chen and Juenger [4] heated limestone, bauxite, desulfurization sludge, class C fly ash, coal combustion residuals at temperature of 1250 °C for 12 h. It was found that at w/c ratio of 0.45, CS at 28 days for CSAB (fineness of 324 m²/kg) and OPC (fineness of 403 m²/kg) was 46.2 and 44.5 MPa, respectively (Fig. 2).

Ma et al. [5] developed CSAB cement by sintering limestone, high-alumina fly ash, desulfurization gypsum (15%) at a temperature of 1250 °C for 30–150 mints. It was noticed that CSAB cement (fineness of 357 m²/kg) showed 28 days CS of 50.8 MPa at w/c ratio of 0.38, while 28 days CS for OPC cement was 53.2 MPa at w/c ratio of 0.50 (Fig. 3). Da Costa et al. [6] manufactured CSAB cement at a temperature of 1250 °C for 30 min, by using limestone, bauxite, SiO₂, gypsum, aluminum anodizing sludge (AAS). Figure 4 depicted the observed results. It was noticed that eco-clinkers produced (CSAB-Bx/AAS and CSAB-AAS) showed higher CS after 28 days as compared to control CSAB cement (CSAB-Ref.).

Da Costa et al. [7] produced CSAB cement using limestone, bauxite, aluminum anodization sludge at temperature range of 1250-1350 °C. It was depicted that optimum sintering temperature was about 1250 °C to avoid decomposition of phases with sulfur compound and their related SO₂ emissions. Rungchet et al. [8] used hydrated lime, Class-F fly ash, Al-rich sludge, and desulfurization gypsum to produce CSAB cement at temperature of 1150 °C with soaking time of 1 h. It was noticed that OPC and CSAB paste developed CS of 60.5 and 41.5 MPa at w/c ratio of 0.45



Fig. 2 CS of PC and MS/MC/MF kind of CSAB cement [4]



and 0.80, respectively. In another study by Shen et al. [9], CS of CSAB paste was reported as 38.5 MPa at w/c ratio of 0.50. In the study, CSAB cement was developed by giving secondary heat treatment to primary raw materials such as—limestone, bauxite, phosphogypsum, at 1100–1200 °C for 1 h. El-Alfi and Gado [10] used kaolin (25%), gypsum (20%), marble sludge waste (55%) to develop CSAB cement at temperature of 1200 °C for 1 h. The 28 days CS of CSAB cement paste was observed as 36.0 MPa at a w/c ratio of 0.50. In the similar manner, Rungchet et al. [11] produced CSAB cement by using hydrated lime, Al-rich sludge and desulfurization gypsum, Class-F fly ash and bottom ash. The heating was done for 1 h. at temperature of 1050 °C. It was noticed that CSAB cement paste developed 28 days CS of 41.0–43.5 MPa.

ii. Alkali-Activated Cement (AAC)

AAC or popularly known as geopolymer cement is low carbon cementitious binder which contains higher amount of aluminosilicates phase. Aluminosilicates phase consists higher amount of amorphous content which gets activated in alkaline medium and forms 3-D polymeric structures. After gaining maturity, AAC develop superior load-bearing ability and excellent durability/environmental performance as compared to conventional OPC ((Li et al. [12]; Shi et al. [13]). To develop AAC, different kinds of industrial by-products/supplementary cementitious material (SCMs) can be used along with alkaline solution and silicates (Li et al. [12]). The chemical activation of fly ash, in presence of alkali, is depicted in Fig. 5.

The descriptive model/steps for alkali activation is shown in Fig. 6.

In the long term, AAC binders showed better durability and good modulus of resilience as compared to conventional OPC (Naqi and Jang [14]). However, the cost of production (per m³) of AAC material is calculated as 25–30% higher than that of OPC binder. Therefore, due to cost constraint, AAC materials are not commonly used cementitious binder in the building infrastructure. Based on composition of primary raw materials, majorly, five kinds of AAC are available, i.e., slag-based/pozzolan/lime-pozzolan-slag/calcium aluminate blended/Portland blended—AA cements (Shi et al. [13]). During the development of AAC binder, most sensitive parameter is curing temperature as it affects the activation energy of AAC binder matrix. It was found that due to increased rate of reaction with the increment in curing temperature from 40 to 95 °C, CS was developed more quickly (Khale and



Fig. 5 Alkali activation of fly ash (Shi et al. [13])



Fig. 6 Steps for alkali activation of aluminosilicates (Shi et al. [13])

Chaudhary [15]. Jang et al. [16] found that AAC showed the ability to immobilize heavy metals in stabilized products with desirable CS.

iii. Reactive Belite-rich Portland Cement (RBPC)

RBPC, also known as high belite cement (HBC), is often considered as family of OPC binder and contains more than 40% of belite content and less than 35% of alite content. RBPC contains lower alite to belite ratio (Gartner and Sui [17]). As C_3S synthesis requires more consumption of specific energy and CO_2 emission than the synthesis of C₂S Phase. Therefore, the production of RBPC requires lesser specific energy, and 10% lower contribution to greenhouse gas (Scrivener et al. [18]). The lime saturation factor (LSF) decreases from 100 to 75%, which reduces energy requirement by 12% and reduction in CO₂ emission by 6% (Figs. 7 and 8). To develop RBPC, similar kind of raw materials is used (with less limestone) as that for OPC, but clinkering temperature is maintained at 1350 °C, i.e., 100 °C lower than OPC. Therefore, low-grade kiln fuels can also be used. To form reactive belite in the clinker, 0.5–1.0% SO₃ is often added. RPBC showed similar 28 days CS as that of OPC and even higher at later ages (Sui [19]). RBPC depicted lower heat of hydration (HOH) as compared to OPC as cumulative HOH of belite was measured as half that of alite (Taylor [20]). In China, HBC cements are used in number of construction projects for over 15 years.



iv. Magnesium Oxides-based (MgO) Cement

Eco cement, as reactive MgO cements (RMC), has been manufactured first in Tasmania. MgO cements are found in two chemical forms, i.e., magnesium carbonate and magnesium silicate. When hydration of MgO cements take place, then magnesium carbonate based MgO cement forms magnesium hydroxide (also known as brucite), while magnesium silicate-based MgO cement form magnesium silicate hydrates. Further, brucite can form magnesium carbonate, after carbonation process. MgO is produced after heating (pyro-processing) magnesium carbonate at dissociation temperature of about 800–1000 °C, which can be recarbonated further as per the anticipated theory. The carbon emission was noted less for magnesium silicate cements because no chemical bound CO₂ was emitted when silicates were heated (Lawrence [21]). The main binding phases formed were 2Mg(OH)₂·MgCl₂·4H₂O, 3 Mg(OH)₂.MgCl₂.8H₂O, and 9Mg(OH)₂.MgCl₂.H₂O (Maravelaki-kalaitzaki and Moraitou [22]). Hay and Celik [23] concluded that RMC can be used as CO₂ sequestration material and can show comparable CS as compared to OPC.

v. Belite-Ye'elimite-Ferrite (BYF) Cement

BYF cement is also known as Belite–calcium sulfoaluminate ferrite (BCSAF) cement, which contains three phases, i.e., C_2S , Ye'elimite/calcium sulfoaluminate (C_4A_3S) and ferrite/calcium alumino-ferrite/brownmillerite (C_4AF). Out of these three phases, C_2S and C_4A_3S phases are considered as major phases (Naqi and Jang

[14]). The most reactive phase is considered as belite, followed by Ye'elimite and ferrite. This is an intermediate technology which falls between traditional OPC technology and CSA cement technology (Gartner and Sui [17]). Dienemann et al. [24] observed that belite and ferrite phases can be replaced with "ternesite" (C_5S_2 \$, sulfate spurite) phase. The following reactions during the hydration of BYF cement were noted (Gartner [25]).

When AH₃ is available-

$$C_2S + AH_3 + 5H \rightarrow C_2ASH_8$$
(strätlingite) (1)

At later ages-

$$C_2(A, F) + C_2S + C_2ASH_8 \rightarrow 2C_3(A, F)SH_4(katoite)$$
(2)

$$2C_2S + 7H \rightarrow C_3S_2H_6 + CH \tag{3}$$

$$2C_2S.CS + 7H \rightarrow C_3S_2H_6 + CH + CS \tag{4}$$

BYF cement was made to reduce the production cost of CSA cement with lower carbon footprint as compared to OPC (Gartner [26]). The BYF cementitious binders are expansive than OPC. Because all the raw materials need for the manufacturing of OPC are available near OPC plant. However, BYF cements require extra aluminum rich material, which is transported from distant sites. Therefore, the cost of production of BYF cement increases. However, less energy is required to produce per unit of BYF clinker (www.aether-cement.eu [27]).

vi. Carbonatable Calcium Silicate Cements (CCSC)

Calcium silicate (Ca₂O₄Si) clinkers can be produced using low-lime Ca₂O₄Si minerals like wollastonite (CaSiO₃, CS). These types of clinkers required lime up to 40%, while OPC requires CaO content up to 70%, which results decrement of CO₂ emission by 30% (Atakan et al. [28]). CCSC requires low clinkering temperature, i.e., 1200 °C which is 250 °C lesser than that is required for OPC manufacturing. Therefore, this kind of cement consumes less amount of fuels, and thus lower greenhouse gas emission. The produced cement clinker is hydrated in CO₂ gas environment at controlled temperature and relative humidity (RH). The CCSC cement develops higher CS even at 24 h of curing irrespective to OPC which develops desirable CS at 28 days. CCSC consumes less water because it captures water which is evaporated during curing process (Gartner and Sui [17]). This kind of special concrete can be used only for cement products without reinforcement due to the typical curing procedure adopted (Scrivener et al. [18]; Gartner and Sui [17]), which lowers down the pH of concrete mass up to 9.0.

vii. Limestone Calcined Clay Cement (LC³)

As shown in Fig. 10, the LC^3 is a recently developed low carbon cement that is synergically developed by intergrinding limestone (15%) and calcined clay (30%) with kaolinite content ranging from 40 to 70%, OPC clinker (50%) and gypsum (5%). The developed cementitious binder have about 50% low clinker factors and showed similar CS than that of conventional OPC binder (https://www.lc3.ch/the-material/ [29]). Apart from the hardened properties, the durability properties of LC^3 cement have found superior in chloride and sulfate exposure condition as compared to OPC. Mishra et al. [30] performed the hydration study at curing temperatures of 27 and 50 °C; for OPC, composite cement (CC), LC³–5%G and LC³–8%G cementitious binders. As shown in Fig. 9, it was found that for CC, $LC^3-5\%G$, and $LC^3-8\%G$ binders, HOH after 24 h., was insignificantly increased at 50 °C than that at 27 °C. It was found that DoH of LC³ binder at 50 °C was less as compared to that that at 27 °C from 7 day onward (Fig. 10). However, at curing temperature of 27 °C, DoH for OPC and LC³ binders was almost same at 28 days. At 28 days and curing temperature of 50 °C, DoH for OPC was 78%, while lowest DoH was observed for CC binders (67%).

Mishra et al. [30] did the quantification of hydration products for OPC, CC, LC^3 ($LC^3-5\%G$ and $LC^3-8\%G$) cementitious binders, at curing temperatures of 27 and 50 °C (Fig. 11). It was concluded that at 50 °C, carboaluminate phases (hemicarboaluminate (Hc) and monocarboaluminate (Mc)) were not observed for $LC^3-5\%G$ and $LC^3-8\%G$ systems. For $LC^3-5\%G$ binder, Aft content was reduced with time due to its conversion into "alumina, ferric oxide, monosulfate" phase (Afm). Mishra et al. [30] showed the effect of higher curing temperature on the porosity of OPC, CC, LC^3 ($LC^3-5\%G$ and $LC^3-8\%G$) pastes hydrated up to 28 days, as shown in Fig. 12. It was concluded that overall porosity and the pore entry diameter was increased with high curing temperature of 50 °C. For OPC and CC mixes, the pore



Fig. 9 HOH of composite cement [30]





Fig. 10 DoH of different binders cured at 27 and

Fig. 12 Porosity graph at 28 days [30]

50 °C [30]

entry diameter was largely increased. The overall porosity for $LC^3-5\%G$ was higher than other mixes, while $LC^3-8\%G$ mix has comparable porosity with the OPC and CC binder mixes due to increased formation of Aft phase.

Pillai et al. [31] confirmed that the carbon footprint of LC³ concrete was lower than that of OPC concrete with same 28 days CS. Nguyen et al. [32] concluded that additional calcium-rich phases in LC³ cement delayed the ASR gel formation. Yang et al. [33] performed the numerical simulation for chloride diffusion in LC³ binder system and concluded that LC³ concretes contain more pore tortuosities even at higher water-binder ratio, than fly ash concretes. Mishra et al. [30] conducted the quantitative backscattered image analysis for LC³ (LC³–5%G and LC³–8%G) binders at different curing temperatures. A clear ring of hydration product (probably, C–A–S–H) was observed around the grains of LC³–5%G specimens (Fig. 13). It was further analyzed through experimentation that intermixing of other hydration products with C–A–S–H was higher in LC³ systems at higher curing temperature. Through BSE-EDX analysis, it was concluded that for LC³ specimens at high curing temperature of 50 °C, there was a significant increase in the Al/Si ratio due to increase in quantities of Aft and carboaluminate phases.

Life cycle analysis reveals that LC^3 production can reduce carbon footprint up to 30% due to lower clinker factor and save up to 50% limestone as compared to conventional OPC. Thus, the replacement of OPC clinker with limestone/limestone slurry and clay in LC^3 blends will curtail not only the production cost but also greenhouse gas/non-renewable energy impact.

viii. Calcium Sulfoaluminate (CSA) Cement

CSA cements were produced by China in late 1970s. Initially, this kind of cements was used to manufacture self-stressed concrete pipes (Habert [34]). CSA contains higher alumina content and produced by sintering bauxite, limestone, and gypsum in rotary kiln (Phair [35]). CSA cements contain 30% belite, 35–70% Ye'elimite and gypsum as major phases (Chatterjee [36]). The following reaction takes place during hydration of CSA cements (Older [37]):

Without calcium hydroxide (CH):



Fig. 13 SEM-BSE images of LC3-5%G cured at 27 °C (left) and 50 °C (right) at 28 days [30]

$$2C_4A_3S + 2CSH_2 + 36H \rightarrow C_6AS_3H_{32} + 2AH_3$$
 (5)

With calcium hydroxide (CH):

$$C_4A_3S + 8CSH_2 + 6CH + 74H \rightarrow 3C_6AS_3H_{32}$$
 (6)

During the production of CSA cements, thermal energy reduces up to 25% along with the reduction of CO_2 emissions by 20%, as compared to OPC. Different kinds of industrial byproducts can also be utilized in the production of CSA cements (Ambroise and Pera [38]). Thus, CSA cements can be considered as sustainable solution for future cement industries.

ix. Calcium Hydrosilicate Based Cement (Celitement)

Celitement binders were developed by the Karlsruhe Institute of Technology (KIT) and considered as a newer cementitious binder. The raw materials used are similar as that for OPC which is carbonates (limestone with 70% CaCO₃ fraction) and silicates (GGBS, fly ash, etc.) (Naqi and Jang [14]). The CaO/SiO₂ ratios were maintained in-between 1 and 2 (Stemmermann et al. [39]). To develop Celitement binder, two stage processes are adopted. In the first stage, all the raw materials are treated hydrothermally (150–200 °C) which produces α –C₂SH. In the second stage, produced α –C₂SH is blended together with silicate compounds which produces amorphous calcium hydrosilicates (Schneider et al. [40]). Stemmermann et al. [41] observed that Celitement binder containing mortar developed CS of 80 MPa at 28 days. However, the production process, to develop Celitement binder, is little bit complex as compared to OPC binder (Scrivener et al. [18]). Use of this kind of cement can reduce the carbon footprint up to 50%.

3 Comparison With OPC

Overall, it can be inferred that most critical parameter while developing or synthesis of any binder are—selection of desirable raw material, raw material proportion, clinkering temperature, fluxing agent (to reduce the sintering time and temperature) along with CO₂ reduction potential to meet sustainable development goals. Such as, for synthesis of OPC, only, calcareous (e.g., limestone, chalk etc.) and argillaceous (e.g., clay, shale etc.) raw materials are required. But, for special cementitious binders, as per the requirement of their physico-mechanical attributes, some other kind of raw feed kiln materials are also required. Such as, for CSAB development, gypsum is required in higher amount by 20% as compared to OPC. Fly ash can be used as conventional primary raw materials for CSAB development. While, during OPC manufacturing, fly ash is only be used as replacement of clay. Apart from the above, CSAB synthesis takes place at temperature of 1200–1250 °C which is 200–250 °C less as compared to OPC. Early age strength development takes place due to Ye'elimite phase in case of CSAB cement, while the same is caused by Alite phase

in OPC cement. In comparison to OPC, process and manufacturing of CSAB cement save energy up to 25%, along with reduction of limestone quantity and CO_2 by about 60 and 20%, respectively.

However, the development of AAC cement does not require sintering process and thus not an energy-intensive process as compared to OPC. In AAC cement, only, aluminosilicates are used as binder phases. Therefore, different kind of SCMs such as fly ash and slags can be used with alkaline solution and silicates. Typically, strength inversion takes place after increasing the curing temperature from 40 to 80 °C, while, in case of AAC cement, rapid development of CS takes place when curing temperature increases from 40 to 95 °C [15]. Similarly, RBPC/HBC binder can be developed with 15-20% lower amount of limestone and less sintering temperature as compared to OPC. By doing so, energy requirement reduces by 10-15% with CO₂ reduction of 6-10% [18]. Synthesis of MgO cement takes place at temperature of 800-1000 °C, i.e., 400-450 °C lower as compared to OPC. Strength development of MgO takes place with carbonation curing unlike OPC for which water curing is required for 28 days [21]. BYF cement contains belite as most reactive phase, while C_3A is considered as highly reactive phase for OPC cement [14]. CCSC binder develops at lower clinker temperature, i.e., at 1200 °C and requires lower lime contain up to 40% which is 30% lower than that of OPC. Apart from these parameters, CCSC cement formation requires less energy/fuels as compared to OPC. However, in case of LC^3 binder, typically, 50% of clinker is being used which is about 45% less as compared to OPC. Therefore, due to less consumption of limestone for production of LC^3 , CO₂ reduces by about 30%. In the similar way, CSA and Celitement cement reduce carbon footprint by 20 and 50%, respectively, as compared to OPC.

4 Conclusions

The presented review has discussed about the new cementitious binders as an alternative to OPC. Nine alternative cements have been discussed in details regarding their production process, required raw materials for synthesis, sintering temperature along with environment impact. Based on detailed literature review, few conclusions are as follows:

- 1. Some of the binders such as CSA cement and MgO cement can replace conventional OPC clinker.
- 2. The conventional raw materials and fuels, used to produce OPC clinkers, can be replaced fully or partly to develop newer cements.
- 3. CSAB cement can save energy up to 25% with reduction of limestone quantity and CO₂ by 60 and 20%, respectively.
- Compressive strength of alkali-activated slag-based cement has been found comparatively higher than OPC and also increased when curing temperature was increased from 40 to 95 °C.

- 5. RPBC binder showed similar 28 days CS as that of OPC and even higher at later ages.
- 6. 24 h CS of CCSC was 10–12% higher as compared to OPC binder.
- 7. Production of LC^3 can reduce carbon emissions up to 30% due to lower clinker factor and thus save up to 50% limestone than that of OPC.

Concerning the above, there is need to established cement standards and practical guidelines also, before the production of some of newer cementitious binders. Ultimately, to meet out the sustainability goals in cement production, the suitable techno-economic, strategic planning vision from industry owners are much needed in the current scenario.

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