



## Limemud resources in the Western Indian offshore: An alternative to high grade limestone and some viable proposals

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Several studies have been carried out by various authors on limemud that occurs in the western Indian offshore which is compositionally  $\text{CaCO}_3$ , amorphous, homogeneous, creamy white, soft and non sticky sediments mainly consisting of ooids. The chief mineral constituent of limemud is aragonite. The age, occurrence, formation and estimation of limemud resource are compiled from various published literatures and briefly presented. Bench scale beneficiation and agglomeration studies for limemud were conducted by Geological Survey of India at the lab of Indian Bureau of Mines and found limemud may be used in steel and filler industries, pharmaceuticals, food, cement etc. The physical and chemical properties of limemud suggest it may be a suitable raw material for the production of stone paper. There are several advantages in manufacturing stone paper in comparison with that of wood pulp paper. A brief comparison between the two compiled from the published literature is presented in the paper. The striking difference is in the consumption of water and emission of  $\text{CO}_2\text{e}$  from the production of wood pulp paper and that of stone paper. A difference of about 1000 kg  $\text{CO}_2\text{e}$  emission between the two helps to possess carbon credits. Sustainable mining technology and transportation of limemud from Gujarat offshore to nearby beach facilities are discussed. The approximate costs for limemud mining and transportation are estimated and compared with that of imported high grade limestone from UAE/Oman and is found to be much cheaper. Advantages of limemud and future propositions are briefly submitted for the consideration of the planners.

[**Keywords:** Carbon credit, Limemud, Pneumatic dredging, Stone paper]

### Introduction

Indian onshore is bestowed with about 200 billion tons of limestone resources of which about 1.6 billion tons are placed under reserve category. Limestone has several uses like cement, steel, filler, agriculture, pharmaceuticals, animal feeds, construction etc. Out of the total production of limestone in India, 97 % is of cement grade, 2 % is of iron & steel grade and 1 % is of chemical grade. India's industrial requirement for steel, blast furnace and chemical grade limestone are mainly met from imports<sup>1</sup>. In 2020, India imported limestone worth USD 370 million, becoming the first largest importer of limestone in the world<sup>2</sup>.

Limemud, compositionally  $\text{CaCO}_3$ , is amorphous, homogeneous, creamy white, soft and non sticky sediments mainly consisting of ooids followed by skeletal matter and mud aggregates with minor detritals. The chief constituent of limemud is aragonite with minor amounts of calcite, quartz, clay minerals etc. as revealed by XRD analysis and were first reported in the continental shelf edge and slope off Kachchh in 1993 by Vaz *et al.*<sup>7</sup>. On the basis of grain size, the silt sized ooids is termed as limemud

where as limesand is mainly composed of sand sized ooids (Fig. 1a – f)<sup>3</sup>. The internal structure of ooids is made up of aragonite needles (Fig. 1e and 1f)<sup>3</sup>. Hereafter, these sediments are collectively being called as 'limemud'. The limemud occurs mostly as basin fill deposits and the limemud bearing cores exhibit fining up sequences suggesting its deposition under suspension<sup>3</sup>. The age of limemud estimated by carbon dating is within a range between 11,000 and 18,000 (approx.) years and the corresponding sea levels were around 55 and 120 m of water depth during that period<sup>4</sup>. According to Bathurst<sup>5</sup>, limemuds are produced by several mechanisms: mechanical disintegration of biogenic skeletal components, disaggregation of calcareous green algae, inorganic precipitation, bioerosion, erosion of tidal-flat deposits and bio-geochemical processes<sup>5</sup>. The carbonate platform reported by several workers is the largest topographic feature on the northwestern outer continental shelf of India and is isolated from the coast by a huge clastic depocentre in which pro-delta sediments have been deposited since Eocene<sup>4,6</sup>. Limemud is deposited in this northwestern outer

continental shelf and also in the slope<sup>7</sup>. Though it occurs both along the east and west coasts of India, we discuss here the large resource of limemud occurring off Gujarat and Maharashtra states at water depths between 55 and 120 m within the western Indian offshore<sup>3</sup>. Vaz *et al.*<sup>7</sup> observed that CaCO<sub>3</sub> content varies from 71 – 77 % in the upper slope to 92 – 94 % on the shelf edge and Rao *et al.*<sup>4</sup>, reported 60 – 75 % CaCO<sub>3</sub> in limemud from continental slope and > 90 % at shelf break. Limemud occurs with varying thickness between 6 and 23 m. The thickness of overburden ranges from 0 to 3 m. Estimation of limemud resource has been carried out using digitized

sub-bottom profiles. These digitized profiles were validated using lithologs prepared from core samples. Quality wise fine tuning was also done by incorporating CaCO<sub>3</sub> content in limemud. Thickness rasters were created using digitized limemud layers and multiplied with area and bulk density to obtain limemud resource using ArcGIS software. Thus the inferred resource of limemud has been estimated by Geological Survey of India (GSI) about 72,000 million tonnes within 23,000 sq km area<sup>3</sup>.

### Materials and Methods

As a part of systematic seabed mapping and mineral exploration of GSI, samples are being collected at different grid intervals within Indian offshore. Limemud samples weighing about 300 kg were submitted to Regional Mineral Processing Laboratory of Indian Bureau of Mines (IBM), Bangalore for conducting bench scale beneficiation and agglomeration studies for limemud<sup>3</sup>.

### Results

The original limemud and limesand samples were separately assayed at IBM Lab. The assay results of bulk limemud and limesand samples are given in the Table 1. Mineralogical studies of the sample revealed that aragonite is the major mineral present in the sample, with subordinate amounts of calcite, quartz and feldspar. The samples were wet sieved at different mesh sizes and found that +200 mesh size fraction of limesand and -500 mesh size fraction of limemud are enriched in CaO and depleted in MgO, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, etc. (Table 1). Size separation of different mesh sizes

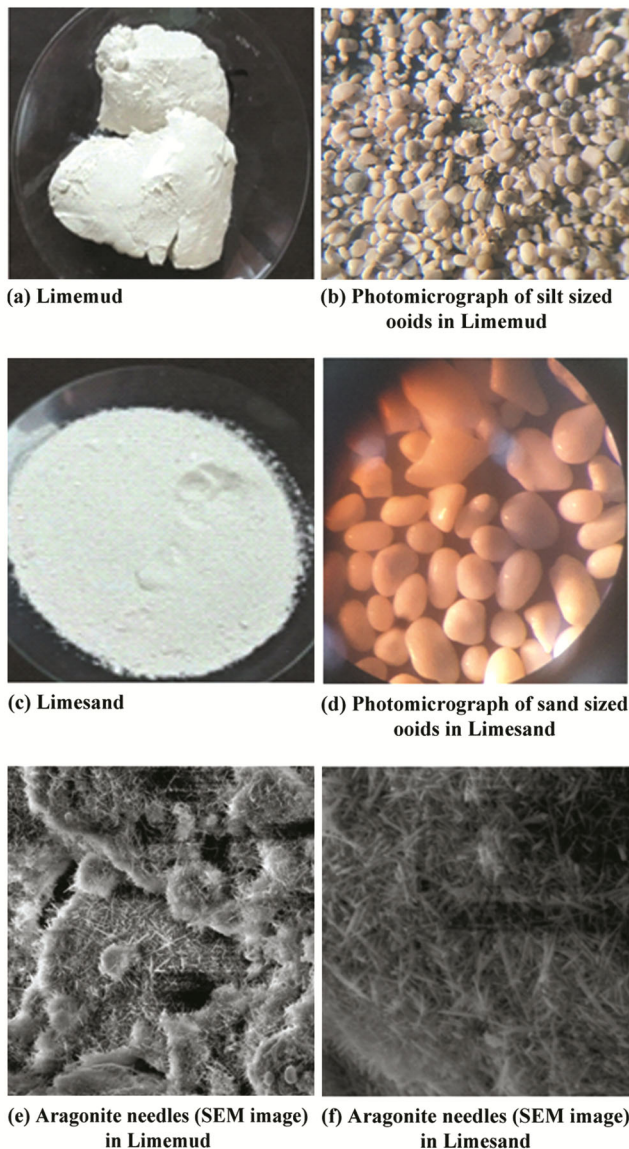


Fig. 1 — (a-f) Images of Limemud (Courtesy: GSI Brochure on Limemud)<sup>3</sup>

Table 1 — Chemical Composition (%) of limemud and limesand analyzed at IBM Lab

	Limesand			Limemud		
	Bulk	+200 mesh	-200 mesh	Bulk	+500 mesh	-500 mesh
CaO	49.73	51.15	49.69	48.62	46.36	51.98
MgO	0.7	0.5	0.47	0.71	0.77	0.49
Fe <sub>2</sub> O <sub>3</sub>	0.34	0.35	0.55	0.52	1.5	0.28
SiO <sub>2</sub>	3.76	2.95	5.08	4.08	9.37	2.69
Al <sub>2</sub> O <sub>3</sub>	0.55	0.47	0.89	0.74	1.22	0.41
K <sub>2</sub> O	0.06	nil	nil	0.05	nil	nil
Na <sub>2</sub> O	0.6	nil	nil	0.27	nil	nil
Cl	0.7	nil	nil	0.31	nil	nil
TiO <sub>2</sub>	0.34	nil	nil	0.34	nil	nil
P	0.03	nil	nil	0.03	nil	nil
Mn	0.03	nil	nil	0.01	nil	nil
S(T)	traces	nil	nil	TRACES	nil	nil
LOI	42.41	41.64	39.37	42.99	37.74	42.69

may be achieved using hydro cyclones or high frequency screens in industrial scale. The + and -500 mesh size fractions of limemud and + and -200 mesh size fractions of limesand were separately assayed and found considerable enrichment of CaO in -500 size fraction of limemud and +200 size fraction of limesand (Table 1). The enriched fractions are found to be suitable for filler industry and the rest can be utilized in cement manufacturing. This suggests that simple wet screening could upgrade the limemud and limesand suitable for both filler and cement industries' applications. The entire material of limemud/limesand can be utilized and generate "ZERO WASTE".

The -500 mesh limemud and +200 mesh limesand concentrates were agglomerated to pellets of different size ranges using molasses. Agglomeration of fine concentrate into pellets makes it as a value added product, by alleviating the material handling problems, so that it can be utilized for industrial application. The highly demanding low silica, high calcium limemud can be utilized as flux after calcination in blast furnace in steel plants, and steel melting shop (second major consumer industry after cement) to lower the temperature of melting and to form calcium silicate by combining with silica of the iron ore, which comes out as slag. The pellets were evaluated for strength by drop shatter test, compressive strength, bulk density, and attrition test. The pellets of all size ranges withstood more than 30 drops from the height of 18 inch on a steel plate. The pellets of all size have compressive strength in the range of 5 to 10.75 kg/pellet for limemud and 5 to 23 kg/pellet for limesand.

The pellets of -3/4+1/2 inch size have maximum strength of 10.75 kg/pellet. The attrition loss of limemud/limesand pellets of different sizes is less than 2 % (Table 2). The agglomerated pellets may find use in steel making as per Bureau of Indian Standards (IS: 10345-1982) for flux limestone<sup>3</sup>.

The studies at IBM mineral processing lab also revealed that limemud as high value powder can be used in coated and uncoated ultrafine fillers, pigments for paper coating, paint, rubber and plastics, pharmaceuticals and food apart from several other uses<sup>3</sup>. Aragonite PCC (Precipitated Calcium Carbonate) is a new kind of functional filler in the paper and plastic industries, nowadays; aragonite PCC synthesis is the most exciting and important industrial application due to numerous attractive properties<sup>8</sup>. It may be noted that limemud mainly consists of ooids which are basically made up of aragonite needles (Fig.1e – f). Hence, a study may be conducted to understand the suitability of limemud in place of aragonite PCC.

## Discussion

### Stone paper vis-à-vis wood pulp paper

In 2021, China produced over 125 million metric tons of paper and paperboard, making it the leading pulp paper producing country worldwide. The United States was the second-largest paper producing country that year, with an output of 67.5 million metric tons<sup>9</sup>. India is the 15<sup>th</sup> largest paper producer in the world. India has emerged as the fastest growing market when it comes to consumption of pulp paper, posting 10.6 % growth in per capita consumption of paper in 2021 – 2027. The domestic market/consumption of pulp paper is over 16 million tons per annum (TPA), with over 2 million TPA being imported. By 2025 – 26, under the baseline scenario, domestic consumption is projected to rise to 23.5 million TPA<sup>10</sup>. However, Pulp and Paper (P&P) industry is one of the most polluting industries, as identified and categorized by Central Pollution Control Board (CPCB), India. The use of wood raw materials is related to deforestation which causes Green House Gasses (GHG) emissions<sup>11</sup>. Plantation forest, from where the majority of wood for pulping is obtained, is generally a monoculture and this raises concerns over

Table 2 — Evaluation of pellet properties analyzed at IBM Lab

Size (inch)		-1+3/4	-3/4+1/2	-1/2+1/4	-1/4+3 mm
Bulk density (kg/m <sup>3</sup> )	Limesand	1135	946	1092	1133
	Limemud	-	930	921	801
Compressive strength (kg/pellet)	Limesand	23 (max.)	13 (max.)	9 (max.)	5 (max.)
	Limemud	7.75 (max.)	10.75 (max.)	6.25 (max.)	5 (max.)
Drop strength	Limesand Limemud	The pellets of all size range withstood more than 30 drops			
Attrition test	Limesand Limemud	The attrition loss of pellets of different sizes viz.; +3/4, +1/2 inch and +6 mm and +3 mm is less than 2 %			

the ecological effects of the practice. Out of the world's 37 billion tonnes of man-made CO<sub>2</sub> release, 5 billion tonnes is released from the manufacture of pulp paper<sup>12, 13</sup>.

It is estimated that one million trees will absorb approximately 24,000 tons of CO<sub>2</sub> each year that is equivalent of taking 6,000 cars off the road each year<sup>14</sup>. The pulp paper industry consumes a significant quantity of water and chemicals and produces large volumes of effluent. Use of significant amount of chemicals, such as sodium hydroxide, sodium carbonates, sodium sulfide, bi-sulfites, elemental chlorine or chlorine dioxide, calcium oxide, hydrochloric acid, etc. in manufacturing of paper results in generation of larger quantities of effluents containing organic and inorganic salts and toxic pollutants, which are let out<sup>11,12</sup>. According to Zhang *et al.*<sup>15</sup>, the 'lime mud' (may not be mistaken with the limemud that's dealt in this paper) from wood pulp papermaking process (LMP) is a solid waste originated from the causticization. Due to fine particle size and high alkalinity of LMP, its recovery is expensive and adverse effect is still uncertain. They reviewed several current techniques proposed for LMP utilization in order to minimize its toxic effect on the environment<sup>15</sup>.

Water is used extensively in the pulp paper business. Around 100 tonnes of water are required to produce one ton of newsprint, and because vast volumes of pure, chemical-free, fresh water are required, fresh water must be available near to the site<sup>16</sup>. According to current Fisher Solve data, 2017, the average water usage is 39 m<sup>3</sup>/ton in India. This is quite high when compared to 16 m<sup>3</sup>/ton for similar paper mills in USA and 8 m<sup>3</sup>/ton for similar mills in EU (European Union) countries<sup>17</sup>. In the process of making paper, trees are the most critical raw materials. It is estimated that 24 trees with a combination of softwoods and hardwoods each about 40 feet tall with a diameter of roughly 6 – 8 inches is required to make 1 ton of standard office paper when the pulp mill uses chemical pulping. If mechanical pulping process is preferred, it uses fewer raw materials (12 trees, a mixture of softwood and hardwood)<sup>18</sup>. In case if the paper is produced from 100 percent virgin wood pulp, 3000 kg wood is required for every 1000 kg of paper, this equals 18 cut trees<sup>19</sup>.

China invented wood-pulp paper production two millennia ago and China re-invented (stone) paper in

the 21st century. In the late of 1990s, a company in Taiwan, Lung Meng Technology Co., began to develop new papermaking technologies called "rock paper" (stone paper). At present, more than 40 countries have patents on this type of paper and their products have been marketed under various trade names<sup>12</sup>. The creation of water repellent paper that requires neither tree fibers nor water is a breakthrough. At this juncture, stone paper is a major alternative to wood-pulp paper. The starting point is the supply of rocks. While calcium carbonate rich stones are the reference, it is possible to use a wide variety of mineral sources provided that the input material meets the standard size of fine dust. The stone dust is mixed with 20 % polyethylene (PE), today a virgin material to be replaced by a recycled PE, and ultimately by a bio-based PE generating more value for the local farmers and creating a higher impact in the region that now can reconnect farming and mining with industrial production<sup>20</sup>.

Stone paper containing 80 % limestone and 20 % recycled polyethylene have been produced by a printing industry in Germany. The mixture of raw materials used are claimed environmentally friendly and can be used for several printed products such as posters, leaflets, up to bag products, and used by farmers for labels and coatings of pot. This paper can be used as a substitute for polypropylene because it is more water resistant, ultra violet (UV) resistant, with high tear strength, even higher than ordinary paper. In addition, stone paper can be even written when it is in wet condition<sup>12</sup>.

The density of stone paper ranges from 1.0 g/cm<sup>3</sup> up to 1.6 g/cm<sup>3</sup>; slightly higher compared to conventional pulp paper. The surface of stone paper is smoother than traditional paper. Stone paper products are compatible with inkjet or solid ink printers (e.g., offset, letterpress, gravure, flexographic) but do not respond well to very high temperature laser printers<sup>21</sup>. Unlike the conventional paper made from fiber which is biodegradable, the stone paper is not biodegradable but it is photodegradable and can be decomposed after being exposed to sunlight for a certain period of time. The time required varies from 12 months to 18 months depending on the type of products. The main competitiveness is (under the same production capacity as wood pulp and paper mills): raw materials that are 50 % lower than wood pulp paper and 50 % lower investment cost than wood pulp paper. Stone paper can be easily recycled back into stone paper or

become Type 2 plastic<sup>12</sup>. However, recycling of sheet metal, the use of biodegradable polymers and UV-resistant additives and arrangements to separate it from recycled paper, will make it possible to replace it with some more paper products<sup>22</sup>.

M/s KIWA which is an authorized testing, inspection and certification company in the Netherlands, tested the stone paper samples produced in Lung Meng Technology (TLM) located in Taiwan supplied and imported by Gaia Concepts B.V. in the Netherlands. The Kiwa Covenant proclaims that stone paper bears good tensile strength, tear resistance and water repellent. Furthermore, Stone Paper is free from wood elements and acids. The cradle to gate carbon footprint, as a partial carbon footprint of a product, is reported as an emission of 474 kg CO<sub>2</sub>e per ton stone paper and for pulp paper as a reference a typical value of 1431 kg CO<sub>2</sub>e per ton is quoted<sup>23,24</sup>.

The production of stone paper does not require any water; consumption is 0 m<sup>3</sup> per 1000 kg stone paper. To produce 1000 kg of pulp paper, European producer need an average of 35 m<sup>3</sup> of water, of this 7.9 % (2.77 m<sup>3</sup>) is consumed; the remainder is reused later on<sup>25</sup>. As per the data given above, 1 million trees approximately absorb 24,000 tons of carbon dioxide<sup>14</sup> means one tree is capable of absorbing 24 kg CO<sub>2</sub> per year. The present consumption of wood pulp paper in India is 16 million tons per year<sup>10</sup>. If an average 15 trees are cut for producing 1 ton paper, the total amount of CO<sub>2</sub> absorption inhibited would be 5.76 million tons.

The size required for granule production which is the initial stage of stone paper making is about 10  $\mu$  (1250 mesh). This is easily attainable since limemud occurs within mean size ranges between 4  $\mu$  and 500  $\mu$ . The physical and chemical properties of limemud suggest it may be a suitable raw material for production of stone paper. The approximate cost of machinery required for stone paper manufacturing unit that includes granule production, cast lining, splitting and cutting for a capacity of 7,000 TPA is about USD 1 million apart from installation and this unit can be operated by 7 – 10 employees<sup>26</sup>. The global stone paper market reached a value of USD 809.3 million in 2021. Looking forward, IMARC Group expects the market to reach USD 1044.4 million by 2027 exhibiting at a compound annual growth rate (CAGR) of 4.29 % during 2022 – 2027<sup>(ref. 27)</sup>.

#### **Mining methodology and transportation**

Conventional dredging technologies, both mechanical (open clamshell bucket, excavators, etc.)

and hydraulic (e.g. suction centrifugal pumps), are still commonly used in several places. These technologies suffer from low operational efficiency, large operating cost and less life time of the main components. These technologies are now dated, and can no longer remove contaminated sediments adequately in ports and harbours throughout the world. Alternative sand removal technologies must be developed and tested in order to remediate those areas. One such alternative tested was the pneumatic lifting of sand during dredging process<sup>28</sup>. The pneumatic dredgers work on the same principle as a vacuum does. They consist of a chamber with high vacuum pressure inside and the chamber is suspended with the help of a crane. Bed material is pumped in the inlet chamber through vacuum pressure and the chamber is lifted up through crane and emptied at the requisite site and the cycle is repeated. The unit is generally suspended from a crane on land or from a pontoon or barge in the sea. These types of dredgers are opted only for easily flowing material<sup>29,30</sup>.

Sustainable mining of limemud from Gujarat offshore may possibly be achieved by pneumatic dredging. The unit can be suspended from a barge and operated more than 100 m water depth. Pneumatic dredgers drawing up to 90 % solids in the dredged mixture with an output up to 2000 m<sup>3</sup>/h are available in the market. These are capable for long distance direct reflow. The approximate cost of a total pneumatic dredging system that includes compressor, pump, barge, A-frame etc. is USD 12 million and the operation and maintenance cost would be about 10 % of the total dredging system cost annually. The system would be lasting for 25 years and can be dredged up to 200 m water depth. If one pump works for 15 hours/day with a lifting capacity of 1800 m<sup>3</sup>/h with 30 % (conservative figure) solids in the dredged mixture, the total mined out material would be 27,000 m<sup>3</sup> per day that's equivalent to about 21,000 tons of limemud<sup>31</sup>. If 220 fair-weather days are considered in an year, the total mined out limemud would be about 4.6 Million Metric Ton (MMT). Hence, the estimated cost of mining limemud would be approx. USD 0.50/ton. All these make pneumatic dredging system cost effective<sup>32</sup>. The sucked in limemud may be pumped into large barges/vessels (capacity  $\geq$  50,000 tons) with water draining facility. The mined material may be transported to nearby beach facilities and pump out from a convenient distance. The cost of transportation of limemud from mining site (80 – 100 km offshore) to beach facilities near Porbandar, Gujarat would be less than USD 5/ton<sup>33</sup>.

The inferred category limemud resource occurs beyond Territorial Waters (TW) within the Western Indian offshore in very large quantity and needs to be explored in detail to bring out the mineable reserves. Though mining restrictions are unwarranted beyond TW, a successful Environment Impact Assessment (EIA) study is mandatory before mining. Dredging possess a huge threat to the marine environment and is required to be carried out quite carefully aided only with the help of the right dredgers and dredges<sup>34</sup>. Multiple potential stressors associated with conventional dredging activities such as mechanical dredges (e.g. grab and excavator) and hydraulic dredges (e.g. trailer suction hopper and pipeline cutter head) are sediment stress (suspended and deposited), release of toxic contaminants, hydraulic entrainment, vibration, noise pollution, reduction of dissolved oxygen, etc.<sup>35,36</sup>. At this juncture, the pneumatic dredging technology may be a right choice. Since pneumatic pumps work on compressed air technology as piston pumps, and no rotating cutters are required, turbidity is almost near to non-existent by maintaining the induced turbidity below 20 NTU (Nephelometric Turbidity Unit) making a marked difference from that of other dredging systems. Further, underwater noise is practically nil, no release of toxic elements and not facilitating hydraulic entrainment and causing reduction of oxygen level. Since limemud is soft and non-sticky that enables the pneumatic pump to suck the material at great ease. Thus pneumatic dredging creates a perfect sustainable mining scenario<sup>31</sup>. Further, the limemud may be spilled over and fall back into the sea while filling barges. However, unlike ordinary mud, it will be settling on the seabed too fast because limemud is not a clay mineral but made of  $\text{CaCO}_3$  and occurs in the form of globular ooids. Advantages of limemud mining and future propositions are listed below.

- 1 The cost of high grade limestone imported from UAE/Oman to ports along the west coast of India would be approx. USD 16 – 18/ton<sup>33</sup>. Whereas the cost of mining and transportation of limemud from Gujarat offshore would be approx. USD 5 – 6/ton that signifies a marked difference.
- 2 The production of stone paper starts with grinding and followed by other processes<sup>26</sup>. Since the limemud occurs in the form of powder (4 – 500  $\mu$ ), grinding is not required, saving large amount of power. This advantage would be applicable to steel and other industries as well.

- 3 Limemud from Gujarat offshore may be able to cater the needs of major and minor steel plants in the west coast states of India that produce about 30 MMT of steel per year<sup>37</sup> and also several filler and chemical industries. The cost of mining and transportation would be competent enough to supply the material even to the east coast of India.
- 4 The difference in emission of  $\text{CO}_2\text{e}$  from the production of wood pulp paper and that of stone paper is about 1000 kg. If 25 % of the wood pulp paper (approx. 4 MMT as per the current production<sup>10</sup>) is replaced by stone paper in a phased manner, it equals 4 million carbon credits<sup>38,39</sup>. This may contribute about 11 % of the India's total carbon credits (35.94 million) available at present<sup>40</sup>. This is significant in the view of our nation's reaffirmed commitment to the UNFCCC (United Nations Framework Convention on Climate Change) towards our long term goal of reaching net-zero by 2070.

### Conclusions

Limemud is found to be suitable for steel, filler and cement industries, stone paper and several other uses. There are several advantages in manufacturing stone paper in comparison with that of wood pulp paper. The striking difference is in the consumption of water and emission of  $\text{CO}_2\text{e}$ . Comparing to conventional dredging methods, pneumatic dredging is found to be environmental friendly. Cost of limemud mining and transportation have been estimated and considered to be much cheaper than that of imported high grade limestone. The decision makers/planners may consider all these factors discussed above to initiate the process and thereby bringing positive transformation in the country.

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### Conflict of Interest

We declare no conflict of interest.

### Author Contributions

ACD: Conceptualization, preparation of manuscript, literature consultation; CJ: Collection of data, literature consultation, presentation of manuscript, cost estimation of limestone import; and PP: Cost estimation for limemud mining and transportation, correspondence and discussion with different agencies in connection with mining technology, transportation.

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