In the context of achieving sustainable development goals, continuously pushing possible extended research and attempting to implement the respective outcomes in expanding a circular economy for a broad range of products are high priorities. In this paper, we considered disposable tannery sludge as a matter of concern and proposed an innovative framework for placing tannery sludge in the economic value chain via an encapsulation technique-based construction product development. We used polypropylene plastic and a cement-water matrix as encapsulation materials, and the encapsulated tannery sludge bodies were tested for their drop strength, water absorbing behaviour, ability to leach chromium and tendency to lose weight upon ignition. Value of water absorption for the prepared double layered encapsulated tannery sludge bodies was 1.332%, the drop strength performance index arrived was 90% and 0.0001 µg/g of Chromium leaching was found in Toxicity Characteristic Leaching Procedure (TCLP) and shown 67% weight loss in thermogravimetric (TGA) analysis. The results confirmed the possibility of ecocompatible disposal and recirculation of tannery sludge for the sustainable production of building blocks in the form of encapsulated bodies. The outcomes of our work add upon a new perspective to the existing literature regarding the environmentally positive utilization of tannery sludge in the production of building blocks.

Keywords: tannery sludge, leachability, encapsulation technique, circularity, sustainability

1. INTRODUCTION

The circular economy (CE) is a multilevel system of resource utilization that mandates the complete closing of all resource loops. The concept of CE is intended to replace the ‘end-of-life’ segment in all business models with the terms reusing, recycling and/or repurposing. [1,2] While advancing toward a ‘net zero waste’ strategy aided by circular economy principles, waste management optimization can

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significantly minimize pollution-heavy activities such as trash incineration and landfills of waste products generated by the end-of-life industry produces. [3] The use of circular tools denotes the existence of a restorative and regenerative system in which the streams of resources and goods are not linear. Major industries have realized the need to restructure their production chains into circular ones since they are more sustainable and take generated trash into account on behalf of parallelly emerging social demands. [4] Taking the industry wastewater treatment units under consideration, achieving either sustainability (environment-economy-social aspects) or circularity primarily involves the rejection of final end products such as sludge and reverse osmosis reject in the case of zero liquid discharge (ZLD) plants. Trying to produce consumable energy from digested sludge or converting it into value-added products are the two ideal solutions that are practiced with industry wastewater sludge as circular measures. [5] On the other hand, the application of circular economy principles in the domain of construction materials research and practices is actively increasing through policy intervention and common global interest initiatives such as the UN-Sustainable Development Goals (UN-SDG) 2030. [6]

1.1 Tannery wastewater treatment and sludge

Tannery wastewater treatment is usually performed at common effluent treatment plants (CETPs) by collecting all the quantitatively varying effluents from member tanneries. The CETP comprises three major treatment concepts: physiochemical coupling with biological treatment systems, reverse osmosis-based tertiary treatment systems and total dissolved solids (TDS) management systems. For the works concerned with this paper, a tannery-based CETP located in Chromepet, Chennai, Tamilnadu, was used. The process layout of the Pallavaram Tanners Industrial Effluent Treatment Company Limited (PTIEC) is shown in Fig 1. [7]

In PTIEC, sludge is produced primarily from the primary clarifier, pretreatment unit and secondary clarifier. The received sludge is initially dewatered and then filtered through a filter press unit. The filtrate is then directed toward the equalization tank, whereas the filtered sludge cakes are collected, transported and deposited at a landfill site located nearer to the CETP. Despite having a chromium recovery unit in the concerned CETP, the technical personnel stated that the filtered sludge prevails because of the significant presence of toxic chromium substances.

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Fig. 1. Pallavaram Tanners CETP process flow diagram [7]
(Source: The PTIETC Website: [http://www.ptietc.org/project_upgration.php](http://www.ptietc.org/project_upgration.php))
1.2 Objectives

The purpose of this paper is to: 1) Conduct a state-of-the-art review on the management of tannery wastewater treatment plant sludge, 2) attempt to develop an encapsulation method as a new disposal technique for chromium containing tannery sludge and 3) discuss the possible extended utilization (circular approach) of encapsulated tannery sludge in the construction sector.

2. STATE-OF-THE-ART REVIEW

To understand the existing practices and research findings on the management of industry sludge, especially those emerging from tannery wastewater treatment plants, a comprehensive review was carried out. The scheme of the executed review study is presented in Fig 2.

To understand the field practices of tannery sludge handling PTIEC, Chromepet was used. It was observed that the treated sludge that emerges from filter press units is regularly collected and transported to a nearby landfill site in trucks. It was noted that the landfilled sludge is then periodically procured by the State Government of Tamilnadu for use as subgrade material in road construction projects. The sludge deposition landfill site is shown in Fig. 3. As shown in Fig. 2, the literature review was carried out under three major headings, sludge treatment techniques, sludge pelletization and recycling of tannery sludge into bricks.

2.1 Sludge treatment techniques

L. Alibardi and R. Cossu demonstrated a novel method for pre-treatment of tannery sludge, which comprises stabilization in the presence of oxygen coupled with compaction and drying. [8] Thus, pretreated tannery sludge was found to benefit the disposal process by reducing the sludge disposal volume and by reducing possible organic and inorganic leaching in landfills, thus restricting long-term landfill emissions. P. Celary and J. S. Szoltysek proposed a vitrification technique against the commonly existing landfilling method for tannery sludge disposal. [9] They presented a flotation followed by a chemical precipitation with different material proportions to achieve better hardened vitrified products with less leaching. The results showed that incinerating tannery sludge before the plasma vitrification process led to better vitrified products.
A P Vig et al., [10] also tested the possibility of converting tannery sludge into value-added products using earthworms. The experiments were intended to co-mix tannery sludge and cow dung in different weight-based proportions and to study the influence of earthworms in converting them into nutrient-rich manure. The results demonstrated the potential use of Eisenia fetida for the effective eco-friendly conversion of toxic tannery sludge into value-added products. In another similar attempt at co-mixing primary tannery sludge (limed) and cattle dung, G Malafaia et al., [11] reported that various proportions of combinations of vermicompost earthworms resulted in the conditioning of objectionable elements in tannery sludge, and the final product could be used in agriculture as soil conditioners.

Researchers from China [12] demonstrated a bioleaching procedure for solubilizing tannery sludge with 99.7% removal efficiency. They treated tannery sludge, which was bioleached during the incubation period, with sulfur-oxidizing A. thiooxidans for six days using a bubble column bioreactor. The results proved that bioleaching can be a less energy intensive process for stabilizing tannery sludge than incineration followed by vitrification. Kavourus et al., [13] presented a chromium speciation study that focused on incinerating tannery sludge, without any specific, pretreatment under the abundant presence of oxygen and absence of oxygen at various temperatures. The results showed that increasing the incineration temperature under oxic conditions, increased the conversion rate of Cr-(III) to Cr-(VI), but under anoxic conditions, the conversion rate decreased by 30% for the same temperature range. During the incineration processes, the observed crystalline phases that formed were calcium chromate and magnesium chromate for oxic and anoxic conditions respectively. A cementitious material-based stabilization/solidification process was attempted by M T Montanes et al., [14] on tannery sludge to assess leachability and chromium retention efficiency. The results revealed that parameters such as the

Fig. 3. Photos taken during the CETP visit - (a) Sludge loaded truck from filter press unit (b) Treated sludge depositing landfill site (c) Collecting sludge for research work (d) Entrance of PTIEC, Chromepet, Chennai
quantity of cement, type of cement and water/cement ratio played significant roles in the final mixture. Additionally, compared to the addition of fly ash and pozzolans, the addition of raw cement-led to a better solidified product when mixed with tannery sludge but was expensive.

In another attempt to vitulate tannery sludge, S Varitis et al., [15] carried out a microstructural investigation on the transformational connection between chromium-rich incinerated tannery sludge ash and nontoxic vitrified products. Six proportions of incinerated tannery sludge ash and glass-forming oxides such as Na₂O, calcium oxide and SiO₂ were prepared for the study. Analytical studies revealed that the conversion of ash-vitrified products into glass-ceramics under controlled temperatures was a sustainable way of managing the tannery sludge disposal, issue as it resulted in further recyclable products. Moreover, J Xu-guang et al., [16] demonstrated that incineration of tannery sludge at 800°C resulted in volatilization of potential heavy metals such as Zn, Cd, Cu, Pb, Mn and Cr. Cr was reported to be volatile only above 900°C, which is highly fuel intensive and not feasible.

S Swarnalatha et al., [17] experimented the starved air combustion at 800°C followed by cement-gypsum based solidification of tannery sludge to destruct the Cr (III) and convert the tannery sludge ash into a construction block with adequate compressive strength. Whereas Y L Wei et al., [18] attempted a complete combustion of Cr rich clay between 500 - 1100°C for 4 hours to remove the objectionable Chromium substance. In both the above attempts, incineration of tannery sludge under 1000°C resulted in major destruction of Cr (III) substance before letting it get converted into Cr (VI). Similarly, in both the attempts, leaching of Cr elements was restricted due to the favorable phase changes with Cr elements.

S M Contreras-Ramos et al., [19] experimented the composting of tannery effluent along with cow manure and wheat straw with the aim of utilizing the available nutrients in the tannery effluent. The compost was prepared in different proportions for 90 days and final compost product was tested for electrical conductivity, cation exchange ability, absorbance and CO₂ respiration rate and germination index. The results shown that the compost product could give reduced germination index due to the large salt concentration and made the compost undesirable. In another similar approach executed by M A Hashem et al., [20] sludge produced from liming unit of a tannery was used along with cow dung, straw dust and chicken manure to prepare a compost product using bamboo aerator. The composting process was held for 60 days. The final compost product was tested and declared suitable for germination with necessary nutrient availability.

Codigestion of tannery sludge was performed with slaughterhouse sludge to assess the improvement in the dynamics of the inhibited anaerobic digestion process. [21] This study aimed to address the issues of abundant nutrient availability and intervention with toxic metals in the ordinary tannery sludge digestion process. The results demonstrated a favourable decrease in solid waste and a significant increase in biogas generation. In another study, [22] the multiplication of the storage capacity of dried tannery sludge powder was assessed through the pelletization method. The study demonstrated that the compaction of tannery sludge powders into dense pellets resulted in a 25% cost savings over the existing practice of storing the tannery sludge. The latter process led to self-heating issues of the deposited sludge mass due to open contact reactions with air and moisture.

In the past investigations regarding the treatment and management of tannery sludge, biological techniques were attempted more as they significantly reduced the objectionable heavy metals. The resultant end products derived from such biological treatment techniques remained to be compost which can find their utilization in ecofriendly farming. On the other hand, significant quantity of research works was carried out on high energy intensive thermal treatment techniques to convert the tannery sludge into compressed ash which was destined to be dumped at landfill. Thermal immobilization of tannery sludge along with clay to produce burnt bricks was also witnessed despite of higher carbon footprint associated. Aerobic and anaerobic condition-based stabilization processes also resulted in promising reduction of chromium – Cr (VI) contents in the tannery sludge, though not economically
viable for large scale processing. None of the past investigation attempted on energy efficient physical processing of tannery sludge to address their environmentally safer disposal.

A summary of the reviewed practices of tannery sludge handling and management is presented in Table 1.

**Table 1. Summary of tannery sludge handling and management techniques**

<table>
<thead>
<tr>
<th>S. No</th>
<th>Authors</th>
<th>Year &amp; Country</th>
<th>Sludge handling technique adopted</th>
<th>Brief outcome</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>L Alibardi and R Cossu [8]</td>
<td>2016 &amp; Italy</td>
<td>Aerobic stabilization followed by compaction and drying</td>
<td>Chemically and physically reduced tannery sludge</td>
</tr>
<tr>
<td>3</td>
<td>A P Vig et al. [10]</td>
<td>2011 &amp; India</td>
<td>Vermicomposting</td>
<td>Compost based agricultural manure</td>
</tr>
<tr>
<td>5</td>
<td>Y S Wang et al. [12]</td>
<td>2007 &amp; China</td>
<td>Bioleaching</td>
<td>Chromium solubilized tannery sludge</td>
</tr>
<tr>
<td>6</td>
<td>P Kavourus et al. [13]</td>
<td>2015 &amp; Greece</td>
<td>Oxic and anoxic incineration</td>
<td>Pre-treated tannery sludge for further solidification / stabilization.</td>
</tr>
<tr>
<td>7</td>
<td>M T Montanes et al. [14]</td>
<td>2014 &amp; Spain</td>
<td>Stabilization / solidification using cement</td>
<td>Environmentally safe tannery sludge for final landfilling</td>
</tr>
<tr>
<td>8</td>
<td>S Varitis et al. [15]</td>
<td>2016 &amp; Greece</td>
<td>Vitrification</td>
<td>Chromium stabilized ash-vitrified monoliths</td>
</tr>
<tr>
<td>9</td>
<td>J Xu-guang et al. [16]</td>
<td>2010 &amp; China</td>
<td>Combustion</td>
<td>Compacted tannery sludge ash</td>
</tr>
<tr>
<td>10</td>
<td>S Swarnalatha et al. [17]</td>
<td>2006 &amp; India</td>
<td>Starved air combustion</td>
<td>Solidified tannery sludge bricks</td>
</tr>
<tr>
<td>11</td>
<td>Y L Wei et al. [18]</td>
<td>2012 &amp; Taiwan</td>
<td>Thermal immobilization with clay</td>
<td>High thermal treated tannery sludge</td>
</tr>
<tr>
<td>12</td>
<td>S M Contreras-Ramos et al. [19]</td>
<td>2004 &amp; Mexico</td>
<td>Composting</td>
<td>Chromium reduced, pathogen free compost</td>
</tr>
<tr>
<td>13</td>
<td>M A Hashem et al. [20]</td>
<td>2021 &amp; Bangladesh</td>
<td>Composting</td>
<td>Stabilized compost product</td>
</tr>
<tr>
<td>14</td>
<td>A B Mpofu et al. [21]</td>
<td>2019 &amp; South Africa</td>
<td>Co-digestion with slaughterhouse sludge</td>
<td>Reduced tannery sludge with biogas production</td>
</tr>
</tbody>
</table>

### 2.2 Sludge pelletization

The blends of secondary sludge from a paper mill and sawdust from a wood processing unit were subjected to pelletization with different sludge proportions. [23] The results showed that the higher the sludge proportion was, the greater the ash produced and the lower the calorific value. The study also revealed that the increased presence of paper mill sludge in the produced pellets led to a higher ash melting temperature, which made the pellets suitable for low-ash melting temperature combustion. In a similar approach carried out by R Nosek et al., [24], paper mill sludge was blended with wood straw in various proportions to produce pellets. The aim of the study was to increase the low ash melting temperature of raw straw pellets with the addition of paper mill sludge as an additive. The results showed
that the addition of 10% (by weight) of paper mill sludge with 90% (by weight) of wood straw pellets led to an increase in the ash melting temperature up to 1260°C above 1020°C compared to that of raw wood pellets.

E. Yilmaz et al., [25] investigated the process of copelletizing sludge obtained from municipal sewage treatment plants and agro-based biomass. The parameters influencing the physical and energy characteristics of the pellets were assessed and reported. The results proved that the resultant copelletized products satisfied the calorific requirement in the combustion process along with coal and exhibited good physical material behaviour, such as absorbability, water resistance and drop strength.

In another study conducted in Ghana, J Nikiema et al., [26] pelletized fecal sludge, which was said to be rich in nutrients and organic elements. The aim of the study was to increase the market reach of the fertilizing potential of fecal sludge in the form of compressed pellets. Five different fertilizer samples were formulated that involved the gamma irradiation process. The moisture content, sludge concentration and binding agent type were the important parameters that were analysed for the different fertilizer compositions. The results showed that the presence of 3% starch as an additive led to the production of better fertilizer pellets.

2.3 Recycling tannery sludge in bricks

In Bangladesh, efforts were made to sustainably produce bricks using tannery sludge. [27] Clay was combined in weight, in amounts of 10%, 20%, 30%, and 40% with tannery sludge. The resulting compressive strength and water absorption largely complied with the ASTM and Bangladeshi standards. Additionally, it has been claimed that the usage of sludge decreases firing energy ranges by 10–40% and up to 15–47%, respectively. The immobilization of heavy metals was demonstrated by TCLP tests on sludge-added bricks, which showed that higher firing temperatures were necessary to achieve this goal. The study demonstrated that bricks created under laboratory circumstances outperformed those made under field settings.

M.A. Abreu and S.M. Toffoli [28] performed an extensive characterization study on chromium containing tannery waste sludge. They interpreted the behaviour of tannery sludge exposed to high temperatures in the range of 1000-1100°C. The results showed that highly thermally treated tannery sludge waste has the potential to act as a ceramic pigment after washing the soluble chromium salts, which favours ceramic tile manufacturing from tannery sludge. In another experimental attempt to immobilize tannery sludge by incorporating it as a raw material for making ceramic product, T Basegio et al., [29] prepared clay-based ceramic blocks with different proportions of tannery sludge aided by a high-temperature firing process and studied the physical properties of those final products. They evaluated the produced blocks against the quality standards of construction bricks as per Brazilian standards. The results obtained resembled the interpretations received in a similar study performed very earlier by M Giugliano and A Paggi [30].

Australian researchers A Ukwatta et al., [31] demonstrated the modified physical and mechanical properties of fired clay bricks supplemented with biosolids (tannery sludge) from effluent treatment plants. The bricks were produced by mixing tannery sludge at various concentrations ranging from 5% to 50% by dry weight. The firing temperature was maintained between 1000 and 1020°C for 180 minutes. The test results satisfied the Australian standards for the compressive strength of building blocks. It was noted that the leachate test showed insignificant metal leaching, and 25% biosolid addition resulted in considerable energy savings regarding the burning temperature requirement. Similar experiments carried out by P Amsayazhi and K.S. Raja Mohan [32] showed that a maximum of 20% tannery sludge added during clay brick production met the requirements of Class I bricks, as per Indian standards.
Terasa et al., [33] demonstrated the effectiveness of partial replacement of clay with several proportional combinations of tannery sludge and bagasse ash for producing burnt clay bricks and evaluated their mechanical behaviour against conventional bricks. The results proved that a cumulative replacement of 20% of both tannery sludge and bagasse ash (with 5% tannery sludge and 15% bagasse ash) with clay could produce favourable bricks according to Indian Standards with respect to compressive strength, water absorption and hardness properties. These results significantly replicated a similar investigation performed earlier [34] by Chinese researchers on the use of industrial wastewater sludge as a raw material for producing burnt bricks. Later, the maximum favourable sludge replacement with clay achieved was 20%, which met the Chinese Standard for minimum qualities of building blocks.

A stabilization technique was evaluated for utilizing tannery liming sludge in the production of bricks as a partial replacement with clay. [35] The liming sludge that emerged from the tanning process was added in different proportions in the range of 2 to 12%, with an eventual increase of 2% in consecutive mixes. The final products, clay bricks burnt at 1000°C, were tested for engineering properties as per ASTM standards and found to be favourable with the addition of 6% lime sludge. The results revealed insignificant heavy metal leaching with the toxicity characteristic leaching procedure (TCLP) test also occurred with 6% lime sludge. In a similar investigation on the utilization of tannery liming sludge performed by Bangladesh researchers, [20] compost products were produced, and their chemical and biological characteristics were tested and reported.

In a Bangladeshi case study, [36] investigated a solid waste management approach for handling leather buffing dust (LBD) that emerges in typical tannery units. LBD was mixed in various proportions ranging from 1% to 12%, in place of clay to produce bricks under high-temperature firing, and the final products were tested for their necessary physico-chemical and environmental properties according to ASTM standards. The results suggested that adding 4% LBD, to the clay brick raw material matrix, helps in producing statutory bricks, as per Bangladesh Standards for Construction Bricks.

The past investigations conducted on the alternative material choices for producing bricks revealed a fact that a greater number of attempts were made with the combination of clay bricks with tannery sludge. This was to achieve the reduction of Chromium-Cr (VI) element present in the tannery sludge in the process of burning the clay bricks under high temperature, 800°C to 1200°C. Even though the burnt bricks demand more energy than unburnt bricks, cementitious bricks were not preferred for recycling tannery sludge. This was due to the possible heavy metal leaching associated with the unburnt bricks, in turn served as a potential research gap.

A summary of the reviewed practices of tannery waste/sludge utilization in the production of alternative bricks is presented in Table 2.
Table 2. Summary of utilization of tannery waste/sludge in brick production

<table>
<thead>
<tr>
<th>S. No</th>
<th>Author(s)</th>
<th>Year &amp; Country</th>
<th>Waste handled</th>
<th>Processing technique(s) adopted</th>
<th>Parameters tested on final product</th>
<th>Achieving circularity with the final product</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>M A I Juel et al. [27]</td>
<td>2017 &amp; Bangladesh</td>
<td>Tannery sludge</td>
<td>Immobilization into bricks</td>
<td>Compressive strength; water absorption; firing shrinkage; weight loss on ignition; bulk density and leaching behaviour</td>
<td>Yes</td>
</tr>
<tr>
<td>2</td>
<td>M A Abreu and S M Toffoli [28]</td>
<td>2009 &amp; Brazil</td>
<td>Tannery sludge</td>
<td>Immobilization into ceramics pigment</td>
<td>Chemical composition; microstructural analysis; organic carbon content; thermal behaviour; leaching behaviour</td>
<td>No</td>
</tr>
<tr>
<td>3</td>
<td>T Basegio et al. [29]</td>
<td>2002 &amp; Brazil</td>
<td>Tannery sludge</td>
<td>Immobilization into clay bricks</td>
<td>Water absorption; porosity; linear shrinkage; transverse rupture strength and leaching test</td>
<td>Yes</td>
</tr>
<tr>
<td>4</td>
<td>M Giugliano and A Paggi [30]</td>
<td>1985 &amp; Italy</td>
<td>Tannery sludge</td>
<td>Immobilization into clay bricks</td>
<td>Water absorption; bending strength; efflorescence; frost resistance and porosity</td>
<td>Yes</td>
</tr>
<tr>
<td>5</td>
<td>A Ukwatta et al. [31]</td>
<td>2016 &amp; Australia</td>
<td>Biosolids from Effluent Treatment Plant (ETP)</td>
<td>Immobilization into clay bricks</td>
<td>Shrinkage; weight loss on ignition; density; density; water absorption; compressive strength; efflorescence and leaching property</td>
<td>Yes</td>
</tr>
<tr>
<td>6</td>
<td>P Amsayazhi and K S R Mohan [32]</td>
<td>2018 &amp; India</td>
<td>Dewatered sludge from tannery effluent treatment plant</td>
<td>Immobilization into cementitious bricks</td>
<td>Compressive strength and water absorption</td>
<td>Yes</td>
</tr>
<tr>
<td>7</td>
<td>Rose E T A et al. [33]</td>
<td>2021 &amp; India</td>
<td>Tannery sludge and sugarcane bagasse ash</td>
<td>Immobilization into clay bricks</td>
<td>Compressive strength; colour; water absorption; soundness and hardness</td>
<td>Yes</td>
</tr>
<tr>
<td>8</td>
<td>C H Weng et al. [34]</td>
<td>2003 &amp; Taiwan</td>
<td>Sludge from industry wastewater treatment plant</td>
<td>Immobilization into burnt bricks</td>
<td>Brick shrinkage; water absorption and compressive strength</td>
<td>Yes</td>
</tr>
<tr>
<td>9</td>
<td>M A Hasan et al. [35]</td>
<td>2022 &amp; Bangladesh</td>
<td>Liming sludge from tanneries</td>
<td>Immobilization into clay bricks</td>
<td>Compressive strength; loss on weight; water absorption and bulk density</td>
<td>Yes</td>
</tr>
<tr>
<td>10</td>
<td>M S Milu et al. [36]</td>
<td>2022 &amp; Bangladesh</td>
<td>Leather buffing dust</td>
<td>Immobilization into bricks</td>
<td>Physiochemical, environmental and morphological properties</td>
<td>Yes</td>
</tr>
</tbody>
</table>
3. CIRCULARITY AND INNOVATION GAP

3.1 Circularity
Material circularity implies that an already exploited product or a waste stream is pushed further by engineering and/or innovative interventions, into a value chain with a new utilization. [1] From this perspective, the dried sludge obtained from tannery wastewater treatment plants is a potential product for use in circular economies. From Tables 1 and 2, it is evident that past investigations have significantly contributed to the circularity of tannery sludge in the context of developing alternative construction materials, yet innovative ways of incorporating sludge in the manufacturing of building materials need to be identified.

3.2 Innovation gap in utilizing tannery sludge in brick production
Innovation in the context of engineering domain implies that it is born when either an existing unexplored theoretical concept or a whole new creative concept results in an economically enhanced product. [37] According to Tables 1 and 2, tannery sludge has not been handled innovatively in the literature, and it has mostly replaced one of the ingredients of brick’s raw material matrix with the aim of producing alternative building blocks. With this approach, on the other hand, the circularity of tannery sludge has also been limited by the closed-ended fate of producing only bricks and not any other construction-related products. The use of an open-ended, efficient form for tannery sludge disposal has remained a potential gap in the literature. Due to which, waste sludge incorporation into brick raw material would follow an unconventional way, lead to limited or elimination of heavy metal leaching from the bricks, over the time, post construction.

Fig.4. Identified gap from the past literature in efficient handling of tannery sludge disposal

In this work, we developed and tested an innovative pathway to address this gap (Figure 4), aiming to achieve economic efficiency and a scalable process for the disposal of tannery sludge in producing building blocks and other potential building elements.
4. MATERIALS AND METHODOLOGY

4.1 Materials
The dewatered sludge from the filter press unit was collected as a grab sample from the sludge dump yard, as per the guidelines provided by United States Environmental Protection Agency for sampling the biosolids [45], from PTIEC, Chromepet, for the purpose of this study. The grab sampling was preferred and done during mid-day operation of filter press units, to minimize the quality variation of the collected sludge. The grab sampled sludge was crushed into powder and was subjected to weathering for seven days to soften [27]. The pulverized sludge was then subjected to sieve analysis to determine its particle size distribution, and an energy dispersive X-ray (EDX) analysis was carried out on the sludge sample to chemically characterize the tannery sludge. Pycnometer bottles were used to measure the specific gravity of tannery sludge and subsequently to determine a value of 1.71 g/mL as the bulk density of the sludge. The pH of the sludge was evaluated using calibrated Hydrogen Ions sensitive electrode as 7.8±0.2. The collected sludge was also tested for the presence of moisture before being subjected to the encapsulation process, which was found to be less than 2%. M sand of construction grade was acquired from the local market and subjected to sieve analysis to determine the particle size distribution. The curves obtained for the particle size distributions of the sludge and M sand are presented in Fig. 5. For preparing the double layer encapsulants, ordinary Portland Cement (OPC) and plastic straw were used. The cement used was 33-grade, while the polyethylene (PE) straws 5mm in diameter were used for first-level plastic encapsulation. The cement used was ensured of not having any lumps and was sieved with a 90-micron sieve before being incorporated into the second-level encapsulation process. The EDX peaks attributed to the raw materials are presented in Fig. 6, and the chemical compositions are presented in Table 3.
Table 3 shows that tannery sludge contained a significant amount of chromium (as much as 5.5% by weight), whereas all the other used raw materials contained carbon and calcium as the chemically dominant constituents. Scanning electron microscopy images showing the surface morphology of the raw materials are presented in Fig. 7. Potable water with a pH range of 6.5-8.5 was used in the process of encapsulation.
PROPOSAL OF AN ENCAPSULATION-LED DISPOSAL METHOD FOR TANNERY SLUDGE – A CIRCULARITY APPROACH

Fig. 6. EDX peaks as received for (a) Tannery sludge (b) M Sand (c) Cement (d) Plastic capsule

Fig. 7. Scanning Electron Microscope (SEM) images for (a) Cement (b) M sand (c) Tannery sludge shot at 500x magnitude with 15kV electron beams
Table 3. Chemical composition of raw materials from EDX analysis

<table>
<thead>
<tr>
<th>Element</th>
<th>Tannery sludge</th>
<th>M Sand</th>
<th>Cement</th>
<th>Empty plastic capsules</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Weight (%)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>28.1</td>
<td>-</td>
<td>2.2</td>
<td>44.7</td>
</tr>
<tr>
<td>O</td>
<td>43.1</td>
<td>45.1</td>
<td>36.6</td>
<td>39.1</td>
</tr>
<tr>
<td>Na</td>
<td>-</td>
<td>3.8</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Mg</td>
<td>1.1</td>
<td>3.0</td>
<td>0.6</td>
<td>0.8</td>
</tr>
<tr>
<td>Al</td>
<td>2.4</td>
<td>8.1</td>
<td>1.9</td>
<td>-</td>
</tr>
<tr>
<td>Si</td>
<td>2.7</td>
<td>24.0</td>
<td>5.9</td>
<td>1.7</td>
</tr>
<tr>
<td>S</td>
<td>2.0</td>
<td>-</td>
<td>1.7</td>
<td>1.4</td>
</tr>
<tr>
<td>Ca</td>
<td>14.2</td>
<td>5.9</td>
<td>49.3</td>
<td>11.2</td>
</tr>
<tr>
<td>Cr</td>
<td>5.5</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Fe</td>
<td>1.0</td>
<td>10.2</td>
<td>3.7</td>
<td>1.1</td>
</tr>
</tbody>
</table>

4.2 Methodology

Based on the interpretations obtained from the literature review, a methodology was developed to fill the gap identified in past investigations regarding more effective disposal of tannery sludge in the context of circularity. The adopted methodology consisted of two components, namely, interpreting the state-of-the-art practices and research findings in tannery sludge management, and processing and testing of tannery sludge encapsulation. The detailed scheme of the adopted methodology is presented in Fig. 8.

Fig. 8. Adapted framework of methodology
The first component of the methodology framework is discussed in Section 2, whereas the second component is broadly presented in Section 4.3 and Section 4.4.

4.3 Tannery sludge encapsulation – A new proposal

The fundamental objective of the tannery sludge encapsulation process was to produce circular and product-based outcomes to fill the research gap through innovative attempts to improve circularity. The detailed steps involved in the tannery sludge encapsulation process are presented in Fig. 9.

Tannery sludge was obtained from the filter press units of PTIEC, Chromepet. The collected sludge was dried in sunlight for two days and further dried for five days at room temperature in the shaded area. The weathered sludge was crushed and powdered manually using a laboratory hammer. The pulverized sludge was then subjected to sieve analysis and EDX to determine its material properties. Waste plastic straws made up of polypropylene were collected from local juice shops. For uniformity, 5 mm diameter straws were selected for this purpose. Then, the plants were soaked in soap water for 24 hours, washed and dried in sunlight before incorporation into the process. The dried straws were subsequently cut into 6 cm long pieces. The pulverized sludge was then stuffed into the plastic straw, and both ends of the straw were sealed using candle light so that the effective filling of sludge inside the straws remained at 5 cm. Thus, the first and inner layers of tannery sludge were encapsulated. To formulate a more effective and sustainable tannery sludge disposal process, manual stuffing of tannery sludge into straws was preferred over pelletizing tannery sludge and putting pellets into straws, to avoid thermal energy consumption during the industrial sludge pelletization process. [39,40]

For the second and outer layers of encapsulation, several materials, such as cement, lime and molten waste plastics were considered. [46] Cement-based coating or encapsulation was found to be practically compatible and effective at preventing objectionable chemical leaching from the encapsulated body. [42] For the execution of a cement-based outer layer of encapsulation, an exclusive mould was needed, and the same design was conceived and developed using the online tool Tinkercad initially. The final design was developed using the Fusion 360 software tool.[47] The design of the mould for the second layer of encapsulation was affected and limited by the shape and dimensions of the first layer of encapsulation. The final design of the mould was 3D printed using the fusion deposition modelling (FDM) technique. The final mould set consisted of a base and a series of five individual mould cells mounted on to the base. The mould was designed such that it could be assembled and disassembled to ensure easy demoulding. The step-by-step process involved in the development of the mould for the outer layer of encapsulation is presented in Fig. 9. The whole process of designing and 3D printing of the mould was supported by VLOG Innovations Ltd., Chennai.

The second layer of encapsulation was initiated by greasing the inner surface of the mould. The sealed plastic-encapsulated tannery sludge was then placed inside the individual mould cells before being assembled onto the base. Cement mixed with potable water at a ratio of 0.6 was poured into the individual mould cells up to the top to seal it. The mould set was kept undisturbed for 12 hours before being demoulded. The demoulded cement-encapsulated tannery sludge bodies were cured using water for 14 days. After 14 days of curing, the cement-encapsulated tannery sludge bodies were subjected to laboratory testing procedures to assess their suitability for further possible utilization as a measure of circularity.
4.4 Testing of encapsulated tannery sludge bodies

4.4.1 Drop strength test
Circularity demands handling and transporting intended products from place to place in the supply chain. A drop strength test was used to evaluate the resistance of the encapsulated tannery sludge bodies to failure while dropping on a concrete surface from a height of 1 m. [44] The drop strength index was calculated as the number of samples broken relative to the total number of samples dropped during the test. A total of 30 samples were tested, during which the encapsulated bodies were kept upright and dropped from height of 1 m onto the concrete surface. Out of the 30 samples tested, one sample was mildly damaged, and two samples were strongly affected (Figure 10). The drop strength index was calculated as given in Equation (1):

\[
\text{Drop strength index} = \frac{\text{Number of unaffected samples}}{\text{Number of total samples}} \times 100
\] (4.4.1)

The drop strength index reached 90%, which indicated that the final cement-encapsulated tannery sludge bodies were suitable for mass handling and dumping.

4.4.2 Water absorption test
As encapsulated tannery sludge bodies are intended to circulate as new products in various construction-related activities when they are used in pavement subgrades and as fillers in flooring, and weather resistant layers in roofing, water absorption has become an important material property. The Indian Standard Code (IS 3495 Part 2:1992) [45] was used to determine the water absorption of cement-encapsulated tannery sludge bodies, which is usually used for determining the water absorption of burnt clay bricks. To estimate the water absorption capacity, cement-encapsulated tannery sludge bodies were initially dry weighted (W1) and subsequently submerged in cold water within a temperature range of 27 ± 2°C for one day. After 24 hours of immersion, the samples were removed from water and completely wiped to dryness before weighing to obtain the final weight (W2). The water absorption was calculated using Equation (2):

\[
\text{Water absorption} = \frac{W_2 - W_1}{W_1} \times 100
\] (4.4.2)

where W1 is the dry weight of the cement-encapsulated tannery sludge body and W2 is the final weight of the cement-encapsulated tannery sludge body after 24 hours of cold-water immersion. The water absorption value reached 1.3326%, with the standard deviation of 0.0303, which is insignificant for the intended use of cement-encapsulated tannery sludge bodies in various construction activities.
Fig. 9. Tannery sludge encapsulation process (black arrow indicates primary process steps and red arrow indicates sub-constituting process steps for the primary process steps)
4.4.3 Leachability test

The leaching characteristics of the encapsulated tannery sludge bodies are a significant component of their material behaviour in the assessment of their environmental compatibility. As prescribed in U.S. Environmental Protection Agency guidelines, toxicity characteristic leaching procedure (TCLP) analysis [46] was carried out on two levels of encapsulants: (a) on only plastic encapsulated tannery sludge bodies and (b) on both plastic + cement encapsulated tannery sludge bodies to understand the impact of material selection in the encapsulation process of environmentally cautious heavy metals. The solid-to-liquid ratio was maintained at 1:20 [48] in the TCLP rotating shaker/vessel (Fig 11 - a1&a2). The laboratory conditions of the TCLP test used to assess the leachability are listed in Table 4.

Table 4. Laboratory conditions adapted for running TCLP test

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample-Medium ratio</td>
<td>1:20</td>
</tr>
<tr>
<td>Medium for extraction (for 1 litre)</td>
<td>5.7 mL glacial acetic acid and 64.3 mL 1N NaOH</td>
</tr>
<tr>
<td>Running time of extraction vessel</td>
<td>18 hours</td>
</tr>
<tr>
<td>Rotating speed of the extraction vessel</td>
<td>30 revolutions per minute</td>
</tr>
<tr>
<td>pH and Temperature maintained</td>
<td>4.95 ± 0.06 and 21 ± 3</td>
</tr>
</tbody>
</table>
After 18 hours of running the extraction vessel, the liquid from the containers was injected, through a 0.7 µm Whatman glass fibre filter paper with the help of a filter holder, and the filtrate was collected in separate plastic containers (Fig 11 – b). The filtrate was then dried and analysed via energy dispersive X-ray spectroscopy (EDX) to determine the presence of objectionable chromium in the filtrate. Table 5 shows that the filtrate extracted from the plastic + cement encapsulated bodies satisfied the World Health Organization (WHO) standards, whereas the filtrate obtained from the plastic encapsulated bodies contained a significant amount of leached chromium.

4.4.4 Weight loss on ignition
Thermogravimetric analysis (TGA) of the encapsulated bodies was performed for single-layer encapsulation (plastic straw) and double-layer encapsulation (plastic straw + cement-water matrix) to determine the change in weight of the encapsulated bodies with respect to the temperature increase. Both single and double encapsulation resulted in significant amount of weight loss of 65% for double encapsulation and 67% for single encapsulation with standard deviation of 0.8535 and 0.9111 respectively, of the initial weight, during ignition under the supply of nitrogen gas. The temperature was increased at a rate of 10°C/min starting from the initial temperature of 30°C until it reached a maximum of 800°C.
5. DISCUSSION AND SUMMARY

The objective of this work was to develop an innovative process via encapsulation to make treated tannery sludge, available for circularity. Accordingly, double-layer encapsulation with plastic straw and a cement-water matrix was designed and executed.

5.1 Struggles encountered

The materials employed for attempting encapsulation were tannery sludge, M-sand, cement, lime, molten waste plastic and water. In the encapsulation process, lime and molten plastic were initially considered for the second layer encapsulation. However, lime-encapsulated tannery sludge bodies were completely dissolved during the water absorption test. Molten waste plastic also faces trouble when poured into a specially designed mould for second layer encapsulation. A high level of difficulty regarding an appropriate container to hold the molten waste plastic to remain in the molten state was also encountered, which cumulatively led to the withdrawal of lime and molten waste plastic from the consideration.

For the execution of the first layer of encapsulation, the pelletization of tannery sludge before being added to the plastic straw was initially considered and subsequently withdrawn. This was due to the thermal energy requirement in the process of sludge pelletization using an industrial pelletizer. It was observed that either pelletization or copelletization of sludge is preferred primarily for increasing the calorific value, which would benefit incineration-led energy generation methods. [25,39] This led to the manual stuffing of processed tannery sludge into the plastic straws and sealing of the ends using candle light, to derive the first layer of encapsulation. This preserved potential energy consumption for executing our proposed encapsulation method. Moreover, the purpose of our work fundamentally lies in the circular utilization of the proposed encapsulated sludge in the construction material domain only and not in combination with energy production.

![Fig. 12. After submerging in cold water for 24 hours (a) cement encapsulation (b) Washed out lime encapsulation](image-url)
5.2 Testing results for encapsulated tannery sludge bodies

Double-layer encapsulation with plastic straws initially and with a cement-water matrix ultimately delivers more compact and modular products, pushing tannery sludge into the circular supply chain for further utilization but in a completely different form.

The strength of the encapsulated tannery sludge bodies was tested preliminarily by a drop test to demonstrate the ability of these bodies to withstand onsite handling of bulk masses of such encapsulated bodies in the context of dumping and transportation activities. As the second layer of encapsulation was made of a cement-water matrix, the strength attainment of the encapsulated bodies depended on the rate of hydration of the cement in the matrix. The water-cement (W/C) ratio was maintained at 0.6, after several initial trials with 0.45 and 0.5 as the W/C ratio. The attempted trial W/C ratios were not selected due to the lack of workability of the cement-water matrix with the designed moulds. A W/C ratio of 0.6 was helpful for successful casting of the second layer encapsulated with the designed mould set. However, the damage occurred on the encapsulated bodies was due to the lack of bonding between 1st layer and 2nd layer of encapsulations. As the surface of the 1st layer – plastic encapsulation remains shiny as it is brought in to the process, failed to hold the cement matrix applied on to it. This resulted in separation of 2nd layer – cement-water matrix encapsulation out of contact. This can be overcome by increasing bonding between encapsulants through roughening the outer surface of 1st layer before applying the 2nd layer. A drop test was performed on the encapsulated bodies after 14 days of water curing. There is a scope to further investigate the impact of the curing period, and method of curing on the drop test performance. Additionally, there is a potential scope for developing a third and more rigid layer of encapsulation with a suitable material(s) around the cement encapsulation zone to make stronger encapsulated bodies for certain application, and in that case, a usual material compression test can be performed to measure the resistance of the material to loading.

Table 5. Leachability values of encapsulated bodies – TCLP

<table>
<thead>
<tr>
<th>Toxic metal</th>
<th>Permissible concentration of Chromium in soil in µg/g (WHO Standard)</th>
<th>Concentration (µg/g) of Chromium after exposing the ‘cement’ encapsulation under leaching condition for 18 hours</th>
<th>Concentration (µg/g) of Chromium after exposing the ‘plastic’ encapsulation under leaching condition for 18 hours</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chromium</td>
<td>100</td>
<td>0</td>
<td>100</td>
</tr>
</tbody>
</table>
A water absorption test was carried out to determine the moisture absorbance of the encapsulated materials. The water absorbance was 1.33% (<2%), which indicated the unavailability of pore spaces on the external, and second layers of the cement-water matrix. This was also supported by the SEM images (Figure 14) captured on the surface of the outer cement encapsulation layer. As lime was initially considered for the second layer encapsulation, the lime encapsulated tannery sludge bodies were subjected to a water absorption test. Because lime took longer than cement to set, the lime encapsulation agent was removed (Fig. 12) during cold water submergence.

As environmentally objectionable chromium-containing sludge is handled for encapsulation, heavy metal leaching analysis is more important than any other material property. The TCLP test was carried out as per the US EPA guidelines to assess the extent of chromium leaching from the encapsulated tannery sludge bodies. Table 5, shows that the leaching of chromium from a single layer of encapsulated plastic straw resulted in significant chromium leaching, as indicated by the World Health Organization (WHO) guidelines, (100 µg/g). This was due to improper sealing of the ends of the plastic straws. Because, the bodies of plastic straw after encapsulation remained intact, the end zones
became susceptible to leakage of heavy metals. This was supported by the SEM images (Figure 13) obtained from the cross-sectional view of the plastic encapsulation at 100x and 500x magnifications.

On the other hand, the TCLP test results showed that the amount of leachate from double layer encapsulation was negligible. This was because the sealed zones of the plastic straws were completely covered by the cement-water matrix and eventually became rigid enclosures; thus, the resulting leachate was completely arrested, even though the sealed zones were susceptible to weakness. This was supported by a set of SEM images (Figure 14) captured at the cross section of the double-layer encapsulated tannery sludge bodies.
Thermogravimetric analysis was preferred for tannery sludge encapsulated bodies because they are intended to be incorporated in the development of building blocks, and because it is possible to include them in fired brick production. A linear relationship (Figure 15) was found between weight loss during the ignition period and the increase in temperature, for both the single- and double-layer encapsulants. At a maximum temperature of 800°C, single layer (plastic straw) and double layer (plastic straw + cement-water matrix) encapsulated bodies lost approximately 67% and 65%, respectively, of their mass, which implies that approximately 33%-35% of their mass remains intact during the ignition period. This can lead to further investigations of plastic-clay composites that develop during the burning process, while incorporating encapsulated tannery sludge bodies in clay bricks [44] as a measure of circularity.
5.3 Extending disposable tannery sludge into a circular economy

The definition of a circular economy [1] implies pushing a product while at its disposal age toward further utilization to keep the product either in the same or modified form back into the economy again. The basic purpose of the proposed encapsulation technique was to derive a circular pathway for the continuous utilization of disposable chromium-containing tannery sludge in the form of modular products in such a way that it can be used in combination with existing construction materials in the context of various potential construction activities. An innovation gap is emerging [44] in the utilization of industrial sludge as a raw material for producing sustainable building blocks such as low cement fly ash bricks, eco-labelled cementitious bricks. The results of our proposed encapsulation technique can address the lack of innovation in the production of sustainable bricks made with industrial wastewater sludge. Fig 16 demonstrates one of the potential utilization frameworks for the proposed encapsulated tannery sludge bodies in the context of producing eco-compatible bricks, in the form of embedded tannery sludge in clay and fly ash bricks. By adopting such frameworks, disposable tannery sludge, which is found in significant quantities across various geographical regions of a country, can be placed in the economic chain again as a circularity measure.

![Fig. 16. Utilization framework for the proposed encapsulated tannery sludge bodies](image-url)
The amount of tannery sludge that could be recirculated successfully with one encapsulated cylindrical body 5 cm in length and 5 mm in diameter, was estimated to be 5 g. With the help of favourable results obtained in the present study (Table 5), it is possible to develop additional mould configurations to recirculate additional tannery sludge. Attempting such different shape configurations with more than two layers of encapsulation would lead to eco-compatible utilization of objectionable tannery sludge in structural and non-structural components in encapsulated form. Figure 17 demonstrates one of the possible pathways or producing alternative construction aggregates by developing a compatible third layer of encapsulation around our proposed two-layer encapsulated tannery sludge bodies.

6. CONCLUSION

In this work, we proposed an innovative framework for sludge encapsulation process that can be applied to disposable tannery sludge. The behaviour of the prepared encapsulated tannery sludge bodies was tested for water absorption, drop strength, chromium leachability and weight loss on ignition with the aim of developing a novel construction product. The results obtained for the developed encapsulated bodies were 1.332% for water absorption test, 90% as drop strength performance index, 0.0001 µg/g of Chromium leaching in TCLP test and 67% weight loss during TGA analysis, which support the possible application of the proposed framework to redirect tannery sludge into an economical value chain as a circularity measure. However, susceptible breaking of 1st layer of encapsulant in the acidic environment, failure of 2nd layer encapsulant during high impact loading are the projected limitations of encapsulated tannery sludge bodies. As a scope of future work, we intend to investigate the incorporation of proposed encapsulated tannery sludge bodies in fly ash and clay bricks, and to report the benefits, challenges, and final product performance as a separate research publication in the near future.
**Declaration of interests**

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.

**REFERENCE**


