

1 **Potential pathway for recycling of the paper mill sludge compost for brick** 2 **making**

3 Gaurav Goel^{a,b*}, Milica Vidak Vasić^c, Nirmal Kumar Katiyar^a, Kirthika S. K.^a, Milada Pezo^d, P.
4 Dinakar^e

5 ^a *School of Engineering, London South Bank University, 103 Borough Road, London, SE10 AA, UK*

6 ^b *School of Aerospace, Transport and Manufacturing, Cranfield University, Bedford, MK43 0AL, UK*

7 ^c *Institute for Testing of Materials IMS, Bulevar vojvode Mišića 43, 11000 Belgrade, Serbia*

8 ^d *Department of Thermal Engineering and Energy, "VINČA" Institute of Nuclear Sciences - National*
9 *Institute of the Republic of Serbia, University of Belgrade, 11001 Belgrade, Serbia*

10 ^e *Department of Civil Engineering, Indian Institute of Technology Bhubaneswar (IITBBS), Khordha,*
11 *Odisha, India - 752050*

12

13

14 *Corresponding author Email: goelg@lsbu.ac.uk

15

16 **Abstract**

17

18 This study's focus was to develop a potential pathway for recycling of the paper mill sludge
19 compost (PMSC) in brick making. Composting reduces the paper mill sludge (PMS) moisture
20 content considerably and shredding becomes easier. The addition of PMSC leads to an increase
21 of porosities in bricks and makes them lighter, besides delivering energy to the firing process
22 from burning organics. Lighter construction materials help minimize construction outlay by
23 reducing labour and transportation costs and lesser expense on foundation construction. The
24 variability in the experimental data and the brick properties were investigated for two types of
25 soils, typical in the brick industry of India (alluvial and laterite soil), blended with PMSC in
26 five mix ratios (0%, 5%, 10%, 15% and 20%). The samples of oven-dried bricks were fired at
27 two different temperatures (850 and 900 °C) in an electrically operated muffle furnace
28 representing typical conditions of a brick kiln. Various properties of bricks were analyzed
29 which included linear shrinkage, bulk density, water absorption and compressive strength.
30 Conclusions were drawn based on these properties. It was found that the addition of PMSC to

31 the alluvial and laterite soil by up to 10% weight yield mechanical properties of fired bricks
32 compliant with the relevant Indian and ASTM codes. Toxicity characteristic leaching
33 procedure (TCLP) tests showed that PMSC incorporated fired bricks are safe to use in regular
34 applications as non-load-bearing and infill walls. This study is timely in light of the European
35 Green Deal putting focus on circular economy. Besides, it fulfills the objective of UN
36 sustainable development goals (SDG).

37

38 **Keywords: Paper mill sludge compost; Fired bricks; Recycling; Sustainability; Waste-**
39 **to-brick; Laterite soil; Alluvial soil**

40

41 **List of abbreviations:**

42 AAS: Atomic absorption spectrometer

43 DSC: Differential scanning calorimetry

44 ETE: Estimated total emissions

45 HHV: Higher heating value

46 LOI: Loss on ignition

47 PMS: Paper mill sludge

48 PMSC: Paper mill sludge compost

49 SEM – Scanning electron microscope

50 TCLP: Toxicity characteristic leaching procedure

51 TGA: Thermogravimetric analysis

52 XRD: X-ray diffraction

53 XRF: X-ray fluorescence

54

55 **1. Introduction**

56

57 Growing urbanization and sky rise construction of modern buildings have led to the increasing
58 demand for already scarce construction raw materials. Clay obtained from opened quarries is
59 an ideal example that has long been used for brick making. On the other hand, urbanization has
60 also led to the problem of waste management both from domestic and industrial sites.
61 Consequently, there is an arduous need for a collective and joint campaign to develop solutions
62 for waste management as well as to save the precious raw materials which once undergoing an
63 irreversible thermodynamic process will take far too long to recycle. As an example, the
64 construction industry is one area where waste materials are easy to utilize without creating any
65 adverse environmental effect [1, 2].

66 Worldwide, paper manufacturing is responsible for a significant amount of wastes [3, 4].
67 This is inevitable because the paper is at the heart of everyone's life whether it includes flicking
68 through a newspaper to begin the day, notebooks required to teach kids in school or at
69 Universities, documentation in offices or courts, and in more recent time also as serving plates
70 and as packing bags as the paper is far more recyclable over plastic bags. Therefore, researchers
71 have been looking into innovative ways for managing paper mill sludge (PMS). PMS is
72 recycled as a source of energy through incineration and anaerobic digestion [5-8]. Ash is
73 produced in incineration and anaerobic digestion produces digested slurry. One way of using
74 this PMS ash is in the making of concrete slurry, where part of cement is substituted by ash
75 additive [9] to achieve better bonding strength of the slurry mix.

76 Other researchers suggest producing wood-paper sludge boards by substituting a certain
77 amount of wood chips with PMS [10, 11]. However, substitution makes the board weaker by
78 reducing strength [11]. PMS can also be utilized in fired brick manufacturing [2, 12-14].
79 However, inventory control of PMS poses significant challenges. To overcome this problem

80 one way is to convert PMS waste into compost which can be easily stored without requiring as
81 much volume of storage required by raw PMS [15, 16].

82 Unlike other high-value composts, the agricultural use of PMSC is problematic. Numerous
83 soil contaminants may be applied in the agricultural fields. German Environmental Protection
84 Agency does not allow agricultural use as the ecological risk potential of deinking sludge is
85 not adequately known. A recent study [15] has shown that bioavailable heavy metals may be
86 present in PMSC. Leaching of heavy metals poses a health hazard and therefore, the use of
87 PMSC in agriculture cannot be recommended. Compost has been used as a substitute for soil
88 cap on landfills [17, 18], rehabilitation of mine sites and tailings [19], and as fuel briquettes
89 [20]. An alternative substitute being proposed is to use the PMSC in brick making.

90 China, India, Brazil, Spain and Italy are the biggest consumers of fired bricks in that order [21].
91 Out of all the fired bricks made worldwide, 80% of bricks are consumed by China and India
92 alone. As per one estimate, a brick kiln caters to the requirement of 10,000 people in India [22].
93 Brick making comes at the expense of consuming precious fertile soil and a considerable
94 incentive is hidden in saving this soil by developing partial substitutes to it such as dredged
95 sediments [23]. To reduce the consumption of soil, researchers have suggested blending of
96 various solid waste materials in the making of bricks. Glass waste [24, 25], agricultural waste
97 [26, 27], biosolids [28], waste marble sludge [29], degraded municipal solid waste [1], PMS
98 [2] are all been used to create waste incorporated bricks. Since PMS contains a large amount
99 of moisture along with fibrous content, it is not so relevant for direct industrial use. As argued
100 above, composting reduces the PMS moisture content considerably and shredding becomes
101 easier besides providing inventory control. Besides, the fibers strengthen the green products,
102 which is important in transportation and bearing the load in a kiln when firing begins. Hence,
103 using PMSC over PMS became the key motivation for this work and is being explored in this
104 paper as an additive for brick making. The novelty of the work is to make use of PMSC in brick

105 making for the first time. This body of work is the first to report on utilizing PMSC in brick
106 fabrication and to establish the structure-property relationships.

107

108 **2. Materials and methods**

109

110 2.1. Materials

111

112 This laboratory-scale work made use of paper mill sludge (PMS), cow dung and sawdust
113 (mixed in 5:1:1 ratio). The mixture was composted using a rotary drum composter (Capacity
114 500 L), to accelerate the process of composting and the process was accomplished within 15
115 days [30]. PMS was collected from Nagaon Paper Mill, state of Assam, India. Two types of
116 soil (i) laterite soil; (ii) alluvial soil were gathered from the Guwahati region, state of Assam,
117 India. The motive to select these particular two soil types stems from the Indian standards [31,
118 32] and other research works [1, 2, 33] so that the results obtained can be linked retrospectively.
119 These soils are widely used in India for centuries in the brick-making and they were obtained
120 from Guwahati region as they are easily available and abundant. The soil samples were
121 thoroughly oven-dried (median size of 150 μm) before the brick making process.

122

123 2.2. Methods for characterization of soils and PMSC

124

125 As one of the aims of this paper is to establish the structure-property relationship, it became
126 a prerequisite to characterize all the raw materials used so those insights can be gained into
127 linking the results with the properties of the raw materials utilized. Accordingly, soils and
128 PMSC were analyzed to understand their mineralogical, chemical, thermal and plastic index
129 properties using several state-of-the-art instruments. Elemental analyses of total nitrogen,

130 carbon, hydrogen and sulfur were executed to get some idea of the anatomy of the organic
131 matter (Euro Vector, EuroEA3000).

132 The phase composition of raw materials was identified using X-ray diffraction and TGA.
133 The XRD apparatus used was a Rigaku, TTRAX III power X-ray Diffractometer equipped with
134 a monochromator using $\text{CuK}\alpha$ radiation ($\lambda=1.5406 \text{ \AA}$). The scan rate was kept $4^\circ/\text{min}$. The
135 analysis was done using the ICDD database. TGA was carried out on crushed samples heated
136 to 1000°C at a constant rate of $10^\circ\text{C}/\text{min}$ in a static nitrogen atmosphere. The calorimeter was
137 used for measuring the heating values (Parr 1341, 6775 digital thermometer). AAS was used
138 for heavy metals determination (Thermo scientific, iCE 3000). A standard protocol was
139 followed for sample digestion [34]. Atterberg limits and specific gravity tests were conducted
140 as prescribed by BIS: 2720-1985 (revised in 2015). The surface of the samples was analyzed
141 using a scanning electron microscope (JEOL JSM-6610 LV) Before analysis, the samples were
142 covered with a layer of gold for better reflection, using a coating spray device. Finally, XRF
143 (PANalytical, AXIOS) was used to examine the chemistry of the dried material.

144

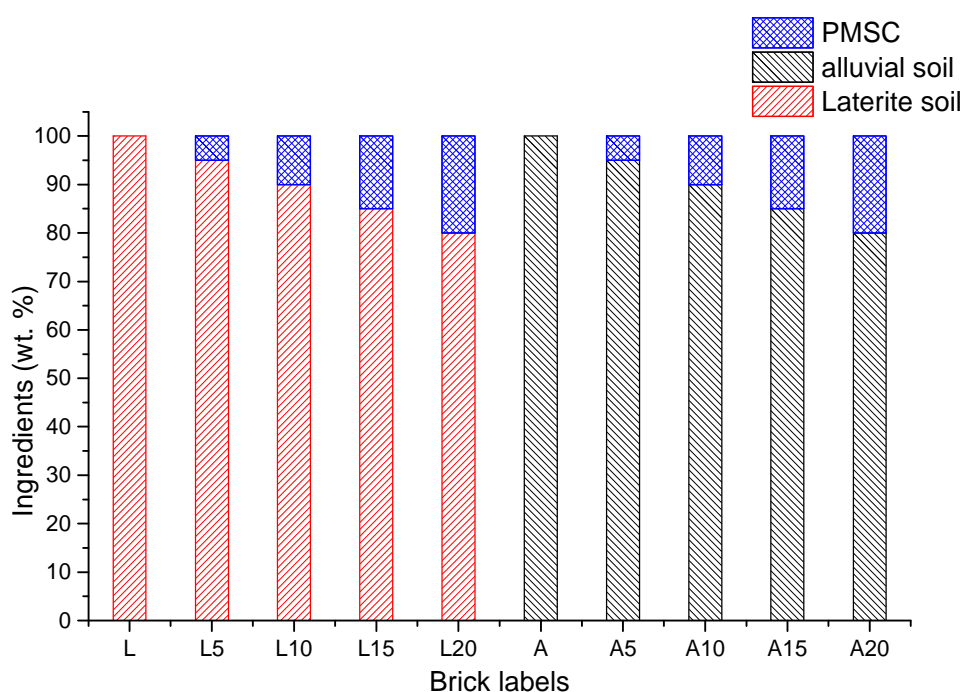
145 2.3. Procedure for brick making and characterization

146

147 PMSC was combined independently in both types of soils in different magnitudes as shown
148 in Fig. 1. Homogenization of the mixture was achieved using mechanical stirring. The
149 minimum quantity of water (20-22%) based on the clay workability chart was added to achieve
150 the state of plasticity for the mixes. To avoid cracking during drying, moulded brick samples
151 were (i) air-dried under ambient conditions for 24 h and, (ii) subsequently oven-dried at 105 ± 5
152 $^\circ\text{C}$ further 24 h. Being a testbed study, the brick specimens made were of dimension $61 \text{ mm} \times$
153 $29 \text{ mm} \times 19 \text{ mm}$ using hand moulding. This size was downscaled from the existing dimension
154 of $230 \text{ mm} \times 110 \text{ mm} \times 70 \text{ mm}$ [35]. Henceforth, the oven-dried bricks are referred to as unfired

155 bricks. Reference brick samples were made purely with the two types of soil samples without
 156 any PMSC for benchmarking the results.

157 The bricks were distinguished by marking them as L (laterite soil) and A (alluvial soil).
 158 Statistical variation of the results was studied by preparing and testing six samples for each
 159 series. The compacted samples were sintered at different firing temperatures (i.e. 850 and 900
 160 °C) mimicking typical kiln conditions. The rate of heating was 2 °C/min and bricks were hold
 161 for an hour at peak firing temperature.



162

163

164 Fig. 1. Compositions of brick specimens.

165

166 To understand the carbon footprint of these fired bricks, gaseous emissions were measured
 167 using the strategy mentioned in our earlier work [33]. The chemo-mechanical identification
 168 procedure enacted in this exercise is similar to the preceding works and is not repeated here for
 169 brevity [1, 2, 33]. Linear shrinkage, loss on ignition, water absorption (5 h boiling test),
 170 apparent porosity, and bulk density were deduced following relevant ASTM standards (ASTM
 171 C326, ASTM C373, and ASTM C67). The compressive strength and modulus of elasticity of

172 fired samples were measured using a universal testing machine (UTM, BISS, Median-250) as
173 described by different standards [36-38].

174

175 **3. Results and discussion**

176 The systematic measurements of the important parameters during the process of brick making
177 and the quality of fired products, following the procedure in agreement with various
178 international standards, are presented in this section.

179

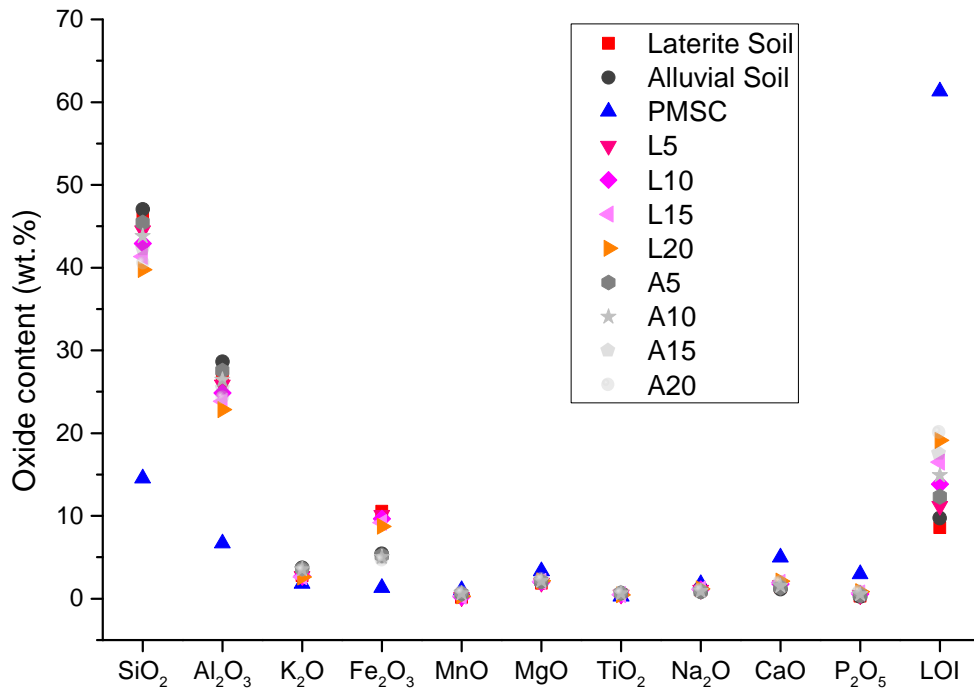
180 **3.1. Determination of chemical composition**

181

182 Chemical composition results (Fig. 2) evince (i) laterite soil and, (ii) alluvial soil are
183 composed of primarily silica and alumina with some amount of flux agents (K_2O , Fe_2O_3 , CaO ,
184 MgO and TiO_2). Laterite soil exhibited a significant quantity of Fe_2O_3 (10.49%) which is also
185 reported by another independent study [39]. The loss on ignition (LOI) values for both types
186 of soils were very low ($< 10\%$). It was established that both soils are kaolinitic, non-calcareous,
187 and low refractory [33].

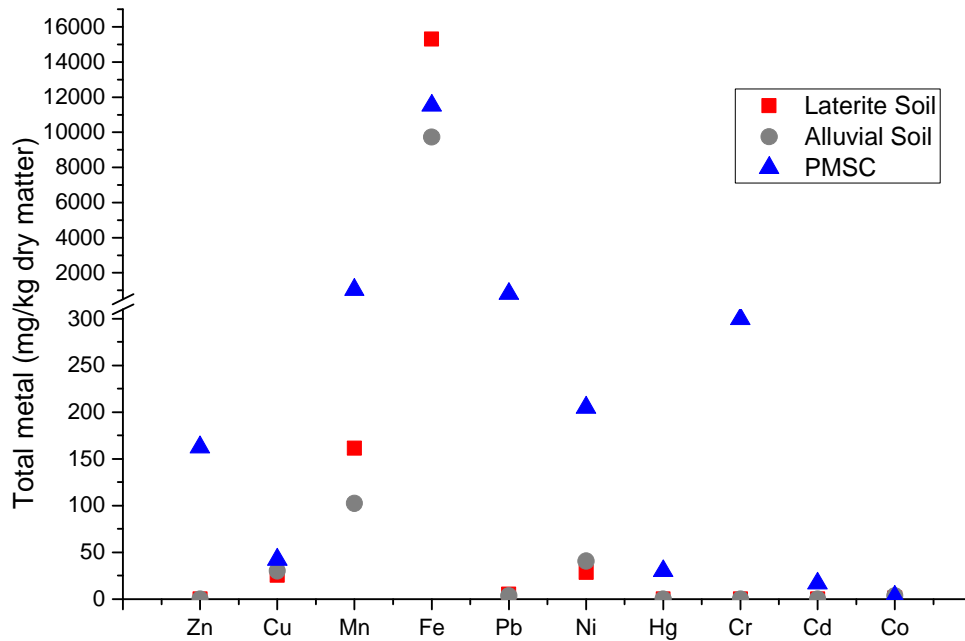
188 PMSC is primarily composed of carbon as disclosed by elemental analysis (hydrogen
189 (2.48%), carbon (25.16%), and nitrogen (3.11%)). PMSC samples were found free from sulfur.
190 The calorific value of PMSC was 2232 kcal/kg. Similar to soils used in this work, PMSC
191 contained mainly silica and alumina. Heavy metal contents for both soils and PMSC are given
192 in Fig. 3.

193



194

195 Fig. 2. Chemical analysis of oven-dried soils, PMSC, and brick compositions.



196

197

198 Fig. 3. Trace metal concentration in both soils and PMSC after oven drying.

199

200 Phase composition results also established that both soils were rich in silica (SiO₂). PMSC

201 primarily consisted of microcline (KAlSi₃O₈). Other minor constituents of PMSC were

202 anorthite ($\text{CaAl}_2\text{Si}_2\text{O}_8$), aluminum orthophosphate (AlPO_4), magnesioferrite (MgFe_2O_4),
203 kutnohorite ($\text{CaMn}(\text{CO}_3)_2$), quartz (SiO_2) and dolomite ($\text{MgCO}_3 \cdot \text{CaCO}_3$). This depicts changes
204 as PMS contained primarily dolomite, carnegieite, microcline, and periclase [2]. Except
205 microcline other phases were transformed.

206 Thermal analysis of PMSC was done and rapid mass loss (~50%) up to 400 °C, possibly due
207 to the volatilization of hemicellulose and cellulose was observed. Beyond 400 °C, mass loss
208 was minor (~10%). The loss on ignition (LOI) values for PMSC was 40%.

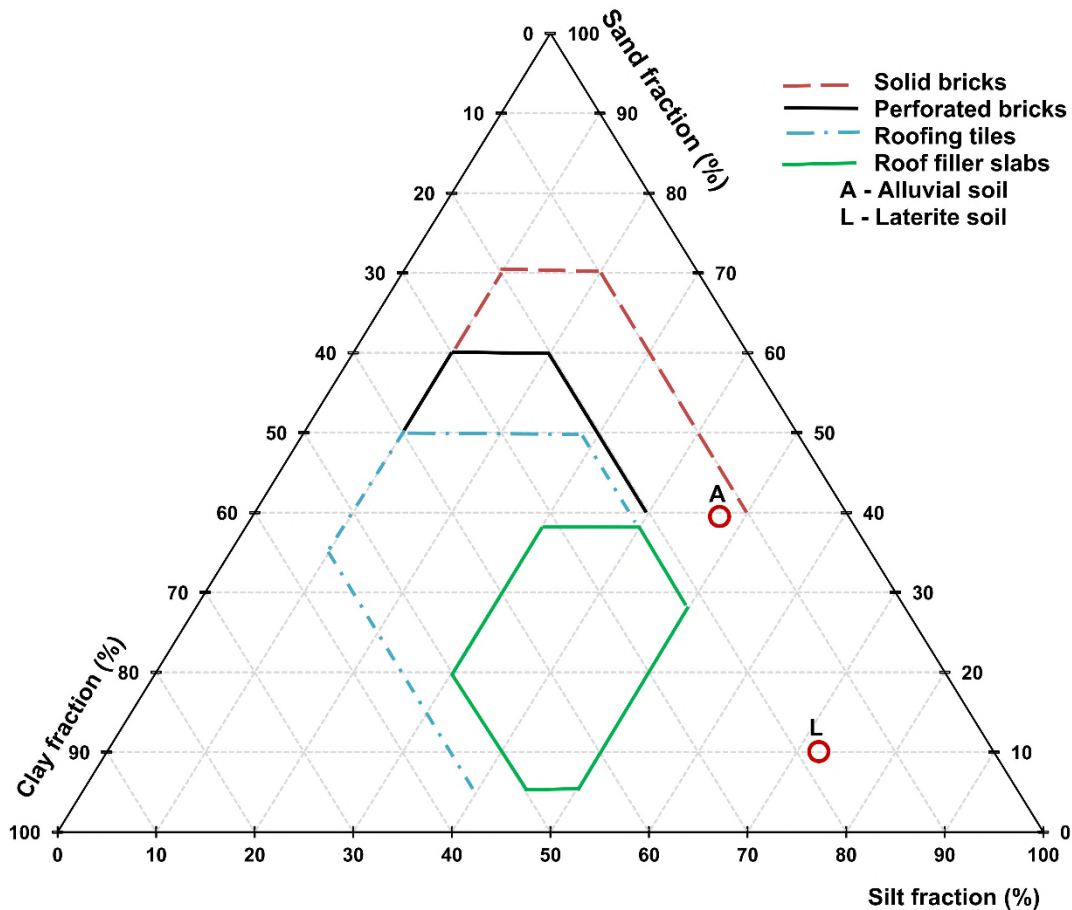
209

210 3.2. Characteristics of the bricks

211 The physical characterization of the developed bricks such as shrinkage, porosity, loss on
212 ignition, dry weight, and bulk density was evaluated on the specimens. The results of the mean
213 dry weight of unfired bricks are presented in Table 1. From the table, it is apparent that the
214 increase in the replacement percentage has a significant influence on the dry weight of the
215 bricks. As the replacement percentage of soils increased from 5-20% the corresponding dry
216 weight also decreased by 15-17% making the bricks lighter. This was observed both in the
217 cases of laterite and alluvial soils used. However, the bricks were lighter when PMSC was
218 used.

219 From the results obtained (Table 1), it can also be seen that the unfired bricks containing
220 laterite soil showed a slightly better packing of particles while moulding, which resulted in
221 higher bulk densities. The reasons for this effect are in the particle size distribution of the used
222 soils presented earlier [2]. Particle size distribution, i.e. texture, plays a significant role in brick
223 raw materials in terms of their behavior in the technological process. A Winkler diagram
224 showing the particle size distribution of both soils has been provided in Fig. 4 [40]. Researchers
225 [41, 42] found that it is necessary to have an adequate particle size distribution to prevent the
226 occurrence of errors in the technological process during the production of bricks. Of course,

227 there is a connection between the particle dispersion and the mineralogical composition of the
 228 brick clays. An increase in the dispersion of brick clays and the content of fine particles causes
 229 greater shrinkage, plasticity, sensitivity to drying, bulk density and compressive strength [43].
 230 The behavior is intensified during the firing process.



231

232 Fig. 4. Winkler diagram showing particle size distribution of soils [44].

233 While performing various procedures, brick specimens were recognized to be
 234 comprehensively fired and free from any defects (i.e. bloating, black coring, cracks, and
 235 efflorescence). The appearance of bricks developed from one type of soil was dark -red whereas
 236 the other type of soil gave a brick-red colour. Ferric oxide in laterite soil (9.49%) and alluvial
 237 soil (6.38%) is mainly responsible for the colour of the bricks since the content of carbonates
 238 was similar in both raw materials (Fig. 5). Both soils belonged to the well-sintering group since
 239 they contained more than 5% of Fe_2O_3 [45].

240 Densification parameters after firing in terms of bulk density, shrinkage, compressive
 241 strength, water absorption, and mass loss on ignition were executed to check the acceptability
 242 of the bricks as construction materials, satisfying the criteria set by different standards.

243

244 Table 1

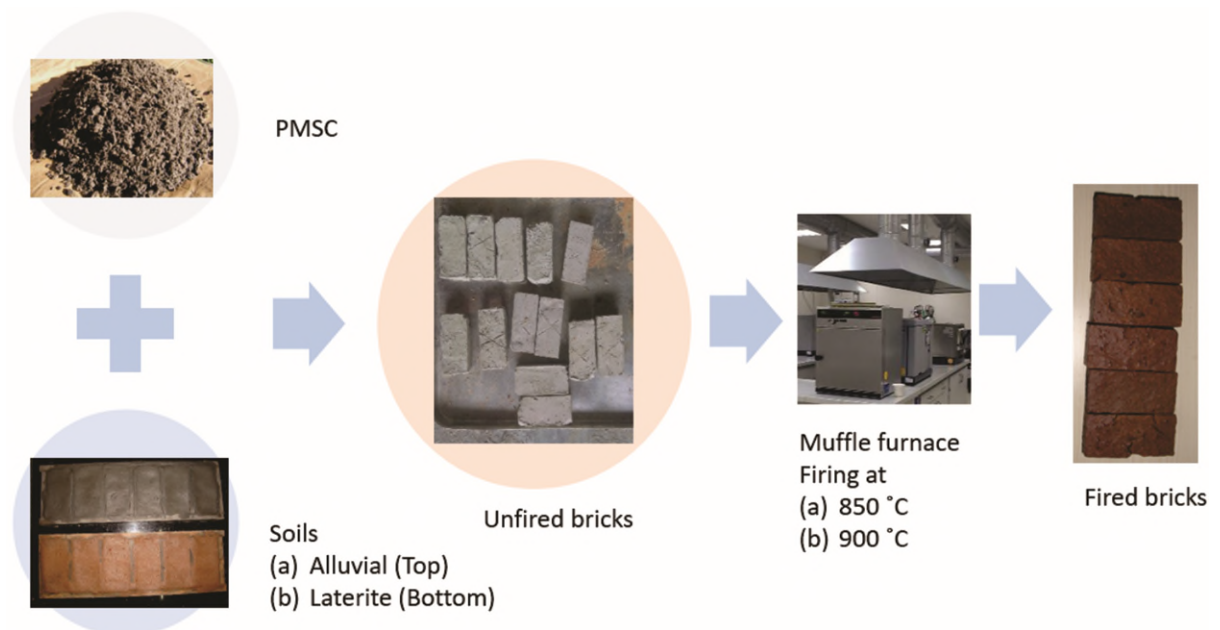
245 The mean oven-dried weights and bulk densities of unfired bricks.

246

Brick labels	Mean oven-dried weight (g)	Bulk density of unfired bricks (g/cm ³)
L	52.69 ± 0.35	1.57 ± 0.12
L 5	46.26 ± 0.29	1.38 ± 0.08
L 10	44.59 ± 0.41	1.33 ± 0.09
L 15	43.52 ± 0.43	1.29 ± 0.13
L 20	41.55 ± 0.37	1.24 ± 0.07
A	49.47 ± 0.51	1.47 ± 0.14
A 5	44.73 ± 0.49	1.33 ± 0.10
A 10	42.87 ± 0.46	1.28 ± 0.11
A 15	41.58 ± 0.55	1.24 ± 0.11
A 20	39.45 ± 0.58	1.17 ± 0.13

247 L- laterite soil, A – alluvial soil

248



249

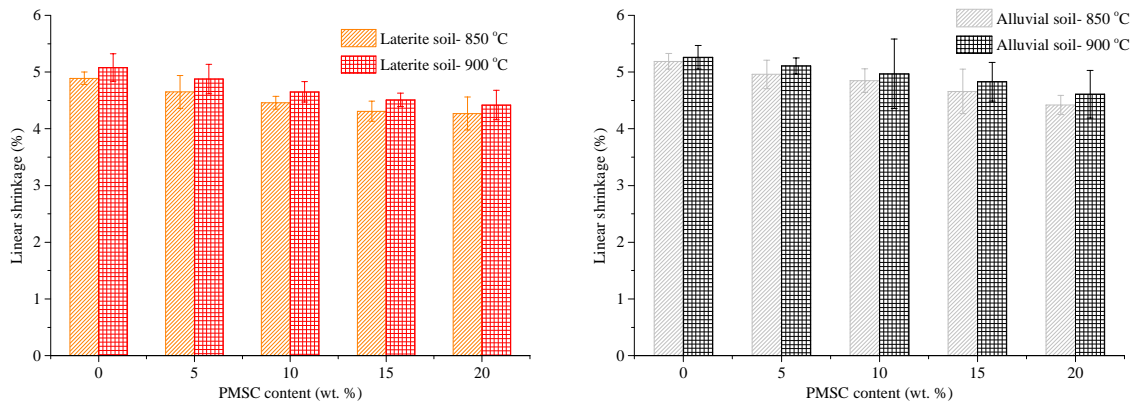
250 Fig. 5. PMSC brick fabrication.

251

252 Figures 6-12 summarise the mechanical characteristics of control bricks and PMSC blended
253 brick samples. Lower firing linear shrinkages were observed for bricks incorporating PMSC
254 for both types of soils. It is observed, as expected, that linear shrinkage, loss on ignition, bulk
255 density, compressive strength and modulus of elasticity increased with the peak firing
256 temperature, while water absorption and apparent porosity decreased. The bricks made of
257 laterite showed lower linear shrinkages since this sample contained more of the so-called filler
258 fraction (0.053 mm – 1.2 mm) that lowers the shrinkage, which was not the case of the alluvial
259 soil [46]. Also, somewhat decreased water absorption and apparent porosity are observed in
260 the case of laterite bricks. As for LOI, bulk density, compressive strength and modulus of
261 elasticity, the laterite bricks showed higher values compared to the alluvial ones. LOI values
262 in laterite bricks were higher because of the higher presence of $\text{Fe}(\text{OH})_3$. The change in bulk
263 density is expected, as explained above (Table 1), as it is intensified by the firing process.
264 Expectedly, linear shrinkage, bulk density and compressive strength are lowered with a higher
265 percentage of PMSC, while LOI increased as observed with other organic wastes [47, 48]. The
266 most intensive differences between the samples, among the studied parameters, are noticed in
267 the case of compressive strength. The compressive strength of the laterite soil (pure or mixed)
268 samples was significantly higher than in the case of products based on the alluvial soil. Besides
269 the better particle packing and higher bulk densities of the products, the reason is in the
270 presence of a small percentage of montmorillonite in laterite soil, which enhanced the
271 mechanical characteristics [46]. From the results, a blend of 10 wt% of PMSC was observed to
272 satisfy engineering properties and optimal solution towards reducing fertile soil consumption
273 in brick making. Also, the minimum compressive strength achieved with this blend of 10 wt%
274 is more than 3.5 MPa, the limit specified by the Indian code of practice, satisfying the
275 requirements of regular clay brick.

276

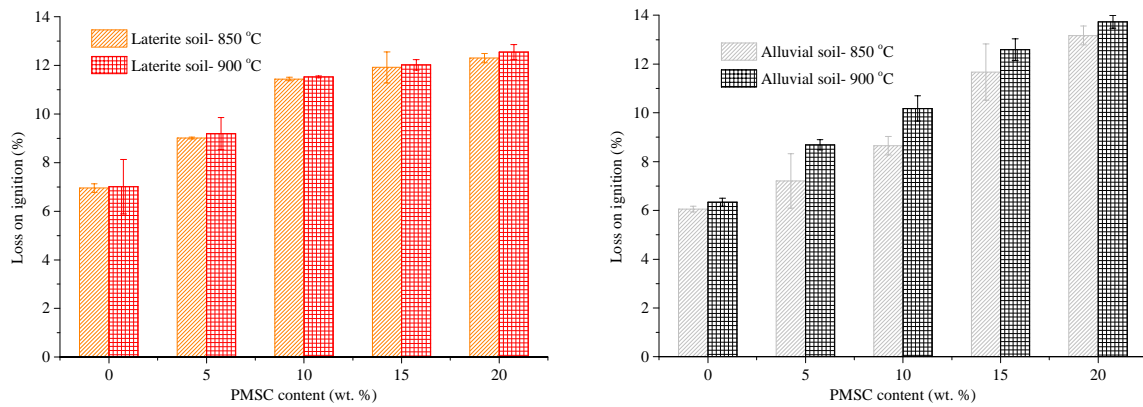
277



278

279 Fig. 6. Linear shrinkage of PMSC mixed bricks (a) Laterite soil blended with PMSC (b)
280 Alluvial soil blended with PMSC – both sintered at (i) 850 °C (ii) 900 °C.

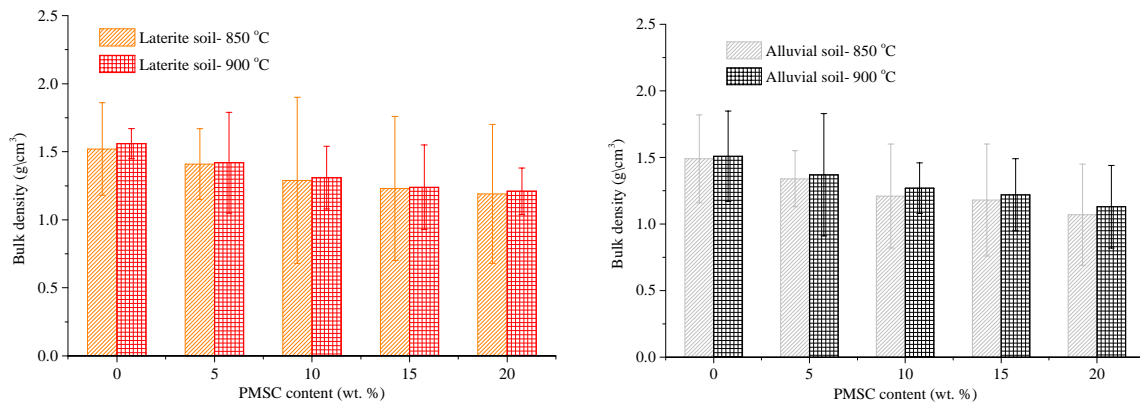
281



282

283 Fig. 7. Loss on ignition of PMSC mixed bricks (a) Laterite soil blended with PMSC (b) Alluvial
284 soil blended with PMSC – both sintered at (i) 850 °C (ii) 900 °C.

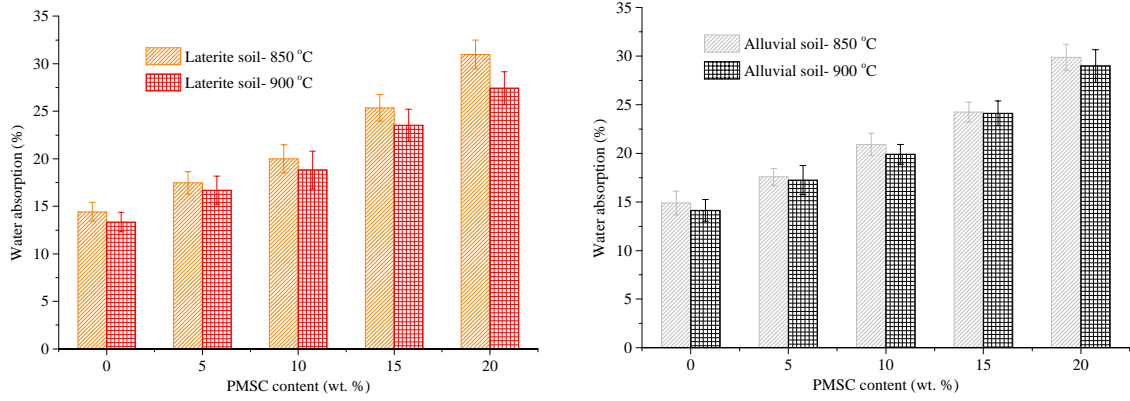
285



286

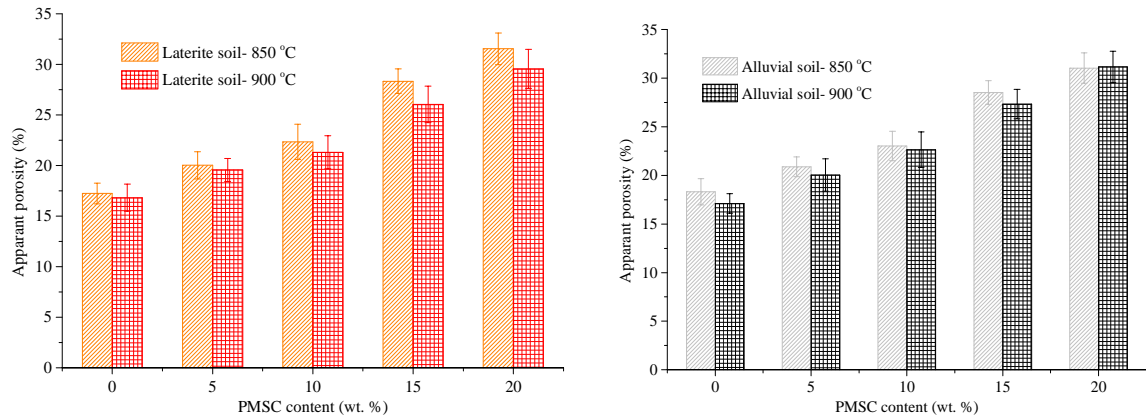
287 Fig. 8. Bulk density of PMSC mixed bricks (a) Laterite soil blended with PMSC (b) Alluvial
288 soil blended with PMSC – both sintered at (i) 850 °C (ii) 900 °C.

289



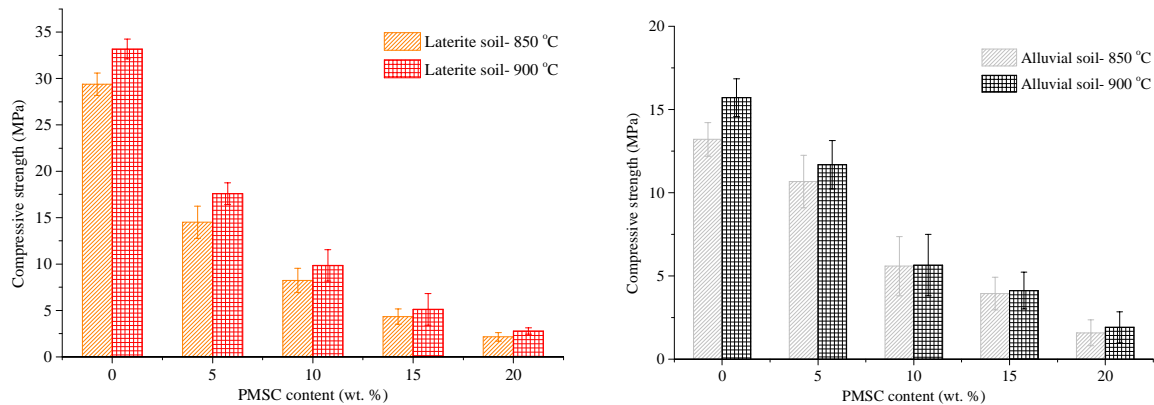
290
291
292
293
294

Fig. 9. Water absorption of PMSC mixed bricks (a) Laterite soil blended with PMSC (b) Alluvial soil blended with PMSC – both sintered at (i) 850 °C (ii) 900 °C.



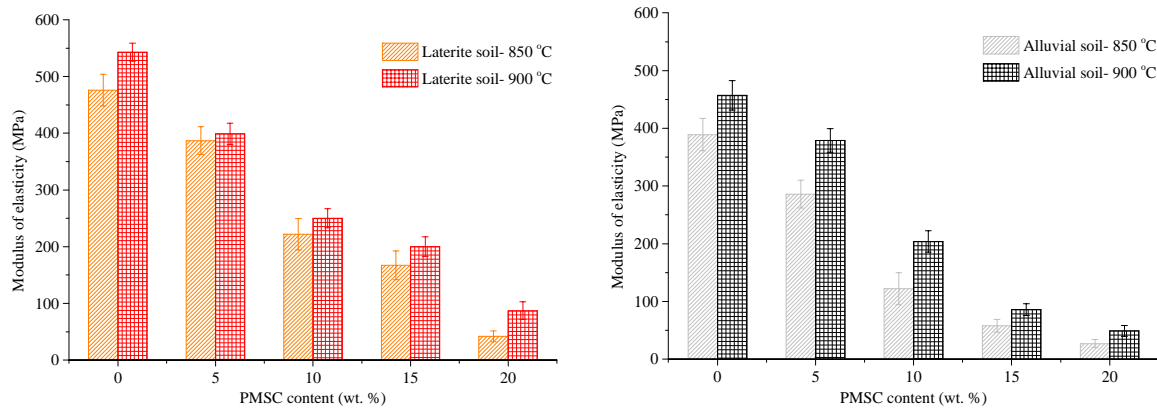
295
296
297
298
299

Fig. 10. Apparent porosity of PMSC mixed bricks (a) Laterite soil blended with PMSC (b) Alluvial soil blended with PMSC – both sintered at (i) 850 °C (ii) 900 °C.

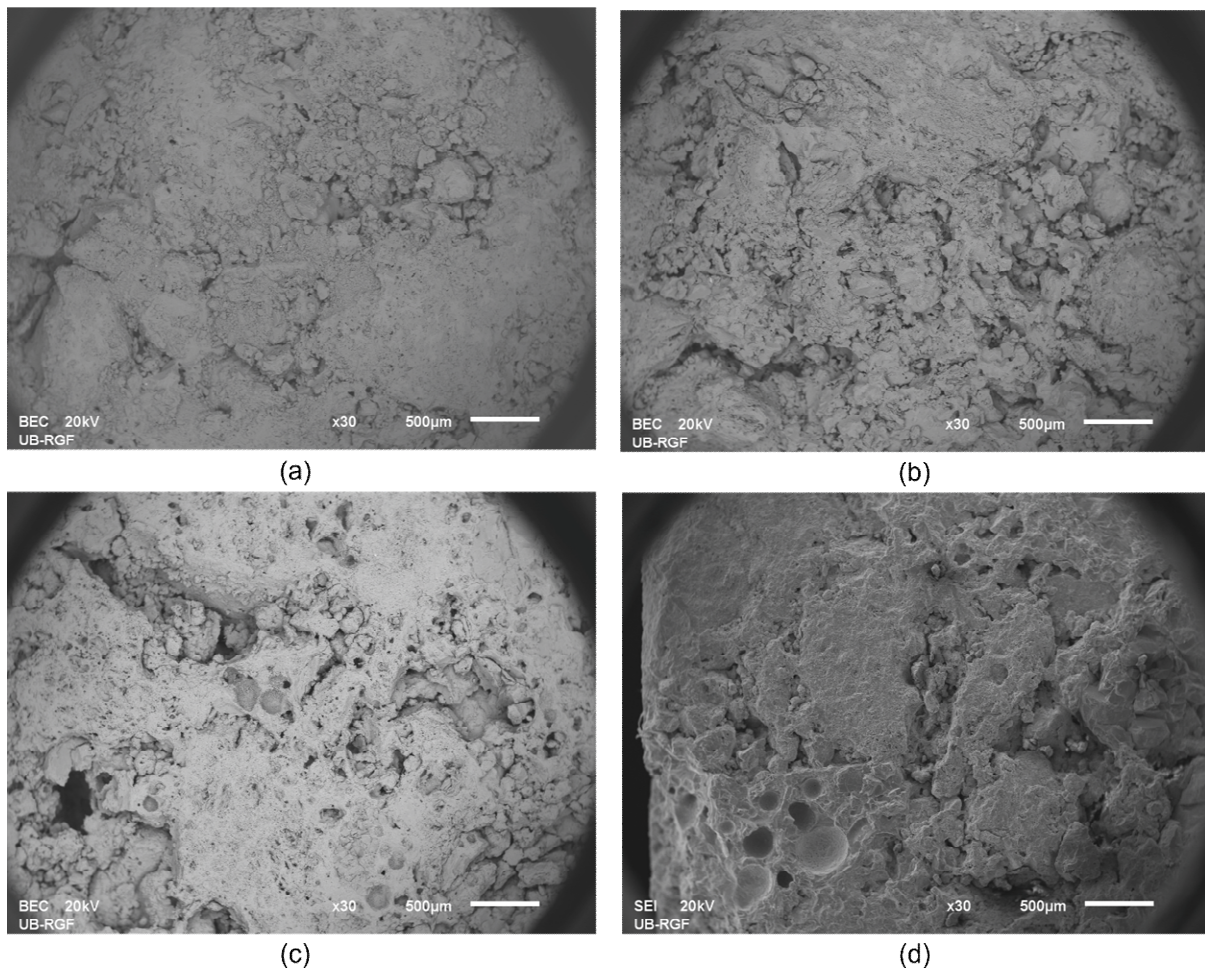


301
302
303

Fig. 11. Experimental compressive strength of PMSC mixed bricks (a) Laterite soil blended with PMSC and (b) Alluvial soil blended with PMSC – both sintered at (i) 850 °C (ii) 900 °C.



304
 305 Fig. 12. Modulus of elasticity of PMSC mixed bricks (a) Laterite soil blended with PMSC and
 306 (b) Alluvial soil blended with PMSC – both sintered at (i) 850 °C (ii) 900 °C.
 307



308
 309 Fig. 13. SEM images of bricks: (a) Laterite soil, (b) Alluvial soil, (c) Laterite soil with 10 wt%
 310 of PMSC, and (d) Alluvial soil with 10 wt% of PMSC. All of these were fired at 900 °C.
 311

312 The microstructure of the bricks made from both soils is shown in Figure 13. The results
 313 of SEM analyses showed that the porosity is improved with the addition of PMSC. The results
 314 confirm higher water absorption/lower compressive strength of bricks made from both of the

315 soils mixed with PMSC. Mineralogical determinations of the fired bricks were conducted using
 316 semi-quantitative analysis and presented in Table 2. Mineralogical composition and their
 317 quantitative presence were found to be almost identical for a soil type fired at a specified
 318 temperature and the occupancy of waste material did not affect it. It is noticeable that some
 319 quantity of quartz is left in all the samples after firing. Some of the $\text{Fe}_2\text{O}_3/\text{Fe}(\text{OH})_3$ was detected
 320 in the raw soil samples [2]. The fired samples contained hematite, which can be of hydroxide
 321 dehydroxylation and break down of the smectite minerals (montmorillonite) in laterite soil [49].
 322 The content of hematite was raised with firing temperature according to the colour of the
 323 products. Calcium and magnesium carbonates are degraded while CO_2 is released. In laterite
 324 samples, Ca is built into the crystal lattice of gehlenite, $\text{Ca}_2\text{Al}[\text{AlSiO}_7]$, after firing at 850 °C
 325 and 950 °C. Mg was introduced to the newly formed spinel phase after firing at a higher
 326 temperature. Aluminosilicates detected in the raw laterite soil transferred to gehlenite at 850
 327 °C, and then partly participated in the formation of spinel after exposure to a temperature of
 328 900 °C. Kaolinite from alluvial soil transformed to mullite after firing at 900 °C.

329
 330 Table 2
 331 Semiquantitative analysis of the mineralogical composition of laterite and alluvial soil bricks
 332 fired at 850 °C and 900 °C.

333
 334

Minerals	ICDD reference card	Laterite soil (%)		Alluvial soil (%)	
		850 °C	900 °C	850 °C	900 °C
Quartz	00-046-1045	61.9	62.1	63.7	63.3
Hematite	00-033-0664	3.0	2.9	-	2.1
Gehlenite	00-035-0755	19.7	12.3	-	1.0
Sillimanite	00-038-0471	15.4	18.6	-	-
Spinel	00-005-0672	-	4.1	-	5.0
Dolomite	00-036-0426	-	-	9.6	-
Carnegieite	00-035-0424	-	-	25.9	-
Rutile	00-021-1276	-	-	0.8	0.5
Mullite	00-002-0415	-	-	-	28.1

335

336

337 3.3. Leaching and emission tests

338 TCLP tests were conducted on crushed samples and outcomes are provided in Table 3.
 339 Permissible values as per EPA regulations and Indian hazardous waste management regulations
 340 are compared. It may be noticed that a 10 wt.% mix of the PMSC is safe to use in fired bricks.
 341 These bricks can safely be used for construction activities and are also safe to dispose of after
 342 the end of life.

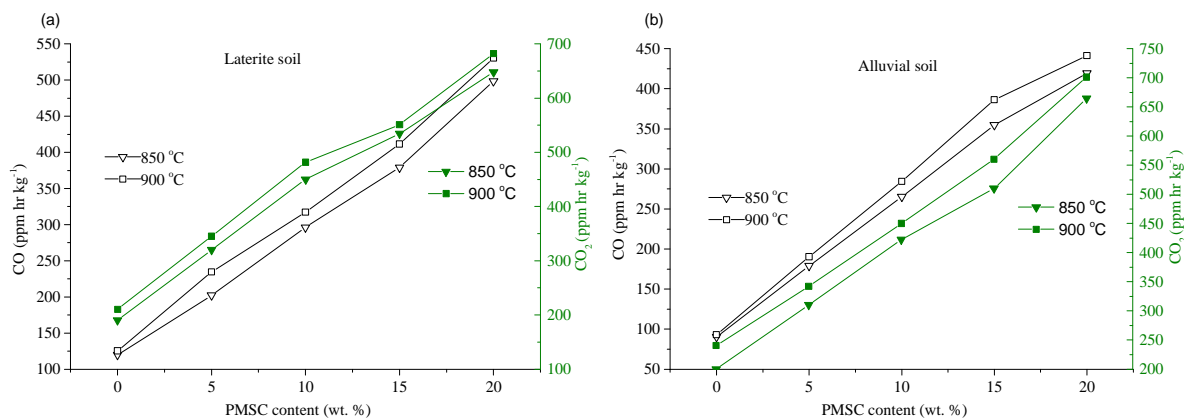
343 Table 3

344 TCLP test results of the laterite soil, alluvial soil bricks incorporating 10 wt% PMSC and
 345 fired at 900 °C.

Elements	Laterite soil (mg/kg)	Alluvial soil (mg/kg)	Permissible limits (mg/kg)
As	6.1	9.3	100
Cd	0.8	0.9	20
Cr	8.3	12.1	100
Pb	14.3	18.5	100
Zn	131.4	234.1	5000
Mn	21.7	28.1	200
Ni	74.5	87.6	400
Co	7.3	11.9	1600

346

347



348

349 Fig. 14. Plots showing estimated total emissions (ETEs) (ppm hr kg⁻¹) during the firing
 350 process.

351

352 There are concerns about pollution due to the firing of bricks, while countries in the Indian
 353 subcontinent rely on biomass and coal for their firing [22]. In addition to greenhouse gases,

354 other gases like sulfur dioxide and oxides of nitrogen may be emitted into the atmosphere. In
355 comparison to an actual brick kiln where the emission will be comprised of fuel and brick
356 components, this study has the advantage of estimating emissions contributed primarily by
357 brick soil and other additives. By measuring the area under the curve "Gas emission per unit
358 mass versus firing time" the emission expenses for CO₂ and CO were determined [33]. These
359 values for (i) alluvial soil, (ii) laterite soil are presented in Fig. 14. The measurements implied
360 lower firing temperature contributes to lower emission, which is in agreement with previous
361 studies. These measurements help get an estimate of greenhouse gas emissions. However, a
362 discerning determination of the emission in real-time conditions could be erroneous as the
363 conditions in the field could vary.

364 3.3.1. Benchmarking of results from previous studies

365 The results obtained in this study have been benchmarked from previous studies in the
366 literature. Engineering parameters of the fired brick such as compressive strength, water
367 absorption, and firing shrinkage have been compared for optimum replacement ratio of the
368 waste. The results are presented in Table 4. Values provided in brackets are a comparison of
369 the respective property concerning control specimen (with no additive) in that study and the
370 values have been rounded to the nearest whole number. It can be observed, all but one studies
371 showed a loss in compressive strength. All the studies show a gain in water absorption with
372 addition of substitute material. Firing shrinkage has been found to be losing in two studies and
373 gaining in one study. Overall trends are linked to increased microporosity within the bricks due
374 to the addition of the organic waste.

375

376 Table 4

377 Comparison of results with previous studies. Values in brackets show the comparison
378 concerning control specimen.

Source	Types of waste and the optimal firing temperature	% of optimal replacement	Compressive Strength (MPa)	Water Absorption (%)	Firing shrinkage (%)
Present work	Paper mill sludge compost (PMSC) @ 900 °C Laterite soil	10	9.97 (- 69 %)	18.65 (+ 41 %)	4.65 (- 8 %)
Goel et al., [2]	Paper mill sludge (PMS) @ 900 °C Laterite soil	10	9.85 (- 70 %)	18.83 % (+ 41 %)	4.61 (- 9 %)
Singh et al., [50]	Deinking Paper mill sludge @ 950 °C	15	5.0 (- 54 %)	28.57 (+ 52 %)	3.06 (+ 8 %)
Arsenović et al., [51]	Sunflower husks, @ 850 °C	5	20.15 (-52 %)	10.48 (+ 14 %)	-
Arsenović et al., [52]	Wooden sawdust, @ 850 °C	2.5	20.97 (- 50 %)	13.9 (+ 33 %)	-
Demir, [53]	Processed Waste Tea @ 900 °C	5.0	22.7 (+ 46 %)	27.30 (+ 52 %)	-

379

380 4. Conclusions

381

382 Increasing number of studies suggest the use of alternative materials in production of fired
383 bricks which is also necessary to meet the obligation towards circular economy. In continuation
384 of this effort, the present study explores the use of paper mill sludge compost (PMSC) as an
385 additive in making fired bricks and proposes guidelines representative of kiln conditions on
386 what parameters should be used to obtain the desired results. This work shows that PMSC is
387 suitable for utilization in fired bricks making. While establishing a strong linkage to the
388 structure-property relationship, this work first of its kind provides useful guidelines to a

389 brickmaker on the use of PMSC in brick making. These discussions lead towards the following
390 reasoning supported by experimental evidence:

391

392 1. The presence of fluxing agents in PMSC as confirmed by XRF is beneficial and helpful
393 in reducing the used energy by lowering firing temperatures which are also critical
394 towards comprehensive firing and minimizing any defects. It was also noticed that
395 cellulose fibers acted as reinforcement and stabilize the magnitude of linear shrinkage
396 in unfired bricks. Laterite soil samples showed a repeatedly higher compressive
397 strength of the brick samples than the alluvial soil samples because of the wider particle
398 size distribution and the presence of a low quantity of montmorillonite.

399 2. From the results, a blend of 10 wt% of PMSC was observed to satisfy engineering
400 properties and optimal solution towards reducing fertile soil consumption in brick
401 making. Also, the minimum compressive strength achieved with this blend of 10 wt%
402 is more than 3.5 MPa, the limit specified by the Indian code of practice, satisfying the
403 requirements of regular clay brick. A higher firing temperature (900 °C) is gainful
404 towards increasing the durability of bricks.

405 3. The commingled 10 wt% PMSC helped lightening of bricks by 14-16% cutback in bulk
406 density. This proposition is a pathway favoring cost-effective construction by the
407 reduction in haulage cost and labor efforts as well as uplifting the state of recyclability
408 while addressing the issue of waste management and promoting the concept of the
409 circular economy. Also due to its calorific value, it reduces the expense of energy
410 during firing.

411 4. Greenhouse gases (CO and CO₂) emission measurements help get the estimate of these
412 values however discerning emission values in real-time conditions could be erroneous
413 as the conditions in the field could vary.

414

415 **Acknowledgments**

416 Gaurav greatly acknowledge the financial support provided by the Royal Academy of
417 Engineering via Grants No. IAPP18-19\295 and EXPP2021\1\277, and EU Cost Action
418 (CA15102, CA18220, CA18224, CA17133 and CA17136). We also acknowledge financial
419 support from the European Regional Development Funds (ERDF) sponsored A2i project at
420 LSBU that has catalyzed several industrial partnerships. Milica V. Vasić acknowledges the
421 financial support received from the Ministry of Education, Science and Technological
422 Development of the Republic of Serbia (Contract No. 451-03-68/2020-14/200012). Nirmal
423 acknowledges the Newton Fellowship award from the Royal Society (NIF\R1\191571).

424

425 **References**

426

- 427 [1] G. Goel, A.S. Kalamdhad, Degraded municipal solid waste as partial substitute for
428 manufacturing fired bricks, *Construction and Building Materials* 155 (2017) 259-266.
- 429 [2] G. Goel, A.S. Kalamdhad, An investigation on use of paper mill sludge in brick
430 manufacturing, *Construction and Building Materials* 148 (2017) 334-343.
- 431 [3] P. Bajpai, *Management of Pulp and Paper Mill Waste*, Springer International Publishing
432 Switzerland 2015.
- 433 [4] S. Seifi, N. Sebaibi, D. Levacher, M. Boutouil, Mechanical performance of a dry mortar
434 without cement, based on paper fly ash and blast furnace slag, *Journal of Building*
435 *Engineering* 22 (2019) 113-121.
- 436 [5] S. Cordiner, G. De Simone, V. Mulone, Experimental–numerical design of a biomass
437 bubbling fluidized bed gasifier for paper sludge energy recovery, *Applied Energy*
438 97(Supplement C) (2012) 532-542.
- 439 [6] A. Demirbas, Progress and recent trends in biofuels, *Progress in Energy and Combustion*
440 *Science* 33(1) (2007) 1-18.
- 441 [7] C. Veluchamy, A.S. Kalamdhad, Enhancement of hydrolysis of lignocellulose waste pulp
442 and paper mill sludge through different heating processes on thermal pretreatment, *J Clean*
443 *Prod* 168 (2017) 219-226.
- 444 [8] Y.H. Yu, S.D. Kim, J.M. Lee, K.H. Lee, Kinetic studies of dehydration, pyrolysis and
445 combustion of paper sludge, *Energy* 27(5) (2002) 457-469.
- 446 [9] H.S. Wong, R. Barakat, A. Alhilali, M. Saleh, C.R. Cheeseman, Hydrophobic concrete
447 using waste paper sludge ash, *Cement and Concrete Research* 70(Supplement C) (2015) 9-20.
- 448 [10] S. Migneault, A. Koubaa, H. Nadji, B. Riedl, S.T. Zhang, J. Deng, Medium-density
449 fiberboard produced using pulp and paper sludge from different pulping processes, *Wood and*
450 *Fiber Science* 42(3) (2010) 292-303.

- 451 [11] S. Kim, H.-J. Kim, J.C. Park, Application of recycled paper sludge and biomass
 452 materials in manufacture of green composite pallet, *Resources, Conservation and Recycling*
 453 53(12) (2009) 674-679.
- 454 [12] M. Sutcu, S. Akkurt, Utilization of recycled paper processing residues and clay of
 455 different sources for the production of porous anorthite ceramics, *Journal of the European*
 456 *Ceramic Society* 30(8) (2010) 1785-1793.
- 457 [13] C.M.F. Vieira, R.M. Pinheiro, R.J.S. Rodriguez, V.S. Candido, S.N. Monteiro, Clay
 458 bricks added with effluent sludge from paper industry: Technical, economical and
 459 environmental benefits, *Applied Clay Science* (2016).
- 460 [14] J.A. Cusido, L.V. Cremades, C. Soriano, M. Devant, Incorporation of paper sludge in
 461 clay brick formulation: Ten years of industrial experience, *Appl Clay Sci* 108 (2015) 191-
 462 198.
- 463 [15] J. Hazarika, U. Ghosh, A.S. Kalamdhad, M. Khwairakpam, J. Singh, Transformation of
 464 elemental toxic metals into immobile fractions in paper mill sludge through rotary drum
 465 composting, *Ecol Eng* 101(Supplement C) (2017) 185-192.
- 466 [16] P. Sonowal, M. Khwairakpam, A.S. Kalamdhad, Stability Analysis of Dewatered Sludge
 467 of Pulp and Paper Mill During Vermicomposting, *Waste Biomass Valori* 5(1) (2014) 19-26.
- 468 [17] A. Lakhout, A.R. Cabral, H. Cabana, Two Novel Biofilters to Remove Volatile Organic
 469 Compounds Emitted by Landfill Sites, *Water, Air, & Soil Pollution* 227(4) (2016) 113.
- 470 [18] A. Khoshand, M. Fall, Geotechnical Characterization of Compost Based Biocover
 471 Materials, *Geotechnical and Geological Engineering* 32(2) (2014) 489-503.
- 472 [19] C.G. Chiochetta, S. Cotelle, J.-F. Masfaraud, H. Toumi, G. Quaranta, F. Adani, C.M.
 473 Radetski, Use of agro-industrial organic sludge amendment to remediate degraded soil:
 474 chemical and eco(geno)toxicological differences between fresh and stabilized sludge and
 475 establishment of application rates, *Environmental Science and Pollution Research* 23(4)
 476 (2016) 3018-3025.
- 477 [20] S. Torii, S. Watanabe, Combustion Characteristics of Combustion Chamber Using
 478 Compost as a Fuel, *Energy Procedia* 61 (2014) 9-12.
- 479 [21] S.N. Monteiro, C.M.F. Vieira, On the production of fired clay bricks from waste
 480 materials: A critical update, *Construction and Building Materials* 68 (2014) 599-610.
- 481 [22] C. Bhushan, D.D. Basu, N.K. Yadav, R. Kumar, National Brick Mission—A scoping
 482 paper, *Roadmap for Brick Kiln Sector: Challenges and Opportunities*, Centre for Science and
 483 Environment, New Delhi, 2016.
- 484 [23] K. Hamer, V. Karius, Brick production with dredged harbour sediments. An industrial-
 485 scale experiment, *Waste Management* 22(5) (2002) 521-530.
- 486 [24] R.V. Silva, J. de Brito, C.Q. Lye, R.K. Dhir, The role of glass waste in the production of
 487 ceramic-based products and other applications: A review, *J Clean Prod* 167 (2017) 346-364.
- 488 [25] S.M.S. Kazmi, M.J. Munir, Y.-F. Wu, A. Hanif, I. Patnaikuni, Thermal performance
 489 evaluation of eco-friendly bricks incorporating waste glass sludge, *Journal of Cleaner*
 490 *Production* 172 (2018) 1867-1880.
- 491 [26] M.V. Madurwar, R.V. Ralegaonkar, S.A. Mandavgane, Application of agro-waste for
 492 sustainable construction materials: A review, *Constr Build Mater* 38 (2013) 872-878.
- 493 [27] S.M.S. Kazmi, M.J. Munir, I. Patnaikuni, Y.-F. Wu, U. Fawad, Thermal performance
 494 enhancement of eco-friendly bricks incorporating agro-wastes, *Energy and Buildings*
 495 158(Supplement C) (2018) 1117-1129.
- 496 [28] A. Ukwatta, A. Mohajerani, N. Eshtiaghi, S. Setunge, Variation in physical and
 497 mechanical properties of fired-clay bricks incorporating ETP biosolids, *Journal of Cleaner*
 498 *Production* 119 (2016) 76-85.

- 499 [29] M.J. Munir, S.M.S. Kazmi, Y.-F. Wu, A. Hanif, M.U.A. Khan, Thermally efficient fired
500 clay bricks incorporating waste marble sludge: An industrial-scale study, *Journal of Cleaner*
501 *Production* 174(Supplement C) (2018) 1122-1135.
- 502 [30] J. Singh, A.S. Kalamdhad, Assessment of bioavailability and leachability of heavy
503 metals during rotary drum composting of green waste (Water hyacinth), *Ecol Eng* 52 (2013)
504 59-69.
- 505 [31] BIS:2117-, Guide for manufacture of hand made common burnt clay buliding bricks (3rd
506 revision), in: B.o.I.S. (BIS) (Ed.) Bureau of Indian Standards (BIS), New Delhi, India, 1991.
- 507 [32] BIS:11650-, Guide for manufacture of common burnt clay building bricks by semi-
508 mechanized process (I revision), in: B.o.I.S. (BIS) (Ed.) Bureau of Indian Standards (BIS),
509 New Delhi, India, 1991.
- 510 [33] G. Goel, A.S. Kalamdhad, A practical proposal for utilisation of water hyacinth:
511 Recycling in fired bricks, *Journal of Cleaner Production* 190 (2018) 261-271.
- 512 [34] J. Singh, A.S. Kalamdhad, Effects of lime on bioavailability and leachability of heavy
513 metals during agitated pile composting of water hyacinth, *Bioresource Technol* 138 (2013)
514 148-155.
- 515 [35] BIS:1077-, Common Burnt Clay Building Bricks -Specification (5th revision), in:
516 B.o.I.S. (BIS) (Ed.) Bureau of Indian Standards (BIS), New Delhi, India, 1992.
- 517 [36] ASTM:C67-14, Standard Test Methods for Sampling and Testing Brick and Structural
518 Clay Tile, in: ASTM (Ed.) Pennsylvania, United States.
- 519 [37] BIS:3495-, Methods of Tests of Burnt Clay Building Bricks, in: B.o.I.S. (BIS) (Ed.)
520 Bureau of Indian Standards (BIS), New Delhi, India, 1992.
- 521 [38] ASTM:D695-15, Standard Test Method for Compressive Properties of Rigid Plastics, in:
522 ASTM (Ed.) Pennsylvania, United States.
- 523 [39] T.K. Dan, B.K. Sarkar, Properties of Bricks from Lateritic Waste, *Ceram Int* 17(6)
524 (1991) 351-357.
- 525 [40] M. Loutou, Y. Taha, M. Benzaazoua, Y. Daafi, R. Hakkou, Valorization of clay by-
526 product from moroccan phosphate mines for the production of fired bricks, *Journal of*
527 *Cleaner Production* 229 (2019) 169-179.
- 528 [41] F. Cardarelli, *Materials Handbook: A concise desktop reference*, Springer-Verlag
529 London, London, 2008.
- 530 [42] H.G.F. Winkler, Bedeutung der Korngrößenverteilung und des Mineralbestandes von
531 Tonen für die Herstellung grobkeramischer Erzeugnisse, *Berichte der Deutschen*
532 *Keramischen Gesellschaft* 31 (1954) 337-343.
- 533 [43] M.L. Diko, G.E. Ekosse, S.N. Ayonghe, E.B. Ntasin, Physical characterization of clayey
534 materials from tertiary volcanic cones in Limbe (Cameroon) for ceramic applications,
535 *Applied Clay Science* 51(3) (2011) 380-384.
- 536 [44] H. Winkler, Bedeutung der Korngrößenverteilung und des Mineralbestandes von Tonen
537 für die Herstellung grobkeramischer Erzeugnisse, *Berichte der Deutschen Keramischen*
538 *Gesellschaft* 31(10) (1954) 337-343.
- 539 [45] A.V. Kornilov, Reasons for the different effects of calcareous clays on strength
540 properties of ceramics, *Glass and Ceramics* 62(11) (2005) 391-393.
- 541 [46] M. Arsenović, Optimization and prediction of the quality of materials, process and final
542 properties of heavy clay products by mathematical modeling of the characteristic parameters,
543 Faculty of technology and metallurgy, University of Belgrade, Serbia, 2013.
- 544 [47] A. Ukwatta, A. Mohajerani, Effect of Organic Content in Biosolids on the Properties of
545 Fired-Clay Bricks Incorporated with Biosolids, *Journal of Materials in Civil Engineering*
546 29(7) (2017) 04017047.
- 547 [48] A. Yaras, Combined effects of paper mill sludge and carbonation sludge on
548 characteristics of fired clay bricks, *Construction and Building Materials* 249 (2020) 118722.

- 549 [49] M. Arsenovic, Z. Radojevic, S. Stankovic, Removal of toxic metals from industrial
550 sludge by fixing in brick structure, *Construction and Building Materials* 37 (2012) 7-14.
- 551 [50] S.K. Singh, S. Kulkarni, V. Kumar, P. Vashistha, Sustainable utilization of deinking
552 paper mill sludge for the manufacture of building bricks, *Journal of Cleaner Production* 204
553 (2018) 321-333.
- 554 [51] M. Arsenović, Z. Radojević, Ž. Jakšić, L. Pezo, Mathematical approach to application of
555 industrial wastes in clay brick production – Part I: Testing and analysis, *Ceramics*
556 *International* 41(3, Part B) (2015) 4890-4898.
- 557 [52] M. Arsenović, Z. Radojević, Ž. Jakšić, L. Pezo, Mathematical approach to application of
558 industrial wastes in clay brick production—Part II: Optimization, *Ceramics International*
559 41(3, Part B) (2015) 4899-4905.
- 560 [53] I. Demir, An investigation on the production of construction brick with processed waste
561 tea, *Building and Environment* 41(9) (2006) 1274-1278.

562