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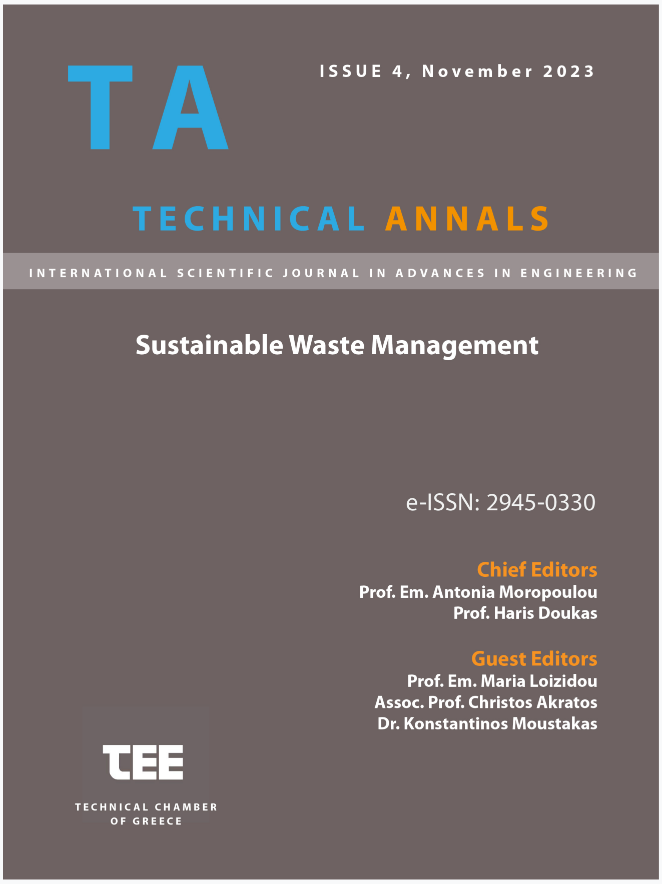

Sustainable management of end-of-life creosote-treated wood poles sawdust into red ceramics for environmental and health protection

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Sustainable management of end-of-life creosote-treated wood poles sawdust into red ceramics for environmental and health protection

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Abstract. In the current original research, sintered red ceramics, incorporated with creosote-impregnated timber (CIT) in the form of sawdust as a useful porous making additive, were manufactured and characterized. Actually, safe management of creosote-treated electricity and telecommunication poles waste and its utilization as a useful secondary raw material into materials of added-value, represents an important priority towards circular economy and equally a great challenge given potential health risks reported in association with reuse of creosote-containing materials. The full life cycle of creosote-treated electricity and telecommunication poles includes: the growth of trees on forested land, harvest of logs, milling to produce poles and lumber; then creosote treatment of poles and their use by the Electricity and Telecommunication Grids; and finally disposal in landfills or valorization as an energy source at the end of their use lives. This paper reports results of evaluation performed to determine the possibility of using end-of-life poles in the form of sawdust as additive (pore making agent) in building ceramic industry. End of Waste (EoW) criteria will be implemented to assess the safe use of this waste by-product as secondary resource, for ensuring environmental and human health protection.

Keywords: Creosote-treated wood poles, end-of-life, sawdust, red ceramics, environmental, health protection, sustainability.

1 Introduction

A combination of coal tar called creosote oil (tar oil) contains, among other things, chemicals from the phenol, cresol, and xylenol groups in different proportions, depending on the production method [1]. It belongs to the oldest impregnating substances used in the preservation of wood, since ancient times. It is a viscous oily liquid with a tar smell, which is produced by the dry distillation of coal [2]. This substance exhibits satisfactory penetration, minimal leakage and strong toxicity towards microorganisms

[3]. Its disadvantages are that it gives the wood a dark brown shade and a strong smell, for which are responsible the drops of oil that appear on the surface of the wood, as a result of which it is not amenable to painting or adhesive substances.

Creosote [4] is a brownish-black oily liquid and is a distillation product of coal tars which themselves are by-products of the high-temperature destructive distillation of bituminous coal to form coke. Creosote is the intermediate cut, ranging from 200 to 355ºC.

Creosote is a complex mixture of hundreds of distinct substances, including bi- and polycyclic aromatic hydrocarbons (PAH's), phenols as well as heterocyclic, oxygen-, sulphur- and nitrogen-containing substances. On average 35-43% of creosote remains unidentified. Creosote typically contains more than 300 compounds that are categorized into five main classes of compounds, as follows [5]:

- Aromatic hydrocarbons including polycyclic aromatic hydrocarbons (PAHs, up to 90% of creosote), benzene, toluene and xylene
- Phenolics including phenols, cresols, xylenols and naphthols (1 to 3 % of creosote)
- Nitrogen-containing heterocyclics including pyridines, quinolines, acridines, indolines, carbazoles (1 to 3 % of creosote)
- Sulphur-containing heterocyclics including benzothiophenols (1 to 3 % of creosote)
- Oxygen-containing heterocyclics including dibenzofurans (5 to 7.5 % of creosote)

European creosotes must comply with EN 13991. In Directive 2011/71/EU this was specified as "Grade B or Grade C creosote as specified in European Standard EN 13991:2003".

Wood as a biological material is attacked by fungi, bacteria, insects, marine organisms wood-eating organisms, which find food and shelter in it. Impregnation of wood with creosote oil (creosote or tar oil) is a method based on in oil-soluble petrochemical fractions (creosote oils), which is widely used to protect pillars of electricity and telecommunication networks from the attacks described above. The under pressure impregnated wood (forest or black pine) has a dark black to brown color, unpleasant smell, especially when it is "fresh", and has excellent dimensional behavior and very extended life time.

However, wood treated with creosote contains several toxic substances. It is still in use in almost all of Europe, even in Scandinavia, although it was predicted that from in 2004 this technique would be phased out. The CIT is a hazardous waste, after its withdrawal from train lines and Electricity distribution as well as Telecommunication Grids. Wooden poles Impregnated with Creosote may offer a lifecycle of more than 40years.

Material Safety Data Sheet (MSDS) [6] for creosote-treated wood in its disposal considerations, proposes that because the smoke and ashes from burning treated wood can contain harmful compounds, it is not recommended to use it in open fires, stoves, fireplaces, or domestic boilers. Alternatively, creosote-treated wood may be incinerated only in commercial or industrial burners or grate-fired boilers and fluidized bed combustion kilns, in accordance with local, regional and national regulations. However, this necessitates that the wood be prepared (shredded) to ensure full incineration and that the temperature at which it is burned is high enough [7].

It should be noticed that reuse of materials that were previously treated with creosote pose a potential danger for environmental and human health, as they contain harmful polycyclic aromatic hydrocarbons (PAHs), nitrogen-containing heterocyclic compounds and other already mentioned substances. Actually, a significant percentage of the creosote total weight is made up of several PAHs, which are considered to be important pollutants and potential carcinogens [8-11]. PAHs can leach into the surrounding water from creosote-treated wood as well as the nitrogen heterocycles that leach more quickly and intensely than the PAHs). Moreover, creosote can contaminate soil in preservation facilities. Because of storage, procedures, tools, and waste treatment of chemicals that are part of the composition of creosote, older wood preservative sites have extensive soil, sediment, and sludge pollution, which makes remediation of these sites more difficult. Thus, risks for human, animal and environmental/ecological health arise from preservatives like creosote that build up in soil and then bioaccumulate in fruits and vegetables [12-15].

Nevertheless, both industries and researchers have tested and experimented with industrial symbiosis solutions that enable the utilization of waste from other industries in ceramic manufacturing [16-20]. Waste valorization technologies in building ceramic industry, are mostly at diverse stages of their maturity (TRL2-3 to TRL7-8) [21]. Sawdust is a common porous making agent in the building ceramic industry [22-26].

In the present research work, safe management of waste poles impregnated with creosote oil, at the end of their life, in the form of sawdust as porous making agent in red ceramics is investigated, complying with the EoW criteria, in accordance to circular economy principles, in the context of sustainable development. For that purpose, clay-based ceramic bodies containing CIT in the form of sawdust were formed, sintered and characterized.

2 Materials and Methods

2.1 Raw Materials

In this study, three different clays A, B and C (received from Central Greece) were utilized. A clay blend, consisting of 50% of clay type A, 33% of clay type B, and 17% of clay type C, commonly utilized for making standard ceramics (Terra SA), was employed for creating prototype (pr) clay samples and samples containing 8% CIT. The chemical analysis of the clays was conducted based on the EN 196-2 standard, and the findings are outlined in Table 1.

Table 1. Chemical analysis of the clayey raw materials

	Composition (%)										
Clay	**Loss on Ignition (LOI)**	**SiO_2**	**Al_2O_3**	**Fe_2O_3**	**CaO**	**MgO**	**K_2O**	**Na_2O**	**TiO_2**	**$CaCO_3$ (eq)**	**CO_2**
A	11.9	49.4	12.9	7.1	8.6	4.9	2.9	1.6	0.8	14.7	6.5
B	9.9	52.8	13.5	7.6	6.3	4.3	3.2	1.6	0.9	-	-
C	16.5	51.0	8.5	4.7	11.6	3.9	1.5	1.4	0.6	24.5	10.7

In Fig. 1, the Creosote-treated wood at the end of its life, which was used in this research, is presented. The impregnated region (wood mavro) is clearly distinguished from the inner region of the pole (wood leuko) – 70% and 30% respectively.

Fig. 1. Creosote-treated wood waste 70% 'mavro' (black) – 30% 'leuko' (white)

In Fig. 2, shredded wood from the above mentioned regions has been used for GC/MS volatile components analysis.

Fig. 2. (a) Shredded wood 'leuko'; (b) Shredded wood 'mavro'

2.2 Ceramic specimen fabrication and characterization

Ceramic bodies were produced by incorporating 0% and 8%wt. CIT sawdust (mixed mavro and leuko) in the aforementioned clay mixture. The particle size distribution of the incorporated CIT sawdust lies between 1mm – 4mm. A schematic diagram of the proposed method is shown in Figure 3.

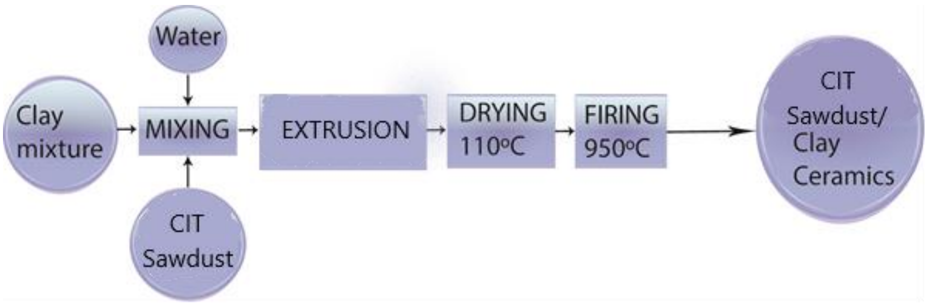

Fig. 3. A schematic representation of the process for the production of CIT sawdust/clay-based red ceramic specimens.

The clay and sawdust blend was mixed with water to create the right malleable texture for shaping specimens with a manual cutter on a pilot-plant vacuum extruder. The specimen bars had a rectangular cross section and measured 80 mm × 43.5 mm × 18 mm. Test specimens were weighed to determine moisture content, then subjected to natural drying at room temperature for 12 hours and forced drying at 110°C until a constant weight was achieved. Test pieces that had been dried were subjected to a controlled temperature increase in a programmable electric chamber furnace, reaching a peak temperature in just 15 minutes to reduce energy spending. The temperature of 950°C was the final sintering temperature that was adopted.

The water absorption and three-point bending strength measured by modulus of rupture (MOR), were assessed on fired samples following the ASTM C67 guidelines. The Anter Unitherm Model 2022 measured the thermal conductivity coefficient (k) at 25°C using the guarded heat flow meter method. The examinations are conducted following the guidelines of ASTM E1530. The results show the impact of adding CIT waste sawdust to the clay mixture, and represent the average of measurements taken from 5, 7, and 3 specimens.

Determination of volatile components contained in CIT and clay/CIT mixture, before and after mixing with clay and sintering was investigated by GC/MS Analysis. Leachates were obtained by following the HS-SPME [27] method for the treated wood and the ceramic bodies containing 8 %wt CIT waste sawdust.

3 Results and Discussion

It was observed during specimen formation that the green clay mixture plasticity and extrusion behavior were influenced in a certain degree by the addition of 8%wt CIT sawdust.

The magnitude of weight loss, cold water absorption, modulus of rupture and thermal conductivity, resulting from the incorporation of 8 %wt CIT sawdust into clay mixture sintered at 950ºC, compared to 0 %wt CIT sawdust specimen are provided in Table 2. The specimen weight loss, cold water absorption, modulus of rupture and thermal conductivity, resulting from the incorporation of 8%wt CIT sawdust into clay mixture sintered at 950ºC, are diverged to a large extend in comparison to 0%wt CIT sawdust specimen. This is an expected consequence of the 8%wt CIT sawdust content in the clay mixture. It is apparent that the embodiment of CIT waste sawdust in considerable proportions (8%wt.) in the clay mixture, slightly deteriorates the modulus of rupture, whereas it decreases the thermal conductivity values, which is beneficial from the thermal insulation properties aspect.

An optimization study will determine the suitable %wt CIT sawdust in the clay mixture and the optimal particle size distribution of the incorporated CIT sawdust, so that article 6 (c) of Directive 2008/98/EC be fulfilled [28].

Determination of volatile components contained in the sintered specimen, compared to the volatile components contained in the CIT waste sawdust (see Table 3), proves that the volatile components are degraded during the sintering process, fulfilling 6 (d) of Directive 2008/98/EC. Thus, potential health risks reported in connection with applications of reused creosote-treated materials are here avoided.

Table 2. Characterization results on sintered ceramic specimens:
(a) Weight Loss (%) and Total Volume Shrinkage, TVS (%)

Tsint (ºC)	**CIT (% wt.)**	**Specimen**	**Sintering weight loss (%)**	**Variation (%)**	**Total Volume Shrinkage (%)**	**Variation (%)**
950	0	40	6,49		2,29	
	8	40	9,47	**45,92**	0,35	**84,69**

b) Water absorption WA (%) (Cold water), WA (%) (Boiled water),
3 – Point Bending MOR (MPa), Thermal Conductivity k (W/m*K)

Tsint (ºC)	CIT (% κ.β.)	Cold Water Absorption (%)	**Variation (%)**	**Boiled** Water Absorption (%)	**Variation (%)**	MOR (MPa)	**Variation (%)**	k (W/m*K)	**Variation (%)**
950	0	15,16		17,60		8,78		0,499	
	8	19,88	**31,13**	**23,51**	**33,58**	5,38	**38,72**	0,398	**20,24**

Table 3. GC/MS Analysis: volatile components of the CIT sawdust and the sintered specimen

No	RT (min)	Volatile compound	Area % LEUKO	Area % MAVRO	Area % Bricks 8% CIT sawdust
1	1.992	Propane	0.21	0.02	-
2	3.106	α-Pinene	27.08	0.05	-
3	3.688	(1R)-α-Pinene		0.03	-
4	4.536	D-Limonene	8.36		-
5	4.547	Indene		0.25	-
6	5.625	L-Fenchone	0.58		-
7	5.849	7-Methylbenzofuran	1.59		-
8	5.936	2-Methylbenzofuran		0.11	-
9	6.326	(+)-Fenchol	0.48		-
10	6.919	(+)-2-Bornanone	1.01		-
11	7.209	Camphor	0.47		-
12	7.267	3-Methyl-1H-indene		0.13	-
13	7.34	1-methyl-1,2-propadienyl-benzene	0.56		-
14	8.206	Naphthalene	13.45	3.18	-
15	8.748	α-Terpineol	6.62		-
16	9.664	Isoquinoline		0.35	-
17	9.739	α-Methylene-benzeneacetonitrile	0.88		-
18	10.388	2-Isopropyl-1-methoxy-4-methylbenzene	0.22		-
19	10.46	Quinoline		0.02	-
20	11.402	6-Methyl-benzothiophene	0.08		-
21	12.08	1-Methylnaphthalene	13.48	5.31	-
22	12.517	2-Methylnaphthalene	6.37	2.61	-
23	13.394	2-Naphthalenamine		0.03	-
24	14.087	7-Methylquinoline		0.11	-
25	14.381	α-Terpinyl acetate	0.38		-
26	14.914	Biphenyl	2.84	3.22	-
27	15.488	1-Ethylnaphthalene	0.88		-
28	15.741	2-Ethylnaphthalene	0.08	1.02	-
29	15.982	2,7-Dimethylnaphthalene	1.48	2.28	-
30	16.471	1,3-Dimethylnaphthalene	1.14	2.90	-
31	16.567	1,6-Dimethylnaphthalene	0.81	1.34	-
32	17.162	1,4-Dimethylnaphthalene	0.70	0.52	-
33	17.607	2,3-Dimethylnaphthalene	0.13		-
34	18.74	Acenaphthene	5.12	8.07	-
35	18.988	3-Methyl-1,1'-biphenyl	0.48		-
36	19.268	4-Methyl-1,1'-biphenyl	0.13		-
37	19.722	1-Isopropenylnaphthalene	0.14	1.62	-
38	19.944	Dibenzofuran	1.57		-
39	20.411	α-Muurolene	0.07		-
40	20.599	1,6,7-Trimethylnaphthalene	0.12	8.88	-
41	20.778	1,4,6-Trimethylnaphthalene	0.11	0.69	-
42	21.323	2,3,6-Trimethylnaphthalene	0.07	0.26	-
43	21.966	1,4,5-Trimethylnaphthalene	0.06		-
44	22.239	1H-Benzonaphthene		4.13	-
45	22.372	Fluorene	0.90	7.89	-
46	25.124	Fluorene-9-methanol		4.52	-
47	25.972	Diphenylketene		5.92	-
48	26.76	Ethionamide		2.48	-
49	27.614	9-Methyl-9H-fluorene		1.30	-
50	28.08	2-Methyl-9H-fluorene		1.38	-
51	28.358	2-Methyl-1,1'-biphenyl	0.31	0.51	-
52	29.946	4,5-Benzothionaphthene		2.78	-
53	31.564	Diethyl Phthalate	0.25	3.32	-
54	33.073	4-Methyldibenzofuran	0.09	5.60	-
55	35.711	4-Biphenylylcarboxaldehyde	0.06	4.66	-
56	39.733	Anthracene	0.10	10.78	-
57	45.348	Isobutyl phthalate	0.53	1.74	-

4 Conclusions

Recent trends in wood processing include safe use of wood preservatives, the development of wood modification methods, and the recycling and disposal of treated materials. In this framework, the incorporation of 8 %wt CIT sawdust into red ceramics was investigated in the present work. Sawdust influences in a certain degree the green clay body plasticity and extrusion behavior during specimen fabrication. The characterization results on sintered red brick products show that the embodiment of CIT waste sawdust in noticeable proportions (8%wt.) in the clay mixture sligthly impacts on strength values, while also decreases thermal conductivity, which will be beneficial to red brick thermal insulation behavior. An optimization study would be necessary to determine the optimal both %wt CIT sawdust in the clay mixture and particle size distribution of the incorporated CIT sawdust in the final ceramic product, in view of fulfilling the technical standards for specific applications and meeting the existing legislation requirements for building materials.

Particularly, the comparison of volatile components contained in the sintered specimen to those identified in the CIT waste sawdust, proves that the volatile components are degraded during the sintering process and, as a result, potential use of the final ceramic product will not cause any negative effects on the environment or human health.

A pilot-scale study (TRL 7–8) should follow the present laboratory scale research, to ensure that possible unburned volatile components in the flue gases will be traced and securely retained, satisfying and fulfilling the End of Waste (EoW) criteria for CIT sawdust through its proper incorporation in clay mixtures to produce high quality and environmentally safe red ceramic building materials while also avoiding overall environmental and human health risks. Furthermore, in addition to addressing current issues, the wood treatment sector should go forward to remediate environmental sites contaminated by wood preservation operations and also develop and embrace innovative recycling and valorization research advancements, practical solutions and supportive policies, towards a sustainable circular economy.

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