



Comparative Study of Heavy Metal Leaching from Clay – Tannery Sludge – Glass Powder Bricks using TCLP and Netherlands Tank Leaching Methods

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Abstract: The work compared the leaching of heavy metals (Cr, Ni, Zn, and Pb) from the hybrid bricks using the Toxicity Characteristics Leaching Procedure (TCLP) and Netherlands tank leaching tests. Hybrid bricks were prepared by partially replacing clay with an increasing weight percentage of tannery sludge (0, 9, 18, and 27% by weight of brick specimen) and a constant weight percentage (10% by weight of brick) of glass powder. The prepared mixes were fired to 900oC, 950oC, and 1000oC to assess the leaching potential of metals from hybrid bricks at different temperatures. The TCLP test was conducted as per the United States Environmental Protection Agency (USEPA) 1311, whereas the Netherlands tank leaching test was performed as per the NEN 7345 standard. In both methods, the leaching of metals increased with the tannery sludge amounts and decreased with an increase in temperature. The release of metals from the TCLP test was slightly higher than that of the Netherlands tank leaching test. The higher amounts of metal released from the TCLP test were due to exposure to more areas of particles due to the crushing of particles. The results obtained from the TCLP test are more appropriate to the actual field conditions arising in the service life of materials.

Keywords: *leaching of metals; USEPA 1311; bricks; NEN 7345.*

1. Introduction

Leather, a commercial material, has multiple purposes and can be used to manufacture bags, belts, foot ware, wallets, etc. The hides of animals are subjected to several processes, such as pre tanning, tanning, and finishing, to produce commercial leather. The leather generation method involves the use of large amounts of water and salts to clean the hides. Chromium is one of the major salts used in the tanneries. During the tanning process, a significant amount of liquid and solid wastes are generated. The solid waste produced in the tanning process is called the tannery sludge and is treated as hazardous waste because of organic matter and high amounts of Cr salts [1]. Because of the toxic nature of the tannery sludge, the sludge is disposed of in secured landfills. The continual increase of tannery sludge each year necessitated reutilizing it to reduce the landfilling [2]. Some of the previous studies showed that industrial sludges could be reutilized in construction materials to reduce the landfilling of sludge [3] The utilization of sludge in construction resulted in a decline in the properties of the materials because of the organic matter

in the sludge. However, using glass along with the raw materials increased the properties of bricks because of the binding action of the molten glass powder [4]. The combined utilization of tannery sludge and glass powder in clay bricks can overcome the harmful effects of using tannery sludge and can be compensated by the glass powder. Considering the loss of adhesiveness because of excess utilization of glass powder, utilization of glass powder in clay bricks was limited to ten percent by weight of wet brick. Variations in the properties of bricks were assessed by changing the weight percentages of tannery sludge in the bricks [5]. Most of the sludges contain heavy metals and hence necessitated the need to study the leaching of heavy metals from the prepared specimens. Previous studies considered the leaching of heavy metals from the materials using the TCLP test or Netherlands tank leaching tests [6]. No prior studies compared the leaching of heavy metals from the TCLP test and the Netherlands tank leaching test. Hence, in the present study, an attempt was made to compare the leaching of heavy metals in both cases. The results help in understanding the leaching process of metals in both cases and help to identify the most appropriate method among the two.



2. Materials and Methods

2.1. Materials

Soil, tannery sludge, and glass powder were used in the present investigation. A common effluent tannery treatment plant in Ranipet, Tamil Nadu, India, provided the tannery sludge. The provided tannery sludge was moist and subjected to air drying, followed by oven drying at 110°C for 24 hours. After oven drying, the material showed lumps. The lumps in the oven-dried material were crushed using a hammer and converted to powder [7]. The lumps in the oven-dried material were crushed using a hammer and converted to powder. The powdered material was preserved in plastic bags to avoid interaction with moisture in the atmosphere. The soil sample was provided by a local brick manufacturer in Kadapa, Andhra Pradesh, India. The soil sample was oven-dried at 110°C for 24 hours to remove the traces of moisture. The dried sample was crushed to make it free from lumps. The borosilicate glass was collected from the Chemistry laboratory of the RGUKT RK Valley campus, Kadapa district, Andhra Pradesh, India. The collected glass was crushed to a fine size using a hammer. All the raw materials were sieved through a 150-micron IS sieve, and the materials finer than 150 microns were used to prepare the bricks.

2.2. Preparation Of Bricks

The raw materials were mixed with water and made into a homogenous mixture. Specimens were prepared to have the glass powder of ten percent by weight of the wet brick, and the tannery sludge was kept at nine, eighteen, and twenty-seven percent by weight of the wet brick [8]. The size of the wet bricks was 0.22 x 0.1 x 0.07 m³. The prepared wet specimens were fired to 900°C, 950°C, and 1000°C at 5°C/min in a muffle furnace. Bricks were prepared only with the soil and were considered as the reference specimens.

2.3. Tests performed on bricks

Tannery sludge contains heavy metals and can leach out from the bricks during its usage. The leaching of heavy metals from the prepared bricks was assessed based on the Toxicity Characteristics Leaching Procedure (TCLP) and the Netherlands tank leaching test [9]. The procedure developed by the United States Environmental Protection Agency was followed to determine the leaching of heavy metals [20]. To conduct the test, brick specimens were broken and sieved through a 9.5 mm sieve. Particles finer than 9.5 mm sieve were used in the test. 0.57% v/v acetic acid was used as a leaching medium.

The solid (broken brick particles) liquid ratio was 1:20 when conducting the TCLP. The solid-liquid mixture was rotated at 30 ± 2 rpm for 18 hours, and the leachate obtained was filtered through a 0.45 µm filter paper.

Heavy metals (Cr, Ni, Pb, Zn, and As) in the filtered solution were analyzed using ICP MS [6]. The Netherlands tank leaching test was conducted as per NEN 7345 [10]. The Netherlands tank leaching test was followed in the European Union and the Netherlands to determine the leaching of metals from the building materials. In this method, a brick sample was subjected to leaching for eight extractions.

The sample was subjected to different contact periods in each extraction. In the first extraction, the brick specimen was placed in a polyethylene container and filled with the leaching solution. The leaching solution used in the test was acidified water, having a pH of 4. The volume of the leaching solution was five times that of the brick used in the test [11]. The brick sample was utterly submerged in the solution, and the brick surface was 5 cm from the leaching solution. The brick sample was submerged in the leaching solution for 0.25 days, and after the completion of the specified time, the brick sample was taken out. The solution was filtered through a 0.45-micron filter paper and analyzed for the heavy metals using ICP MS. Equation (1) was used to calculate the heavy metals leached in the first extraction.

$$E_i = \frac{(C_i - C_0) \cdot V}{1000A} \quad (1)$$

where E_i is the leachability of the heavy metal in the i^{th} extraction in mg/m², C_i is the concentration of a particular heavy metal in the i^{th} extraction in mg/L, C_0 is the concentration of the particular heavy metal in the blank in mg/L, V is the volume of the extracting solution in litres, and A is the surface area of the brick used in m². After the first extraction [12], the same brick sample was again placed in a polyethylene container and filled with a fresh leaching solution, as done in the first extraction. The sample was kept in submergence for the second extraction to have a cumulative contact period of one day. After the specified time, the leaching solution was removed and filtered using a 0.45-micron filter paper. The filtered solution was analyzed for the concentration of heavy metals using "(1)". The same brick sample was used for the subsequent six extractions using a fresh leaching solution for each extraction [13]. The extraction process followed in each extraction was the same in the previous extractions.

The contact period of brick with the leaching solution in the 3rd, 4th, 5th, 6th, 7th, and 8th extractions was 2.25, 4, 9, 16, 36, and 64 days respectively [14]. The contact period was the cumulative time period followed in each extraction [6]. Equation (2) was used to calculate the heavy metals leached at the end of eight extractions.

$$E = \sum_{i=1}^8 E_i \quad (2)$$

Table. 1 TCLP results of various heavy metals leached from TS bricks

Heavy metal	Firing temperature	0 wt% TS bricks	9 wt% TS bricks	18% TS bricks	27 wt% TS bricks	USEPA limits (mg/L)
Cr (mg/L)	1000°C	n.d	0.10	0.48	1.24	5
	950°C	n.d	0.34	0.90	1.78	
	900°C	n.d	0.54	1.35	2.10	
Ni (mg/L)	1000°C	n.d	0.05	0.11	0.20	11
	950°C	n.d	0.07	0.14	0.24	
	900°C	n.d	0.08	0.17	0.27	
Zn (mg/L)	1000°C	n.d	n.d	n.d	n.d	500
	950°C	n.d	n.d	n.d	n.d	
	900°C	n.d	n.d	n.d	n.d	
Pb (mg/L)	1000°C	n.d	0.02	0.09	0.17	5
	950°C	n.d	0.03	0.11	0.20	
	900°C	n.d	0.04	0.13	0.22	
n.d = Not detected						

Table. 2 Heavy metals leached from TS bricks as per the Netherlands tank leaching test

Heavy metal	Firing temperature	0 wt% TS bricks	9 wt% TS bricks	18% TS bricks	27 wt% TS bricks	Permissible limits set by NEN 7345 [25]
Cr (mg/m ²)	1000°C	n.d	0.018	0.027	0.039	150 for U ₁ and 950 for U ₂
	950°C	n.d	0.058	0.077	0.095	
	900°C	n.d	0.110	0.145	0.184	
Ni (mg/m ²)	1000°C	n.d	n.d	n.d	n.d	50 for U ₁ and 350 for U ₂
	950°C	n.d	0.001	0.002	0.003	
	900°C	n.d	0.002	0.004	0.007	
Zn (mg/m ²)	1000°C	n.d	n.d	n.d	n.d	200 for U ₁ and 1500 for U ₂
	950°C	n.d	n.d	n.d	n.d	
	900°C	n.d	n.d	n.d	n.d	
Pb (mg/m ²)	1000°C	n.d	0.003	0.004	0.006	100 for U ₁ and 800 for U ₂
	950°C	n.d	0.008	0.012	0.022	
	900°C	n.d	0.033	0.045	0.062	
n.d is not detected						
If the cumulative value of a particular heavy metal leached was less than U ₁ , the material can be used in constructions without restrictions. If the heavy metal leached was greater than U ₂ , the material is not permitted in constructions. If the results are in between U ₁ and U ₂ the material can be used for constructions and needs to be treated after the service life.						

3. Results and Discussions

Heavy metals leached from the brick were compared with the permissible limits of USEPA 1311 and NEN 7345. Heavy metals leached from brick as per TCLP are mentioned in Table 1 and NEN 7345 in Table 2. Permissible limits set by the USEPA for Cr, Ni, Zn, and Pb were 5 mg/L, 11 mg/L, 500 mg/L, and 5 mg/L, respectively [15]. 2.10 mg/L was the maximum amount of Cr leached from 27 wt% TS bricks fired to 900°C. 0.22 mg/L was the maximum amount of Pb leached from the bricks containing 27 wt% TS fired at 900°C. Zn was not detected

in any of the bricks containing TS. The maximum amount of Ni leached was 0.27 mg/L in 27 wt% TS bricks fired at 900°C. In both methods [16], the leaching of metals increased with an increase in the tannery sludge amounts and decreased with an increase in temperature. It was observed that all the metals (Cr, Ni, and Pb) leached were comparatively higher in the TCLP test than in the Netherlands tank leaching test [17]. In the TCLP test, the brick sample was crushed, and the particles less than 9.5 mm in size were used in the leach test, whereas in the Netherlands tank leaching test, the brick sample was used without crushing [18]. Due to crushing, the surface area of particles increases, hence increasing the chances of getting in contact with the leaching solution.

Due to crushing, heavy metals in the material's inner portions also come in contact with the leaching solution. In the TCLP test, the crushed pieces were in contact with the leaching solution for 18 hours, and were kept in agitation [19]. Due to agitation, each particle comes in contact with the leaching solution in all directions. In the Netherlands tank leaching test, the uncrushed sample was in contact with the leaching solution for a cumulative period of 64 days. In the Netherlands tank leaching test, as the sample was uncrushed, heavy metals on the surface of the brick could be leached out quickly compared to the heavy metals in the interior portion of the brick. The leaching of heavy metals from the inner portions depends on the diffusion of the leaching solution into the inner portion of the brick [20]. The leaching of metals in the Netherlands tank leaching test also depends on the distribution of heavy metals in various brick portions. As the bottom of the brick touches the inner surface of the container, the chance of leaching solution getting in contact with the bottom surface reduces. Also, the material was not in agitation during the leaching period. The rate of release of metals at different periods can be measured from the Netherlands tank leaching test.

In the TCLP test, as the crushed particles were used, the distribution of heavy metals in the brick did not effect on the leaching of heavy metals. Moreover, the particles were agitated during the TCLP test, which increased the chances of contact with the leaching solution at all the surface portions of the particles. Because of all these reasons, the leaching of metals from the TCLP test might be comparatively high as that of the Netherlands tank leaching test. The results obtained from the TCLP test are more appropriate to the actual field conditions arising in the service life of materials. Hence, the TCLP test results could be more reliable than the Netherlands tank leaching test.

4. Conclusions

The following were the conclusions drawn from the results of the TCLP and Netherlands tank leaching tests:

- (a) The leaching of metals was comparatively high in the TCLP test compared to that of the Netherlands tank leaching test.
- (b) The crushing of particles increases the surface area of particles, resulting in increased chances of getting in contact with the leaching solution.
- (c) TCLP test results do not depend on the distribution of metals in the inner portions of the brick, whereas the Netherlands tank leaching test has a significant impact on the distribution of metals in the brick.
- (d) The crushing of particles has a more significant impact on the leaching of metals than the contact period with the leaching solution.
- (e) TCLP test results are more appropriate to the actual

field conditions arising during the service life of materials.

References

- [1]. B. Dhal, H.N. Thatoi, N.N. Das, and B.D. Pandey, "Chemical and microbial remediation of hexavalent chromium from contaminated soil and mining/metallurgical soil waste: a review," *Journal of Hazardous Materials*, vol. 250–251, pp. 272–291, 2013
- [2]. P. Kavouras, E. Pantazopoulou, S. Varitis, G. Vourlias, K. Chrissafis, and G.P. Dimitrakopoulos, "Incineration of tannery sludge under oxic and anoxic conditions: study of chromium speciation," *Journal of Hazardous Materials*, vol. 283, pp. 672–679, 2015
- [3]. G.L. Abdul, I. Azni, A.S. Abdul, H.K.W Calvin, S.J. Mohd, and M.B. Aminuddin, "Reusability of sewage sludge in clay bricks," *Journal of Material Cycles and Waste Management*, vol. 6(1), pp. 41–47, 2004
- [4]. M.A. Abreu and S.M. Toffoli, "Characterization of a chromium-rich tannery waste and its potential use in ceramics," *Ceramics International*, vol. 35(6), pp. 2225–2234, 2009
- [5]. S.K. Amin, E.M. Abdel Hamid, S.A. El-Sherbiny, H.A. Sibak, and M.F. Abadir, "The use of sewage sludge in the production of ceramic floor tiles," *Housing and Building National Research Center*, vol. 14(3), pp. 309–315, 2018
- [6]. M. Ariful Islam Juel, A. Mizan, and T. Ahmed, "Sustainable use of tannery sludge in brick manufacturing in Bangladesh," *Waste Management (New York, N.Y.)*, Vol. 60, pp. 259–269, 2017.
- [7]. T. Basegio, F. Berutti, A. Bernardes, and C.P. Bergmann, "Environmental and technical aspects of the utilization of tannery sludge as a raw material for clay products," *Journal of the European Ceramic Society*, vol. 22(13), pp. 2251–2259, 2022.
- [8]. S.S.B. Begum, R. Gandhimathi, S.T. Ramesh, and P.V. Nidheesh, "Utilization of textile effluent wastewater treatment plant sludge as brick material," *Journal of Material Cycles and Waste Management*, vol. 15, pp. 564–570, 2013.
- [9]. K.Y. Chiang, P.H. Chou, C.R. Hua, K.L. Chien, and C. Cheeseman "Light weight bricks manufactured from water treatment sludge and rice husks," *Journal of Hazardous Materials*, vol. 171(1-3), pp. 76–82, 2009.
- [10]. J.A. Cusido and L.V. Cremades, "Environmental effects of using clay bricks produced with sewage sludge: leachability and toxicity studies," *Waste Management (New York, N.Y.)*, vol. 32(6), pp. 1202–1208, 2012.
- [11]. G. Goel and A.S. Kalamdhad, "An investigation on use of paper mill sludge in brick manufacturing,"



Construction and Building Materials, vol. 148, pp. 334–343, 2017.

- [12].K.M. Hassan, K. Fukushi, K. Turikuzzaman, and S.M. Moniruzzaman, "Effects of using arsenic-iron sludge wastes in brick making," Waste Management (New York, N.Y.), vol. 34(6), pp. 1072–1078, 2014.
- [13].A.G. Liew, A. Idris, C.H. Wong, A.A. Samad, M.J. Noor, and A.M. Baki, "Incorporation of sewage sludge in clay brick and its characterization," Waste Management and Research, vol. 22(4), pp. 226–233, 2004.
- [14].D.F. Lin and C.H. Weng, "Use of sewage sludge ash as brick material," Journal of Environmental Engineering, vol. 127(10), pp. 922–927, 2001.
- [15].L. Mao, Y. Wu, W. Zhang, L. Hu, and Q. Huang, "Effects of electroplating sludge introduction on the morphology, mineral phase and porosity evolution of fired clay bricks," Construction and Building Materials, vol. 211, pp. 130–138, 2019.
- [16].S.N. Monteiro, J. Alexandre, J.I. Margem, R. Sanchez, and C.M.F. Vieira, "Incorporation of sludge waste from water treatment plant into red ceramic," Construction and Building Materials, vol. 22(6), pp. 1281–1287, 2008.
- [17].S.E. Chidiac and L.M. Federico, "Effects of waste glass additions on the properties and durability of fired clay bricks," Canadian Journal of Civil Engineering, vol. 34(11), pp. 1458–1466, 2007.
- [18].S. Kazmi, M.S.S. Abbas, M.L. Nehdi, M.A. Saleem, and M.J. Munir, "Feasibility of using waste glass sludge in production of eco-friendly clay bricks," Journal of Materials in Civil Engineering, vol. 29(8), pp. 1–12, 2017.
- [19].N. Phonphuak, S. Kanyakam, and P. Chindaprasirt, "Utilization of waste glass to enhance physical mechanical properties of fired clay bricks," Journal of Cleaner Production, vol. 112 (Part 4), pp. 3057–3062, 2015.
- [20].USEPA. 1992. Method 1311 toxicity characteristic leaching procedure (TCLP). US Environmental Protection Agency.

How To Cite:

G Venkatesh , S Krishnaiah , " Comparative Study of Heavy Metal Leaching from Clay – Tannery Sludge – Glass Powder Bricks using TCLP and Netherlands Tank Leaching Methods " , International Journal of Computational Sciences and Engineering Research (IJCSEr) , Volume 2, Issue 4 , pp 29 - 33 , November 2025
CrossRef DOI: <https://doi.org/10.63328/ijcser-v2ri4p6>

Declaration

Conflicts of Interest: The authors declare no conflict of interest.

Author Contribution: All authors wrote the main manuscript text and also consent to the submission.

Ethical approval: Not applicable.

Consent to Participate: All authors consent to participate.

Funding: Not applicable, and No funding was received

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Personal Statement: We declare with our best of knowledge that this research work is purely Original Work and No third party material used in this article drafting. If any such kind material found in further online publication, we are responsible only for any judicial and copyright issues.

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Acknowledgements

We thank everyone who inspired our work.