Lead Migration Tendancy in Composite Materials with E-Waste Glass Embedded

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This study analyzed the lead behavior from CRT glass embedded in original composite materials exposed to different pH and temperature conditions. For this study were obtained composite materials with CRT glass (funnel) waste, some of them with zeolite addition, for lead chelating. The experimental results showed the pH and temperature influence lead leachability during the ultrasonic procedure and explained the influence of zeolites in lead behavior.

Keywords: composite materials, CRT waste, lead, zeolite

In the concept of sustainable development, waste represents a secondary raw material. An important waste stream in waste management is represented by Waste Electrical and Electronic Equipment (WEEE) or e-waste. The legislative targets for WEEE recycling/reuse are high and the landfilling is restricted. Therefore, recycling methods are studied and applied in, so the environmental legislative targets are achieved. A modern e-waste recycling method can be based on WEEE incorporation in composite materials. Composite materials have to response concomitant to technical and environmental requirements. The waste CRT glass is classified as hazardous waste, due to its high lead content. Composite materials with hazardous waste content aren't framed in any actual construction standard. To propose such material for construction use, the environmental and human safety must to be certified.

The aim of this paper was to study the behavior of lead leachability from CRT embedded in composite material, at different *p*H and temperature levels, using lead chelating materials (zeolite).

The priorities of the EU Directive no. 65/2011 are the restriction of the amount of hazardous substances in EEE (Electric and Electronic Equipment) and the recycling of , but these substances are still used under the exact limits [1]. Therefore, at the end of life, the electronic equipment became hazardous waste, so the disposal of untreated WEEE separately collected is prohibited [2]. Each Member State has to achieve EU targets regarding the collection and recycling of WEEE [3]. Once the target achieved, a major challenge occurs: recycling the waste CRT glass from dismantling WEEE, with high volume and high recycling cost [4].

In Romania, in 2013, total WEEE collected amount was 23,246.31 tons, from which 3,848.11 tons were IT and telecommunications equipment containing CRT glass (National Environmental Protection Agency statistic data [5]). Considering that in Romania the collected equipment contain high amount of CRT (54%) [6], it follows that in 2013 the collected CRT waste was around 2,077.98 tons.

Romanian WEEE recyclers haven't yet regional or local solutions for recovery of waste CRT glass, therefore the waste is stored temporarily to identify environmentally and economically viable recycling solutions. Only a small proportion of waste CRT glass (front panel or cone) is accepted in the European market as a raw material for new glass products. Manufacturers of objects / glass products, in most cases, do not accept this type of waste as raw material, due to its chemical heterogeneity [7]. The major problem of CRT waste glass is the presence of high amount of lead [8, 9].

Knowing that WEEE are classified as hazardous waste (code 16 02 15*) [2] and their disposal is an environmental problem, the authors performed several studies to find an environmental friendly recycling method for waste CRT glass [2, 9-11].

Thus a new original composite material obtained by embedding waste CRT glass into a cement matrix was proposed as construction material [7]. Similar studies made with different amount of CRT came to support the present proposal [12, 13] and also good environmental results were obtained in author's previous studies on original composite material [14].

Furthermore, the lead leachability studies revealed that, in normal conditions the composite is safe, the lead behavior being improved by addition of zeolites [2, 7].

Experimental part

To understand the lead leaching decreasing tendency in composites with zeolites content [2], the correlation between the lead concentration, pH and temperature during the sonication process was studied. The composite samples were obtained from crushed CRT funnel glass waste (size 2.5 mm) white cement CEM II / AL 52.5 N [15] and zeolite (fig. 1). The used zeolite ratio (10%) was the

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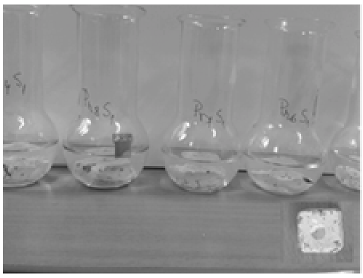

Fig. 1. Composite material samples. Detail: sample

Fig. 2. Ultrasonic tests

Table 1
COMPOSITE MATERIAL
- SET SAMPLES

Funnel Glass diameter 1-3 mm				Funnel Glass diameter 7-8 mm			
SET 1	25%	**SET 2**	50%	**SET 3**	25%	**SET 4**	50%
SET 1Z	25 % + **10% zeolite**	**SET 2Z**	50% + **10% zeolite**	**SET 3Z**	25% + **10% zeolite**	**SET 4Z**	50% + **10% zeolite**

Table 2
*p*H VARIATION OF THE SOLUTIONS, AFTER ULTRASONIC TEST (30 min)

The solution pH prior to samples introduction	*The leachate pH after 30 minutes of sonication*			
	Set 1	Set 2	Set 3	Set 4
3	7.0	6.9	8.3	8.2
5.5	7.1	7	8.4	8.1
9	10	10.1	10.2	10.2
12	10.2	10.1	10.3	10.3

Table 3
*p*H VARIATION OF THE SOLUTIONS, AFTER 30 MINUTES OF ULTRASONIC TEST - ZEOLITE SAMPLES

The solution pH prior to samples introduction	*The leachate pH after 30 min of sonication*			
	Set 1Z	Set 2Z	Set 3Z	Set 4Z
3	9.5	11.2	11.3	11.1
5.5	10.2	10.3	10.4	10.5
9	10.5	10.6	10.3	10.4
12	8.1	8.1	8.1	7.9

same as the used one in heavy metal water purification processes [16, 18-21, 23, 34].

For this study, were made eight composite material set samples (3 samples/set), as can be seen in table 1.

The eight sets were submitted to ultrasonic bath, model S 10 H Elmasonic (fig. 2), at four pH values (3; 5.5; 9; 12), at the same period of time (30 min) and room temperature. Each value of *p*H was adjusted by titration with NaOH or HNO_3. The alkalinity of the cement had a high influence on pH; lead migration analysis at a certain pH requiring a constant adjustment of it [9]; therefore the ultrasonic tests were performed instantly after the samples introduction into the leaching solution (fig. 2).

The analysis of the total lead from leachate was performed by using the Atomic Absorption Spectrometry by flame method and graphite tube method.

Results and discussions

pH changing during sonication at room temperature

To can observe the *p*H changes during the sonication tests (30 min), the *p*H solutions values were measured before and after the test time (table 2 and table 3).

From the results listed in table 2 it can be seen a change in all samples *p*H, after sonication. To be noted that if the initial acid solution after ultrasonic test becomes neutral, in case of the initial basic solution (*p*H = 12), the *p*H decreases.

From the results shown in table 3, a significant change was registered in all *p*H samples with the zeolite content, after sonication.

It is noted that if the initial acid solution, becomes very alkaline after sonication, the initial basic solution (*p*H = 12) becomes almost neutral (*p*H ~ 8). Perhaps this explains the differences between lead concentrations recorded for samples without zeolite content comparative with samples containing zeolite (fig. 3).

The figure 3 represents the comparison between lead concentration (mg/kg) lechability from samples without zeolite (1 to 4) and with zeolite content (1Z to 4Z) at different *p*H levels.

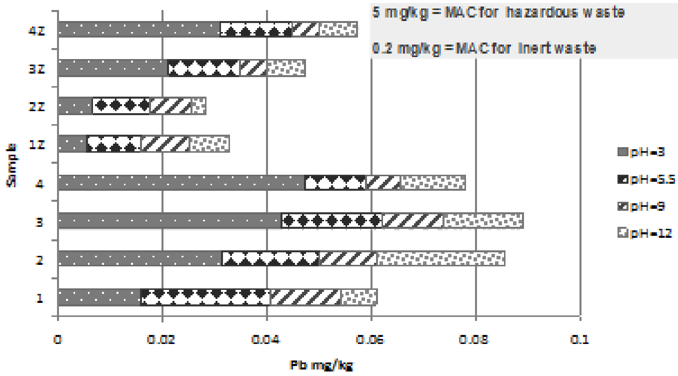

Fig. 3. Comparison between lead concentration (mg/kg) lechability from samples without zeolite (1 to 4) and with zeolite content (1Z to 4Z) at different *p*H levels

pH modification during sonication at different temperatures

Besides *p*H, temperature plays an important role in heavy metals migration.

A similar experiment was performed on **Set 2**, but it was submitted to sonication for 30 min at 5 different temperatures: 10, 20, 30, 40 and 60 °C. This time was started out with *p*H = 3. The samples from Set 2 were immersed in a solution with *p*H = 3 because the more intense lead migration have been observed at this *p*H value. The reason for choosing the Set 2 for further investigation was given by the sum of the favorable results of this composition, especially at *p*H - 5.5, close to natural conditions [7, 9, 10]. The results of the experiment are presented in table 4.

From table 4 it can be seen that solution pH reach neutral values by sonication, only for sample analyzed at room temperature (20°C). The values for all other temperatures, suffered significant changes, the solution became very basic.

Influence of temperature on lead leaching

In order to follow the effect of temperature on lead leaching, Set 2 was subjected to sonication for 30 min at 5 different temperatures: 10, 20, 30, 40 and 60°C. The temperature influence on lead migration is shown in figure

The solution pH prior to samples introduction	The leachate pH after 30 minutes sonication				
	10 (°C)	20 (°C)	30 (°C)	40 (°C)	60 (°C)
3	10.5	7.1	11.3	11.6	10.9

Table 4
*p*H VARIATION OF THE SOLUTIONS AFTER 30 MINUTES OF ULTRASONIC TEST, AT DIFFERENT TEMPERATURES

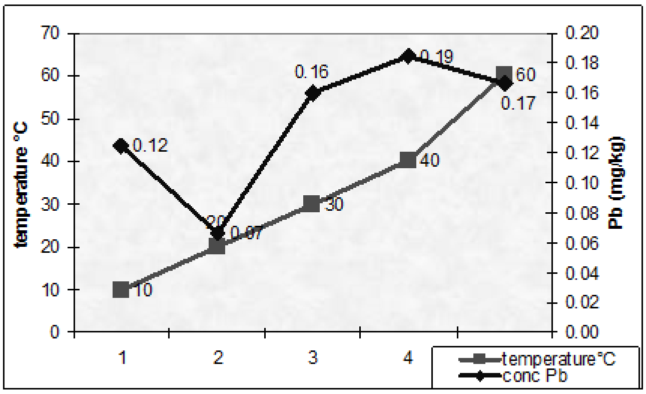

Fig.4 The correlation between lead concentration and temperature, at constant *p*H, during sonication

4. The lead concentration of the lixiviation solution after the ultrasonic test was determined by flame atomic absorption using a spectrophotometer ZEEnit 700 (Analytic Jena).

Figure 4 shows that at room temperature, lead concentration values are lowest; but also at other temperatures, lead concentration values are not high, doesn't exceeded MAC (maximum allowed concentration) for inert waste (0.2 mg / kg) [22]. By increasing the temperature above 40°C, the lead migration decreases.

Conclusions

The addition of zeolite in composite material structure improves the lead behavior.

The results provided valuable information on the lead leachability / mobilization from waste CRT glass. The zeolite addition in the composite material structure improves the lead behavior.

The *p*H and temperature variations explain the differences between lead concentrations recorded for samples without zeolite content comparative with samples with zeolite content.

Further studies for materials containing zeolites are needed; both in terms of composition and mechanical resistance, coupled with economic and technical feasibility studies.

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