# The Induced Impact on the Environment by Combined Plastic and Paper Waste before and after Recovery

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Combined plastic and paper waste (paper envelopes with plastic window, large envelopes made from paper with plastic air bubble, etc.) are not recyclable by current technologies. The storage is the only viable alternative to disposal, but in this case a series of issues arise by the release of pollutant compounds into the leachate. It has been studied the possibility of using this non-recyclable waste by including it in the structure of composite materials that have applicability in the field of constructions. Current paper presents the results obtained in the analysis of leachates from combined plastic and paper waste before being included in the matrices of composite materials and after applying this recovery process. The comparative analysis of the results highlights the environmental impact of each category of waste.

Keywords: combined waste of plastic and paper, composite materials, leaching, environmental impact

The waste, depending on their nature, contributes during the storage process to the pollution of all environmental components: air, soil and water. Their recycling through a succession of operations has as main objective the reduction of the pollution-induced on the environment [1-3]. At the same time, the recycling aims to obtain new raw materials and reaching the demands of the circular economy [4-5]. In the last period, the major development of plastics and paper industries involved in making different types of packaging have triggered a special attention to their recycling, as soon as they become waste [6]. In the particular case of solid plastic waste, their recycling is done according to principles of the circular economy through procedures classified by the type of product manufactured from secondary raw materials. Thus, two major categories of processes are known: closed-loop recycling and open-loop recycling. In the first case, the recycled plastic waste is transformed, alone [7] or in mixture with new plastic material [8-9], into a product similar to the one from which it was recovered. It is not recommended to use mixtures of plastic waste for the application of the closed-loop process [10]. In the second case, the recycled waste is used in the manufacture of a product different from the one from which they were recycled [11]. In the case of a paper waste, its recovery has become an essential component in the production of cellulose. Currently, the recycled paper is an essential raw material for the paper industry [12]. Paper recycling reduces the impact on the environment by increasing paper life cycle together with increasing recycling rates [13]. The concerns about environmental pollution remains due to hazardous substances present in certain categories of waste paper packaging, which cannot be recycled, that could be released during waste paper storage [14]. In order to show the impact of these wastes on the environment, the ecological risk [15] and the degree of hazardous substances [16] migration in the deposit leachate must be analyzed by carrying out similar research studies to those conducted for other types of wastes such as fly ash [17].

There are several distinct categories of combined plastic and paper waste that cannot be recycled by current technologies. The most frequently encountered are those generated from the activities of postal messaging such as paper envelopes with plastic window or packages with an inner surface of anti-shock plastic protection. Other non-recyclable waste came from foodstuffs packaging with Tetra Pak® type structure and single-use cups and plates made of paper with added plastic films or foils. The recycling of these wastes by current technologies involves high labor costs, high energy consumption and generation of new wastes that must be disposed off through storage. The recycling is difficult because in the first step the plastic components must be separated from the paper ones by mechanical methods, applied manually. Sometimes, packages that must provide a barrier to water, water vapor, oils and fats or to light radiation [18] are combined plastic and paper material made by depositing on the surface of the paper film-forming polymers [19], polyethylene, polypropylene, polyethylene terephthalate films [20] or hydrophobicizing resins [21]. The separation of the two components (plastic and paper) for recycling is practically an impossible operation for such waste materials. Based on these aspects, technological

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solutions for the recovery/recycling of these kind of wastes have been investigated by incorporating them into new composite materials that can be used in the field of construction. The technological phases include the collection of the combined plastic and paper waste, sorting and drying along with disinfection by microwave treatment [22] followed by chopping/grinding, mixing with new base polymer and extrusion into new materials with mechanical characteristics specific to the construction materials. [23]. In this context, the purpose of this paper is to highlight the reduction of plastic and paper combined waste impact on the environment by their inclusion in composite materials intended for the field of constructions.

## **Experimental part**

## Materials and methods

Three types of mixed plastic and non-recyclable paper waste and 6 types of materials were included in the study. Sample codes were as follows: P1 - paper envelope with plastic inside; P2 - paper envelope with plastic applied externally; P3 - blend of paper envelopes with plastic window; P4 - polypropylene grinding (PP) with 5% combined plastic and paper waste (CW) ; P5 - high density polyethylene (HDPE) grinder with 5% CW; P6 - composite material obtained by injection of HDPE and 5% CW; P7 - composite material obtained by injection of HDPE blend and 15% CW; P8 - composite material obtained by injection of PP mixture and 5% CW; P9 - composite material obtained by injection of PP mixture and 15% CW. The samples evaluated in this study are presented in Figure 1.

The waste material analyzed in the study was subjected to the leaching process following the procedure described in SR EN 12457-2: 2003 [24]. The leachates obtained were characterized by determining the values of all the indicators specified in Order no. 95/12.02.2005 regarding the establishment of the criteria for the acceptance and preliminary acceptance procedures for waste at storage and national list of waste accepted in each class of waste landfill [25]. The metal content was determined using an inductively coupled plasma mass spectrometer ICP-MS 7900 Agilent Technologies, (Japan), sulphate and chloride anions values were determined using a DIONEX ICS-3000 ion chromatograph (U.S.A.) with AG23 Dionex column and suppressed conductivity detection. A nitrogen/carbon Analyzer N/C 3100, Analytik Jena, (Germany) was used to determine the dissolved organic carbon (DOC) indicator values. The values of the Total Dissolved Solids (TDS) indicator were determined by standardized analytical method.



Fig. 1. The waste materials taken in the study, P1-P9

## **Results and discussions**

The results obtained regarding the content of heavy metals specified in Order no. 95 / 12.02.2005, were presented in Table 1.

It was found that the Zn indicator value from samples P4 and P5 exceeded the limit value imposed for a waste acceptance on non-hazardous waste deposits. However, both measured values for the Zn indicator, 57.769 mg / kg D.M. and 109.21 mg / kg D.M., respectively, were lower than the limit value imposed by Order no. 95 / 12.02.2005 for the acceptance of waste on hazardous waste deposits, 200 mg / kg D.M. The explanation for the high content of Zn in P4 and P5 samples, grinded polymer mixed with the combined plastic and paper waste, was consisted with the fact that a component with high content of Zn has been also involved in the waste collection process. This showed that a special attention must be given during the process of collecting and selecting the combined waste of plastic and paper. However, this metal (Zn) was blocked (insoluble) in the process of transformation into composite material by injection, an aspect demonstrated by the low content (below the value imposed for the storage of waste on non-hazardous waste deposits) in the leachate of the corresponding samples (P8 and P6 respectively). There were no exceedances of the limit value imposed for the storage of waste on non-hazardous waste deposits for the other analyzed metals.

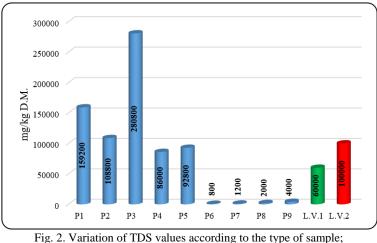
Metal	Sample type / content									
	P1	P2	P3	P4	P5	P6	P7	P8	P9	L.V.*
As	0.0025	0.0076	0.0026	0.4012	0.1693	0.0014	0.0013	0.0007	0.0027	2
Ba	0.3493	0.6586	0.5371	2.2041	2.6454	0.6722	1.2467	1.2701	1.1828	100
Cd	0.0027	0.0006	0.0021	0.0191	0.0197	0.0002	0.0001	0.0001	0.0001	1
Cr	0.0278	0.0308	0.0247	0.0918	0.1289	0.0424	0.0497	0.0343	0.0375	10
Cu	0.0935	0.1263	0.1446	0.1042	0.2731	0.0241	0.0249	0.0236	0.0338	50
Hg	0.0171	0.0113	0.0087	0.0158	0.0187	0.0037	0.0031	0.0025	0.0017	0,2
Мо	0.0176	0.1757	0.6827	0.1313	0.0388	0.0013	< 0.001	< 0.001	< 0.001	10
Ni	0.0381	0.0237	0.0411	0.2141	0.2138	0.0151	0.0089	0.0096	0.0095	10
Pb	0.0164	0.0056	0.0055	0.0079	0.0237	0.0031	0.0021	0.0016	0.0019	10
Sb	0.0077	0.0122	0.0019	0.0659	0.0592	0.0021	0.0009	0.0007	0.0027	0,7
Se	0.0034	0.0027	0.0024	0.0022	0.0033	0.0004	0.0013	0.0016	0.0001	0.5
Zn	0.2318	0.2594	0.2091	57.769	109.21	0.6391	0.3452	0.2001	0.7438	50

 Table 1

 VALUES OF METALS CONTENT IN LEACHATE, MG/KG D.M.

\* L.V. = Limit value for accepting waste in non - hazardous waste deposits.

The obtained results regarding the values of the TDS indicator are shown in Figure 2.



L.V.1 TDS - limit value for accepting waste in non-hazardous waste deposits and
 L.V.2 TDS - limit value for accepting waste in hazardous waste deposits

Their analysis showed that for samples P4 and P5 there were values higher than the limit value imposed for accepting waste on non-hazardous waste deposits, but without exceeding the limit value imposed for acceptance on hazardous waste deposits. The situation was more worrying from the point of view of TDS indicator, in the case of samples P1, P2 and P3, for which TDS values exceed the limit value imposed for the acceptance on hazardous waste deposits. The wastes corresponding to the mentioned samples induced a major negative impact on the environment during the storage period. According to the legislation in force, these wastes should not be accepted for storage on any type of deposit. Samples P1, P2 and P3, combined waste of plastic and paper, cannot be recycled by any conventional technologies. Samples P4 and P5 were milled mixtures in which parts of P1, P2 or P3 type waste were also found. Through the mixing and transformation operation in the mill the impact on the environment was moderate. The TDS values for leachate samples P6 - P9 were 15 to 75 folds lower than the limit value for accepting waste on non-hazardous waste deposits. This demonstrated that the transformation of raw materials of type P4 and P5 by injection and extrusion into finished products (composite materials intended for the field of constructions of type P6 - P9) with an insignificant impact on the environment.

The results obtained regarding the DOC indicator values were shown in Figure 3.

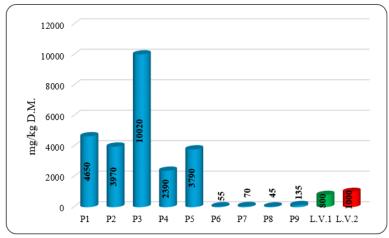


Fig. 3. Variation of DOC values according to the type of sample \*L.V.1 DOC - limit value for accepting waste in non-hazardous waste deposits (mg/kg d.m.) \*L.V.2 DOC - limit value for accepting waste in hazardous waste deposits (mg/kg d.m.)

The values of leachate samples P1 - P5 were between 2.39 and 10 folds greater than the imposed limit for the storage of hazardous waste deposits acceptance. This was due to the release of organic compounds (adhesives, dyes, waterproofing compounds, etc.), used to obtain the combined materials of plastic and paper, which later turned into waste. The results showed that from the point of view of the DOC indicator, samples P1 - P5 generated a major negative impact on the environment. The effect on the environment become insignificant after the inclusion of the combined waste of plastic and paper in matrices of composite materials (samples P6 - P9). The DOC values for these samples were from 5.9 - 17.8 folds lower than the limit value for accepting waste on non-hazardous waste deposits.

Regarding the values of the chloride and sulfate indicators, the values obtained for all samples were much lower than the limit values imposed for accepting waste on non-hazardous waste deposits, as was shown in Figure 4.

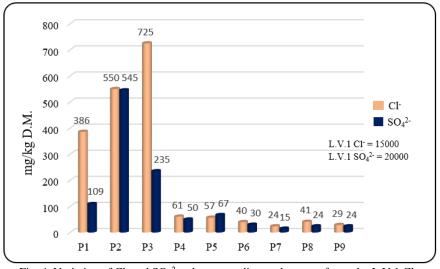


Fig. 4. Variation of Cl<sup>-</sup> and SO4<sup>2-</sup> values according to the type of sample; L.V.1 Cl<sup>-</sup>, SO4<sup>2</sup> - limit value, mg/kg D.M., for accepting waste in non-hazardous waste deposits

Thus, in the case of the chloride ion, the highest value recorded (in the case of sample P3) was 20 folds lower than the limit value and for the sulphate ion, the highest value recorded was 36.7 folds lower than the limit value. Practically, from the point of view of the two indicators, the induced impact on the environment in the storage process was insignificant. It should be noted that the inclusion of the combined waste of plastic and paper in composite materials by injection / extrusion had an effect of reducing the chloride ion values by at least 9.6 folds and the sulphate ion by 3.6 folds (P6 vs. P1).

#### Conclusions

The study revealed that in the process of disposal through storage, the combined plastic and paper waste (samples P1, P2 and P3), which cannot be recycled by current technologies, induced a major negative impact on the environmental components. This aspect was the consequence of the leachate composition generated during storage. Leachate had a high

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organic load evidenced by TDS indicators and DOC values that exceed the limits imposed by Order no. 95 / 12.02.2005 for the acceptance of waste on hazardous waste deposits.

The application of a new technology to capitalize on these wastes by transforming them into composite materials intended for the field of constructions (samples P6 - P9) reduced the impact on the environment down to the level of "insignificant" where TDS decreased 15 to 75 folds compared to the limit value imposed for accepting waste on non-hazardous waste deposits and DOC values decreased 5.9 to 17.8 folds compared to the limit value imposed for accepting waste on non-hazardous waste deposits. It should be emphasized that in order to be transformed into composite materials, the combined waste of plastic and paper were initially transformed from the raw material to the powder form by grinding, which was a mixture of a basic polymer (PP or HDPE) and a certain proportion of the waste itself. The analysis of these raw materials (samples P4 and P5) indicated that if the issues of their storage were raised, a moderate environmental impact from the point of view of the TDS indicator and a major one from the point of view of the DOC indicator would be revealed. Overall, a total transformation of raw materials from samples P4 and P5 into composite materials by a new technology, as opposite to store them as waste, decrease of the environmental impact from the major to the insignificant.

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#### References

1. BUTNARIU, I., CONSTANTIN, N., DOBRESCU, C., HEPUT, T., Rev. Chim. (Bucharest), 69, no. 5, 2018, p. 1066.

2. BURDUHOS NERGIS, D.D., VIZUREANU, P., CORBU, O., Rev. Chim. (Bucharest), 70, no. 4, 2019, p. 1262.

3. DULDNER, M.M., BARTHA, E., CAPITANU, S., NICA, S., COMAN, A.E., TINCU, R., SARBU, A., FILIP, P.I., APOSTOL, S., GAREA, S., Rev. Chim. (Bucharest), **70**, no. 7, 2019, p. 2301.

- 4. \*\*\*European Commision, A European Strategy for Plastics in a Circular Economy, 16.1.2018, p.14.
- 5. IZVERCIAN, M., IVASCU, L., Procedia Econ., 26, 2015, p.717.
- 6. da CRUZ, N.F., FERREIRA, S., CABRAL, M., SIMÕES, P., MARQUES, R.C., Waste Manag., 34, no. 2, 2014, p. 298.

7. KUMAR, S., PANDA, A., SINGH, R., Resour. Conserv. Recy., 55, no. 11, 2011, p.893.

- 8. RAGAERT, K., DELVA, L., VAN GEEM, K., Waste Manag. 69, 2017, p.24.
- 9. SINGH, N., HUI, D., SINGH, R., AHUJA, I.P.S., FEO, L., FRATERNALI, F., Compos. Part B Eng., 115, 2017, p. 409.
- 10. YU, J., SUN, L., MA, C., QIAO, Y., YAO, Waste Manag. 48, 2016, p. 300.
- 11. AZNAR, M., CABALLERO, M., SANCHO, J., FRANCS, E., Fuel Process. Technol., 87, 2006, p. 409.

12. PIVNENKO, K., ERIKSSON, E., ASTRUP, T., Waste Manag., 45, 2015, p. 134.

13. LAURIJSSEN, J., MARSIDI, M., WESTENBROEK, A., WORREL, E., FAAIJ, A., Resour. Conserv. Recy., 54, no. 12, 2010, p.1208.

14. LORENZINI, R., BIEDERMAN, M., GROB, K., GARBINI, D., BARBANERA, M., BRASCHI, I., Food Addit. Contam. A., **30**, no. 4, 2013, p.760.

15. KIM, L., ARAMA, G.M., Environ. Eng. Manag. J., 17, no. 9, 2018, p. 2201.

16. KIM, L., ARAMA, G.M., CUCIUREANU, A., GUTA, D., Environ. Eng. Manag. J., 17, no. 12, 2018, p. 2945.

17. KIM, L., CATRINA, G.A., STANESCU, B., PASCU, L.F., GHEORGHITA, T., MANOLACHE, D., Rev. Chim. (Bucharest), 70, no. 1, 2019, p. 269.

18. BAJPAI, P., Pulp and Paper Industry: Chemicals, Elsevier, Amsterdam, Netherlands, 2016, p.145.

19. HAGIOPOL, C., JOHNSTON, J.W., Chemistry of Modern Papermaking, CRC Press, edited by Hagiopol C. and Johnston J.W., Boca Raton, Florida, USA, 2012, p. 267.

20.ROBERTSON, G.L., Food Packaging: Principles and Practice, 3<sup>rd</sup> Edition, CRC Press, edited by Robertson G.L., Boca Raton, Florida, USA, 2012, p. 178.

21.BIERMANN, C.J., Handbook of Pulping and Papermaking, Academic Press, edited by Biermann C.J., San Diego, California, USA, 1996, p.421.

22. CIOBANU, R.C., URSAN, G.A., ARADOAEI, M., BATRINESCU, G., BANCIU, A.R., 2018 International Conference and Exposition on Electrical and Power Engineering, Proceedings, 2018, p. 1000.

23. CIOBANU, R.C., BATRINESCU, G., URSAN, G.A., CARAMITU, A.R., MARINESCU, V., BORS, A.M., LINGVAY, I., Mater. Plast., 56, no. 3, 2019, p. 475.

24. \*\*\*SR EN 12457:2 – 2003 – Waste characterization. Leaching. Conformity check for leaching granular waste and sludge. Part 2: One stage batch test at a solid – liquid ratio of 10l/kg for materials with a particle size of less than 4 mm (without or with a reduction in size). Romanian Standard Association, 30p.

25. \*\*\*ORDER 95/2005, The criteria for the acceptance and preliminary acceptance procedures for waste at storage and national list of waste accepted in each class of waste landfill, Official Monitor of Romania, Part 1, No. 194/8.

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