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Selective removal of heavy metals from landfill leachate by reactive granular filters

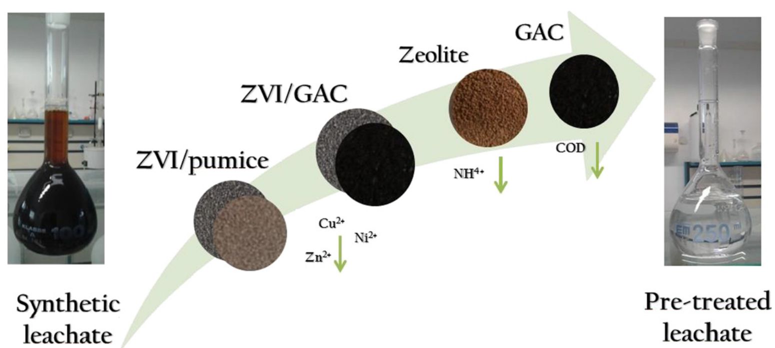
Bilardi Stefania, Paolo Salvatore Calabrò *, Greco Rosa, Moraci Nicola

Department of Civil, Energy, Environment and Materials Engineering (DICEAM), Mediterranea University of Reggio Calabria, Reggio Calabria, Italy

HIGHLIGHTS

- A new chemical-physical pre-treatment for leachate was presented
- ZVI, GAC and a zeolite efficiently remove heavy metals contained into the leachate
- The proposed method could be used as an on-site technology for leachate pre-treatment
- Pre-treatment allows to safely co-treat leachate in municipal WWTPs

GRAPHICAL ABSTRACT



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ABSTRACT

The pre-treatment of landfill leachate prior to its co-treatment in the municipal plants of waste water processing could represent an appropriate and cost-effective solution for its management. Pre-treatment is necessary especially to remove heavy metals, which may be transferred to the excess sludge preventing its valorisation. In the present paper, we propose a chemical-physical pre-treatment of leachate using four different granular reactive media able to selectively remove the contaminants present in the leachate. The efficiency of these materials was investigated using synthetic leachate through batch tests and a column test. In the latter case the four materials were placed in two columns connected in series and fed an under constant upward flow (0.5 mL/min). The first column was filled half (50 cm) with a granular mixture of zero valent iron (ZVI) and pumice and half (50 cm) with a granular mixture of ZVI and granular activated carbon (GAC). The second column, which was fed with the effluent of the first column, was filled half with zeolite (chabazite) and half with GAC. Heavy metals were mainly removed by the ZVI/pumice and ZVI/GAC steps with a removal efficiency that was higher than 98, 94 and 90% for copper, nickel and zinc, respectively, after 70 days of operation. Ammonium was removed by zeolite with a removal efficiency of 99% up to 23 days. The average reduction of the chemical oxygen demand (COD) was of 40% for 85 days, whereas chloride and sulphate removal was negligible.

1. Introduction

One of the most important issues for the overall sustainability (economic and environmental) of a modern landfill is leachate management; in fact, leachate is a complex and highly polluted matrix containing a large amount of dissolved organic matter, which is

* Corresponding author.
E-mail addresses: stefania.bilardi@unirc.it, (B. Stefania), paolo.calabro@unirc.it, (P.S. Calabrò), nicola.moraci@unirc.it (M. Nicola).

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Table 1
Pre-treatment typologies of sanitary landfill leachate.

Pre-treatment typology	Contaminant	Leachate typology	Removal efficiency [%]	Reference
Stripping process	Ammonia (ammoniacal nitrogen) COD	Stabilised leachate (methanogenic phase)	70–90 24–47	(Cheung et al., 1997)
Coagulation–flocculation	COD	Stabilised leachate	42–55	(Amokrane et al., 1997)
Coagulation–flocculation	Organic matter	Raw and partially stabilised	25–80	(Tatsi et al., 2003)
Fenton's reagent	COD	Old municipal landfill leachate	60	(Lopez et al., 2004)
Coagulation–flocculation	COD Heavy metals	Raw leachate	21–28 68–91	(Zazouli and Yousefi, 2008)
Air stripping Coagulation and ultrafiltration	Ammonia nitrogen COD	Raw leachate	88.6 84.8	(Pi et al., 2009)
Precipitation process	COD Heavy metals	Raw leachate	25 79–88	(Zazouli et al., 2010)
Coagulation–flocculation	COD Humic acids	Stabilised leachate	55.87–68.65 53.64–80.18	(Liu et al., 2012)
Coagulation and adsorption	COD	Young and stabilised leachate	25–80	(Gandhimathi et al., 2013)
Air stripping, chemical coagulation, electro-coagulation advanced oxidation with sodium ferrate	COD Ammonia	Stabilised leachate	85 50	(Poveda et al., 2016)
Geotextile filters	COD Heavy metals	Stabilised leachate	42 0–51	(Silva and Palmeira, 2017)

biodegradable or refractory to biodegradation (e.g. humic acid), and of inorganic compounds such as: (i) light metals (Al, K, Na, Mg, etc.); (ii) heavy metals and metalloids (As, Ca, Cd, Cu, Fe, Pb; Zn, etc.); (iii) anions (Cl^-, NO_2^-, NO_3^-, SO_4^{2-}, PO_4^{3-}, S^{2-} etc.), and (iv) NH_3 (Fan et al., 2006; Kjeldsen et al., 2002; Qasim, 2017; Slack et al., 2005; Wiszniowski et al., 2006). Whereas anions and light metals are generally present in non-toxic concentrations, the toxicity of heavy metals and As may be considered a threat (Heyer and Stegmann, 2002; Wiszniowski et al., 2006).

According to landfill age, leachate is generally classified as young or stabilised (or mature):young leachate generally presents low pH values (<6.5) and higher values of organic matter content and biodegradability (i.e. COD up to 50.000 mg/L and ratio between biological and chemical oxygen demand - BOD/COD > 0.4) and of heavy metals. Old or stabilised leachate usually presents higher values of pH (>7.5) and NH_4–N (>400 mg/L) and lower values of COD (<3000–4000 mg/L), of the BOD/COD ratio (down to 0.1) and of heavy metals (adapted from Gandhimathi et al., 2013, Foo and Hameed, 2009, Renou et al., 2008).

Conventional landfill leachate treatments can be classified into four major groups: i) recycling of leachate into the landfill body, ii) combined treatment with domestic sewage in external wastewater treatment plants (WWTPs), iii) biodegradation: aerobic and anaerobic processes, and iv) chemical and physical methods: chemical oxidation, adsorption, chemical precipitation, coagulation/flocculation, sedimentation/flotation and air stripping (Hermosilla et al., 2009; Kurniawan et al., 2006; Renou et al., 2008).

The co-treatment of landfill leachate with municipal sewage in WWTPs, after its transportation by trucks, together with the on-site treatment by reverse osmosis, represents the method most commonly used in many countries and in Italy, in particular, for leachate treatment (Calabrò et al., 2018).

However, as suggested by Calabrò and co-workers (Calabrò et al., 2010, 2018) these solutions still present many issues to be solved, the main problems being the following:

- the transfer of heavy metals and of other toxic substances, during the treatment in WWTPs, in the excess sludge and in purified water;
- the presence of compounds (e.g. ammonium, heavy metals) that could inhibit the biological process in WWTPs.

In this context, an appropriate and cost-effective solution, could be a leachate pre-treatment before co-treatment into municipal WWTPs (Gao et al., 2015; Wiszniowski et al., 2006). The aim of the pre-treatment would be the removal of organic and inorganic inhibitory compounds, such as heavy metals, that, as already mentioned, could reduce treatment efficiency or could be transferred to the excess sludge preventing its valorisation (e.g. composting, direct use in agriculture).

As results from the literature, several studies have focused on the pre-treatment of leachate, prior to biological treatment or reverse osmosis, by applying different methods. The most common pre-treatment method is the coagulation–flocculation process (Amokrane et al., 1997; Liu et al., 2012; Tatsi et al., 2003; Zazouli and Yousefi,

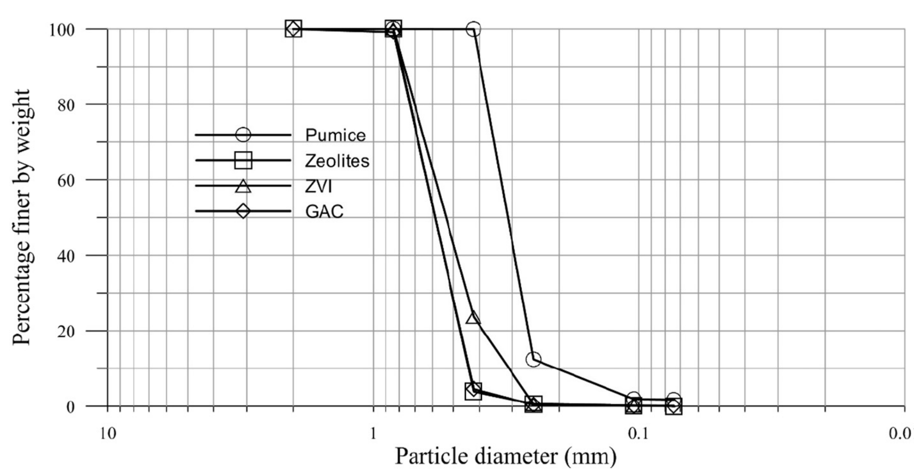

Fig. 1. Grain size distributions of ZVI, GAC, zeolites and pumice.