Leached fine concrete waste's effect on mineral saturation in acid mine drainage

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INTRODUCTION

The difficulty in minimizing pollution derived from acid mine drainage (AMD) has led to incessant scientific work on AMD treatment, seeking to reduce its impact on natural watercourses and surrounding soils. Both passive reaction column-based systems (Delgado et al., 2019) and addition of reactive materials to flooded areas reveal a prolonged positive interaction in the medium and long term (Fernández-Caliani et al., 2022). The current European Union Waste Directive 2008/98/EC includes a reuse of construction and demolition waste (C&DW) of 70%. Despite efforts to reuse concrete waste, the fine fraction (<4mm) is still not included in the circular process for technical reasons in the recycling process (European Commission, 2016). Given this background, this study investigates the chemical speciation of mine waters after percolation through a layer of fine recycled concrete aggregate, which has successfully reduced the metal load of leachate (Barba-Brioso et al., 2022), in order to expand knowledge of the processes that have led to this success, and to be able to implement the experiment at a pilot level in the field.

MATERIALS AND METHODS

A 5L AMD sample was obtained from the Tinto River, on its way through the Cascajal water mill (Huelva), in November 2021. Two all-in-one samples of about 10 kg of C&DW were provided by two Spanish companies dedicated to waste management and revalorization: *Ecoinertes* and *Áridos El Soto*.

150 g of two recycled concrete aggregates < 4mm, called SF and EF, were disposed in filtering vessels, and leached 7 times with 150 ml of AMD over a month. The leachates (AMD-S-*X* and AMD-E-*X*) were filtered at 0.2 μm and analysed for pH, Eh and electrical conductivity with Crison equipment. Major and trace elements (S, Al, Ca, Mg, Fe, Cu, Zn, As, Cd and Pb) were analysed by ICP-OES in a Spectroblue TI instrument at the Mycroanalysis Service of the Research Services department of Seville University (CITIUS). At the end of the experiment, residual solids were studied by high resolution scanning electron microscopy (FEGSEM), to verify the remanent and the precipitated phases, on a FEI Teneo coupled with energy dispersive X-ray spectroscopy (EDS), acquired at 15 kV accelerating voltage to assist in the identification of accessory minerals of environmental concern.

RESULTS

Acidity of AMD (initial pH 2.4) was neutralized since the first leaching (mean pH 7.6). Electrical Conductivity was reduced to natural values (from 11.6 to below 6 mS cm^{-1}) and the extremely high oxidant potential (851 mV) was reduced to 477 mV. Dissolved element concentration in the original AMD, and leachates 1, 4 and 7 are shown in Table 1. Arsenic, cadmium and lead were under detection limits of the technique in all solutions. After leaching concrete aggregates, Al and Fe were depleted in the AMD, while Ca and Mg were added to solution. Sulphur was halved but was still present in leachates. Despite EF grains were coarser than SF, both aggregates behaved analogous, showing no effect of grain size distribution on the interaction with AMD.

The effect of leaching through the aggregates was immediate on AMD (Fig. 1). Original AMD (AMD-0) was rich in elements to form sulphates, oxyhydroxides, but physicochemical conditions only were compatible with saturation of hydroniumjarosite[$(H_3O)Fe_3(SO_4)_2(OH)_6$], lepidocrocite [FeOOH], and schwertmannite [$Fe_{16}(OH,SO_4)_{12\text{-}13}O_{16}\cdot 10\text{-}12H_2O$] (missing from database).

Table 1. *Element concentration in AMD and leachates obtained from the experiment.*

TEST DAY	0	1	7	29	1	7	29
SAMPLE	AMD-0	AMD-S-1	AMD-S-4	AMD-S-7	AMD-E-1	AMD-E-4	AMD-E-7
Al mg L	459	≤ 0.001	0.005	0.021	0.030	≤ 0.001	0.015
Ca mg/L	258	801	679	719	885	733	725
Fe mg/L	1059	≤ 0.001	≤ 0.001	≤ 0.001	0.091	≤ 0.001	0.008
Mg mg/L	376	341	489	388	333	473	468
S g/L	2.67	1.04	1.21	1.19	1.02	1.29	1.29
Zn mg/L	98.6	0.096	0.076	8.78	0.136	0.717	6.13
Cu mg/L	66.5	0.157	0.019	0.122	0.205	0.027	0.142

After reaction with aggregates, output waters saturation indices (Fig. 1) were variable in function of the presence or absence of Fe and Al. When remanent Fe was still in solution (AMD-E-1 and AMD-E-7) amorphous iron hydroxides [$Fe(OH)_3(a)$] and lepidocrocite were possible to form. When Al stayed in solution (except AMD-S-1 and AMD-E-4) pH and redox conditions could allow precipitation of several hydroxylated species like boehmite, diaspore or gibbsite (Delgado et al., 2009). This was extensible for rest of minerals considered to precipitate, which were Cu-retainers species like hydroxysulphates (antlerite, brochantite, langite) and hydroxides [$Cu(OH)_2$].

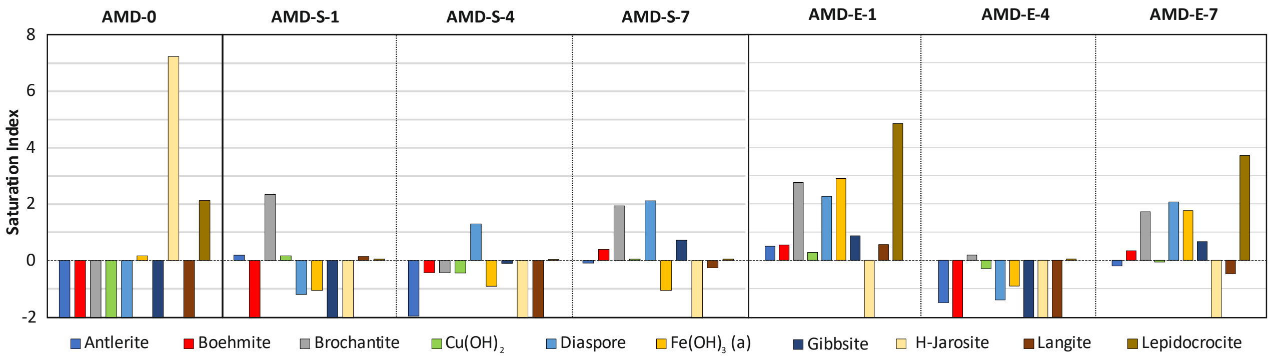

***Fig 1**. Evolution of saturation indices of leachates along the experiment. A saturation index above zero indicates possible precipitation of the phase.*

SEM images and EDS analysis revealed that waters mainly precipitated Al and Fe oxyhydroxides on the gypsum and silicate particles previously present in the aggregates. This oxyhydroxides showed traces of Cu, but Zn could not be seen by microscopy.

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