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Full Length Research Paper

Assessment of short-term leach behavior of waste natural stone slurries mixed into soils in road and earth construction

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The present study was aimed at estimating the leaching behavior of waste natural stone slurries used in earth and road construction. For this purpose, concentration levels of Ca^{2+} and Mg^{2+} ions, which were extracted from the soil and the waste natural stone slurries mixed into soils in road and earthworks by synthetic precipitation leaching procedure (SPLP) were detected. The results obtained from Ca^{2+} and Mg^{2+} analysis implied that each type of the waste materials had a Ca^{2+} and Mg^{2+} concentration lower than the initial concentration for the waste material ratios of 0, 10, 20, 30 and 40% examined after any leaching procedure. In the present study, final pH value of any leaching medium (initially adjusted to three different pH values; 4.2, 5.0 and 6.0) was also considered as an important environmental parameter. The maximum final pH value was found to be 9.8 and it was also found that this value slightly exceeds the permitted value of Turkish regulations.

Key words: Leaching, waste natural stone slurry, soil, road construction, earth construction.

INTRODUCTION

For the sake of avoiding expenditures for transportation and loading-unloading operations of waste natural stone slurries, most of the producers dispose their wastes into vacant lands and valleys located next to their factories and/or store the waste in somewhere else between the factory and the pit even though there can be found several feasible places assigned to the producers in order to dispose the waste. Annually, approximately 2.7 million tons of waste natural stone slurry, which is generated as by-products from processing of large quantities of natural stones such as marble and travertine of 9 million tons in mine industry, are filled into vacant lands in Turkey (Stone, 2007; Yildiz and Eskikaya, 1995). When these waste marble slurries get dry, the dried material causes the lands to occupy, where the lands are vast and feasible for agricultural activities and affects the environment negatively due to the formation of dust-associated pollutions or contaminations in groundwater. Furthermore, disposal of such wastes into agricultural lands reduces the efficiency of topsoil since the waste also causes the physical characteristics of the topsoil to reduce such as porosity, permeability and water adsorption. Also, the places, where marble slurries are disposed, have negative effects on the growing of any vegetation. Therefore, the depletion of the rich arable lands, the increase in costs for the waste disposal and the need for the undamaged earth has increased the need for recycling of these materials in road and earth construction. Although wastes of marble slurries and

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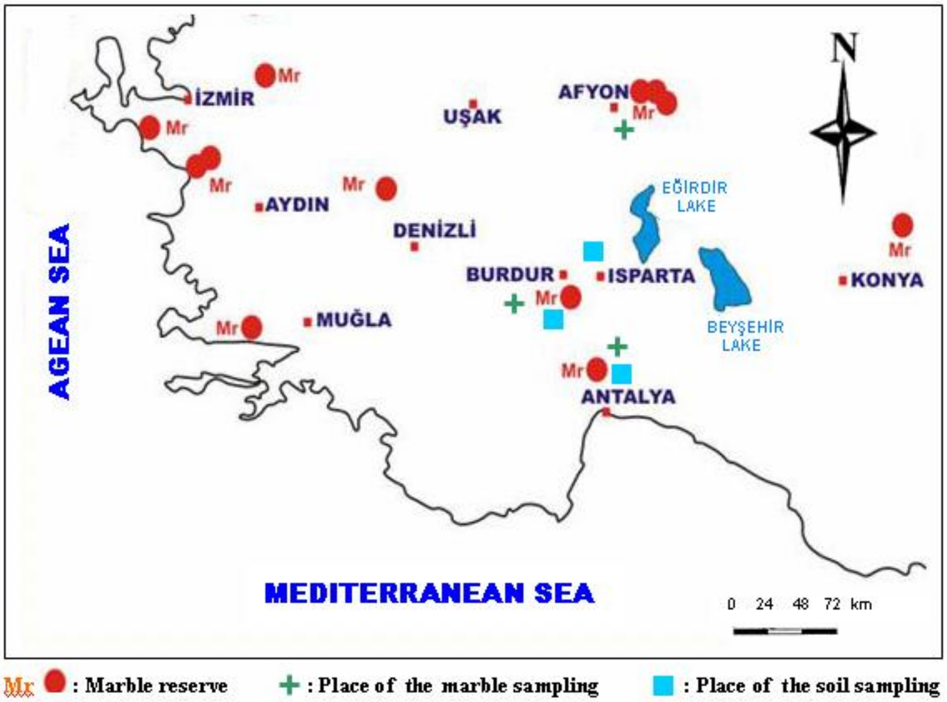

Figure 1. Location map of study area.

small marble pieces are being used in marble workshops as covering and filling materials, such wastes are the industrial wastes generating nearby the factories in bulk.

Utilizing of these wastes undoubtedly contributes great economical values to the finance of countries and factories, and minimizes the costs for the storage of the waste and the negative environmental impacts. However, there has not been any sector or field in which these wastes are utilized extensively so far. One of the barriers to the wide-ranging utilization of the secondary products of energy production and industry in road construction has been uncertainty about the environmental impacts. In order to prevent potentially hazardous effects, it is important to identify all the positive and negative impacts of these waste materials. The leaching analysis for life cycle impacts is also being increasingly used as a selection for products and materials both in industry and in other activities. Leaching tests are used to determine the availability and actual release of elements that can intrude and pollute the groundwater (Fällman and Hartlén, 1996; Tossavainen and Forsberg, 1999). When secondary materials such as slag and waste slurry are to be evaluated in road construction, the focus of attention is on their leaching characteristics. The special characteristics of road construction and earthworks are large volumes of the materials consumed, the long service lives of the finished products, the need to assess construction as a whole rather than comparing alternative materials, and the significant effect of the construction's longevity and need for repair on their life cycle environmental loadings (Laine-Ylijoki et al., 2001). This paper was aimed at detecting the concentration levels of Ca^{2+} and Mg^{2+} ions followed by the leach extraction of three different kinds of waste natural stone slurry mixed into three different kinds of soil used in road and earthworks by using a modification of E.P.A.

Method 1312, synthetic precipitation leaching procedure (SPLP) (USEPA, 1994). Also, this study is a part of more extensive project 'the use of the waste natural stone slurry and pieces in road and earth construction' that is similar to a work carried out by Okagbue and Onyeobi (1999).

MATERIALS AND METHODS

Site description and sample collection

The study area comprise Isparta, Antalya, Burdur and Afyon provinces from natural stone production sites in south of Turkey that is showed in Figure 1 (MTA, 2009). Waste stone slurry came out while treated by used saw and abrasive materials was taken with wastewater collectors from waste basins of three factories treated the different natural stones in the district. Wastes of Bucak Travertine, Afyon Sugar and Burdur Beige were chosen for natural stone sampling because they were generally produced with natural stone variety in the site. The fraction passing the sieve size of 63 µm was used for the tests. Soil samples were taken from three different places near to the roads between provinces. Soil sampling was carried out in pits excavated to a depth in excess of 30 cm to expose fresh samples and avoid the influence of vegetation and sieved from sieve #4 according to road and earth construction. The soil samples were thoroughly mixed in the laboratory and a

Table 1. Particle size distribution of the soil and waste slurry samples.

	Soil type	Clay and silt (%)			Sand (%)	Gravel (%)
	Particle size (mm)	**0.09**	**0.37**		**0.74**	**4.75**
Natural stone dust	Burdur Beige	70	27	3	0	0
	Afyon sugar Beige	35	36	14	15	0
	Bucak travertine	94	6	0	0	0
Soil samples	Antalya highway	38	9	4	21	28
	Burdur highway	65	13	4	18	0
	Isparta highway	3	4	7	76	10

quartering process was used to get representative samples for each test (Okagbue and Onyeobi, 1999; Gertsch et al., 2000).

Sample leach characterization

Natural stones have different categories in themselves. Burdur Beige and Afyon Sugar are included in the marble category while Bucak travertine is included in the travertine category. The results obtained from the characterization analysis in earlier studies (Ersoy, 2005; Aluko et al., 2003) showed that there is a CaO compound of about 54% as the highest quantity and a MgO compound of 0.22 to 0.42% as the secondary highest quantity in the chemical structure of stone slurry. According to these results, only Ca^{2+} and Mg^{2+} analysis was conducted for each waste slurry sample in the present study. The same analysis was applied to the soil samples before mixing them with waste slurry samples in order to make a comparison for test results in case mixing with waste slurry. Ca and Mg ions were analyzed according to the US EPA approved HACH-method 8009 (HACH, 1979; Wik and Dave, 2006) and measured with a DR/4000 U spectrophotometer (HACH, Loveland, CO, USA). Out of the chemical analysis explained earlier, the size of soil samples was analyzed as a physical parameter examined after getting a powder from the dried slurry samples (ASTM D 422, 2000).

Batch leaching tests

The synthetic precipitation leaching procedure (SPLP) (USEPA, 1986, 1994), which is also known as the short-term batch leaching test was used in this study. SPLP is a method designed to evaluate the impact of contaminated soils on groundwater. The leach medium consists of slightly acidified deionized water that is formulated to simulate natural precipitation. A mixture of 60/40 HNO_3 (by weight) was used to achieve the appropriate pH for the leach medium. The solid was extracted at a 20:1 ratio (2.000 g leach medium/100 g of soil-waste composite material) on an end-over-end rotary agitator for 18 h. The solid/liquid slurry was then filtered through a 0.7 μm borosilicate glass fiber filter utilizing a pressure filtration unit (Hageman et al., 2000; Al-Abed et al., 2006). In this SPLP procedure, the leach medium was adjusted to three different acidic pH values (4.2, 5 and 6) depending on the acidity of rain waters measured in the study area.

RESULTS AND DISCUSSION

The change in the results of leach analysis was assessed according to the initial pH (adjusted before the leach extraction) as well as physical features such as the specific gravity and the particle size of the soil and slurry samples which are from important parameters of road construction and also affect the leach procedure due to the surface area. In this way, the particle size distribution of the samples is presented as physical characteristics in Table 1 (Filgueira et al., 2006; Assallay et al., 1998). In addition to this table, Ca and Mg quantities of the raw soil and raw slurry that were not mixed with each other are presented as chemical characteristics in Table 2. In Table 2, it is seen that Ca and Mg quantities of slurry samples are higher than the values for soil samples due to the magnesium and calcium oxide content of natural stones (Ceylan, 2000).

Ca^{2+} and Mg^{2+} analysis

It is seen that slope of the curves for the obtained concentration of Ca^{2+} and Mg^{2+}, which were resulted from the leach extractions applied to the soil-slurry mixture, resembles each other by graphical means (Figures 2 to 7). In case of 10% of waste material usage in the mixture, Ca^{2+} and Mg^{2+} concentrations decreased for each type of the waste materials. Ca^{2+} and Mg^{2+} concentrations showed an increase for travertine and Afyon sugar in case of using 20% of waste material while there was slightly a decrease and even the concentration partially remained constant for Burdur Beige under the same condition. In case of 30 and 40% of waste usage, the leach extraction of travertine and Afyon sugar decreased while the leach extraction of Burdur Beige increased. Unlike the trend observed for wastes of travertine and Afyon sugar, not increasing the leach extraction of wastes of Burdur Beige remarkably in case of 20% of waste ratio and showing an increase in case of 30 to 40% waste ratio may be attributed to the Ca^{2+} and Mg^{2+} concentration existing in its composition. As shown in Table 1, waste material of Burdur Beige contains 33% less Ca^{2+} ions than that of the other two types in the composition. Being the increase in Ca^{2+} and Mg^{2+} concentration of travertine partially higher than that of Afyon sugar in case of 20% waste ratio resulted from

Table 2. Quantities of pollutant in the raw soil and the raw slurry.

	Soil samples			Natural stone slurry		
Total waste material (g/kg)	**Antalya highway**	**Burdur highway**	**Isparta highway**	**Burdur Beige**	**Afyon sugar**	**Bucak travertine**
Ca	0.016	0.029	0.028	88	109.6	118.4
Mg	0.006	0.016	0.009	26.8	29.6	32.8

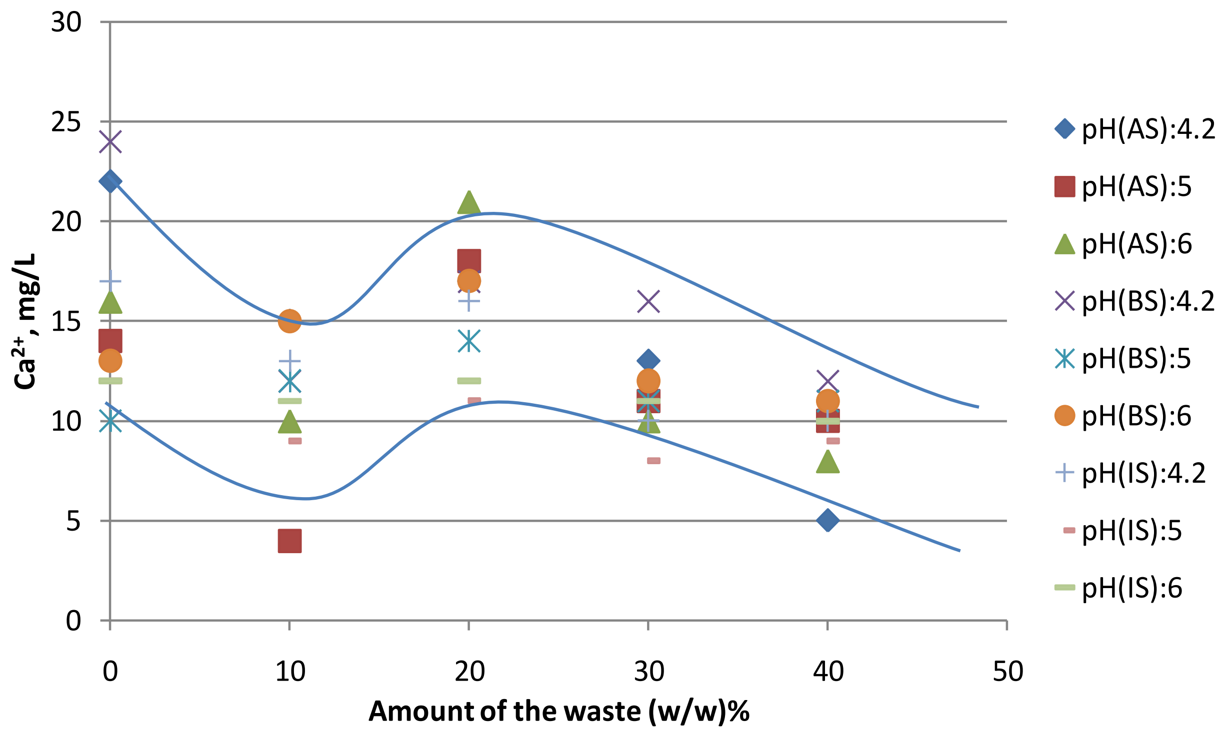

Figure 2. CA^{2+} concentrations of the leach extraction for travertine.

being the Ca^{2+} and Mg^{2+} concentration in the composition of travertine is relatively higher than that of Afyon sugar. In case of increasing the waste ratio above 20%, the decrease for travertine and Afyon sugar caused to obtain similar results.

The effect of the change in the three different initial pH values of the examined waste materials on the results of the leach extraction was different for each of the materials under the different waste ratio conditions. Therefore, a certain constant effect on the results of the leach extraction could not be recorded. Similar to the effect of the change in pH, soil materials used in the mixture had no certain effect on the results of the leach extraction. However, the results of the leach extraction for Ca^{2+} and Mg^{2+} ions were obtained differently for the raw soil materials under the

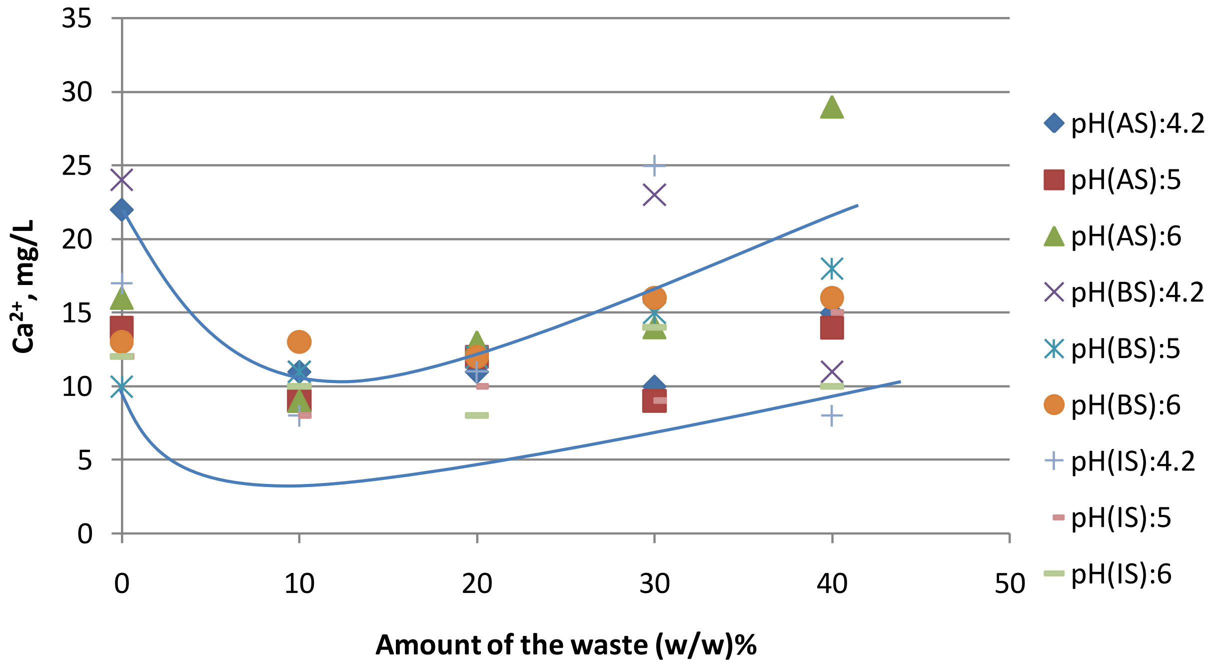

Figure 3. CA^{2+} concentrations of the leach extraction for Burdur Beige.

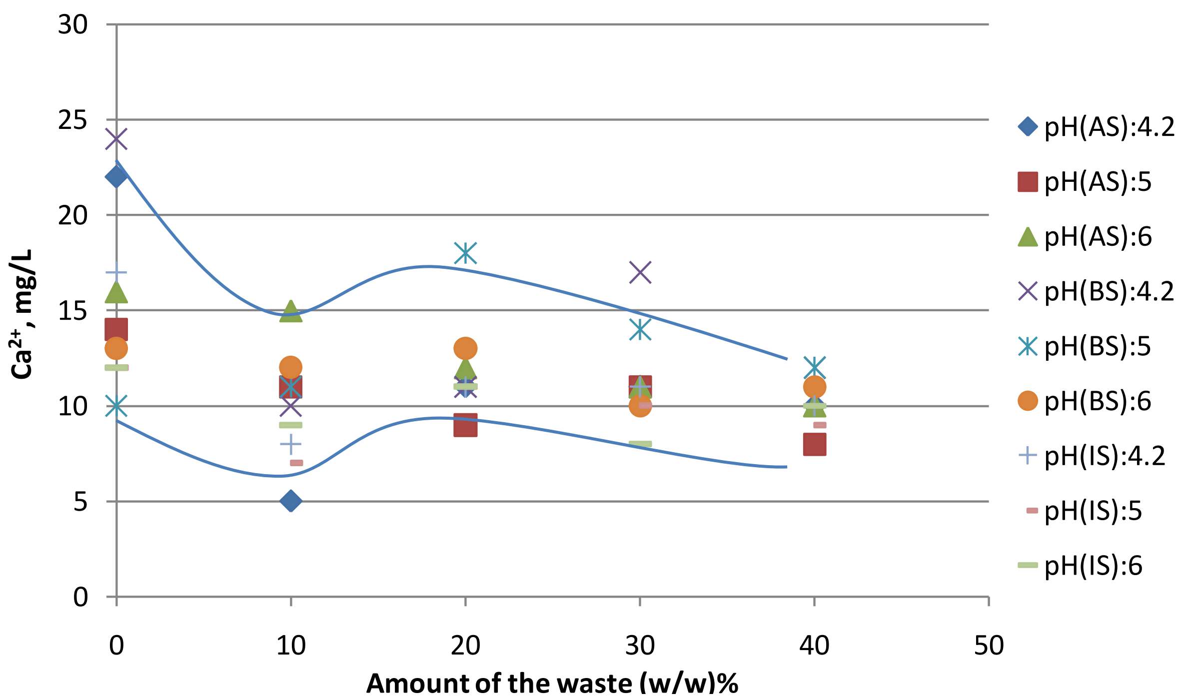

Figure 4. CA^{2+} concentrations of the leach extraction for Afyon sugar.

different waste ratio and marble type conditions. It was observed that the effect of variety in pH and soil samples on the leaching analysis had a complex nature. It is thought that this case took place due to the complexity of the chemical interactions existing between the marble slurry wastes and soil materials. Okagbue and Onyeobi (1999) stated that stabilization of marble slurry wastes carried out with soil has features similar to that of the stabilization carried out with lime. Also, their study reported that $Ca(OH)_2$ complexes existing in the marble

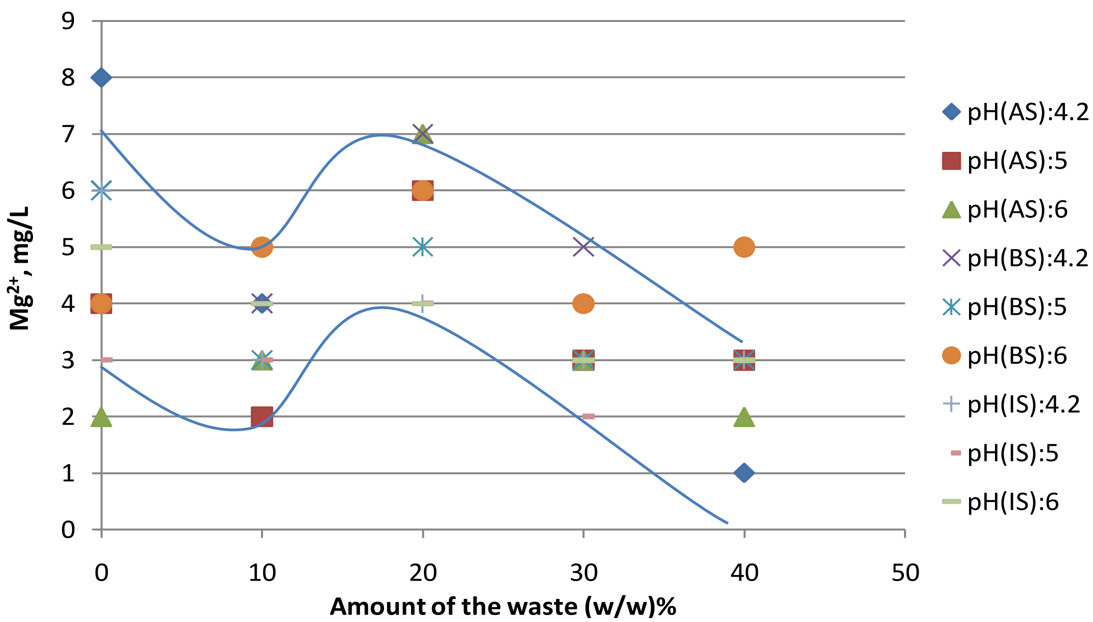

Figure 5. MG^{2+} concentrations of the leach extraction for travertine.

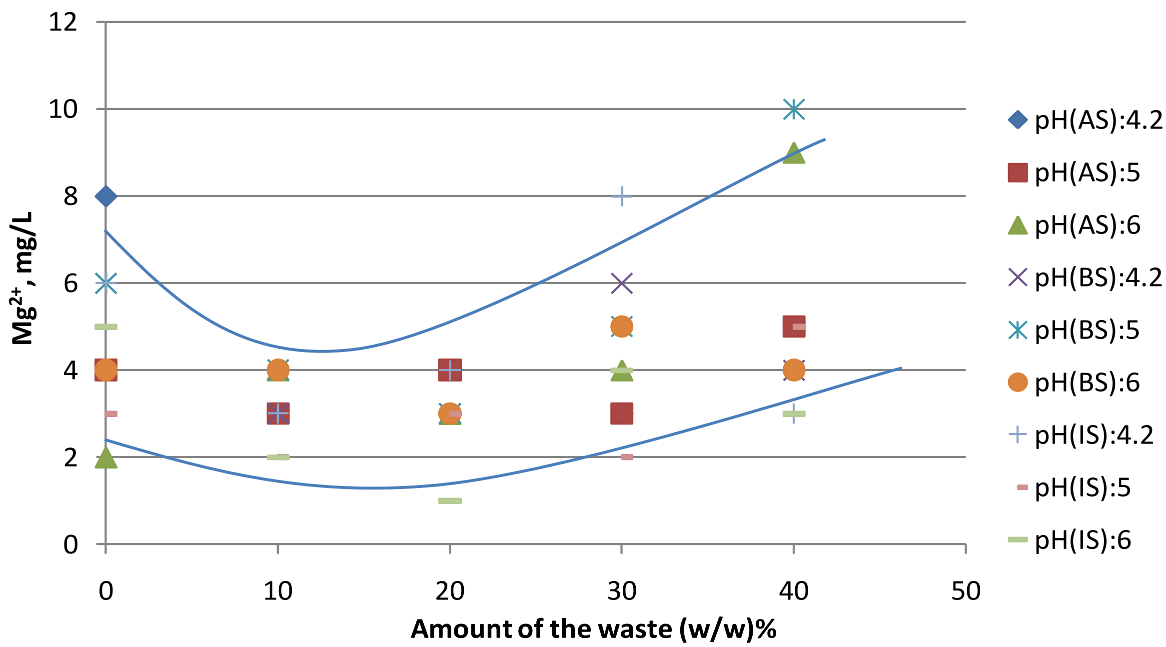

Figure 6. MG^{2+} concentrations of the leach extraction for Burdur Beige.

slurry deionized in water and Ca^{2+} cations formed complexes with the other cations existing in the soil materials. In the present study, showing Mg^{2+} ions, which were investigated, similar features with Ca^{2+} ions implies that $Mg(OH)_2$ complexes existing in the waste materials deionized in the leach medium and Mg^{2+} cations interacted with soil materials. In addition to that, it is thought that decrease in the concentration of Ca^{2+} and Mg^{2+} ions in the leach medium for 30 and 40% of wastes of travertine and Afyon sugar resulted from the mentioned interactions.

pH analysis

The effect of pH is rather high while determining results

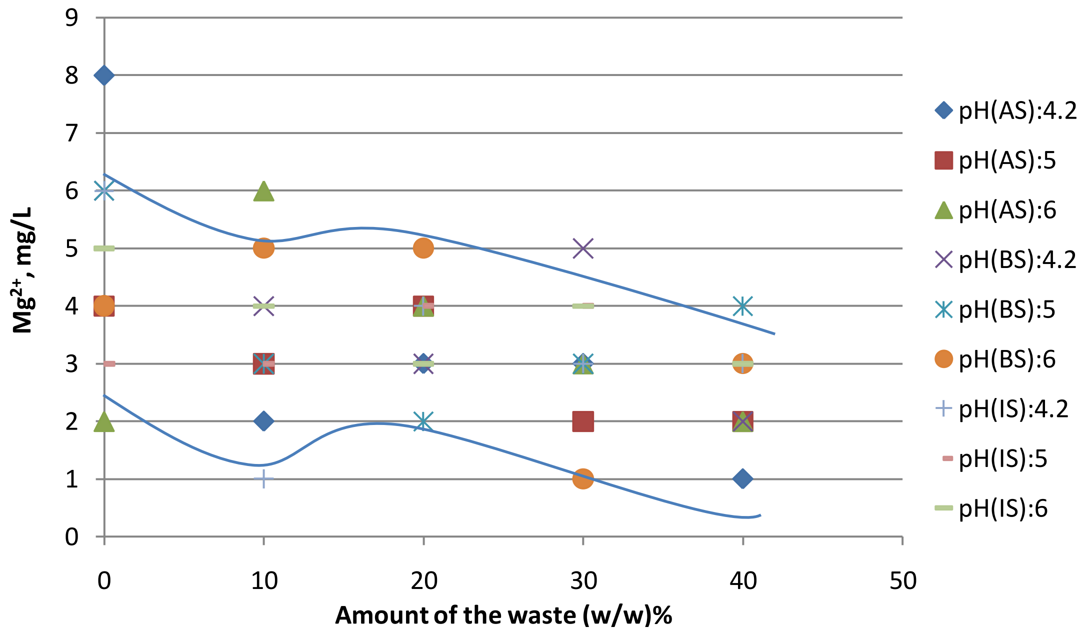

Figure 7. MG^{2+} concentrations of the leach extraction for Afyon sugar.

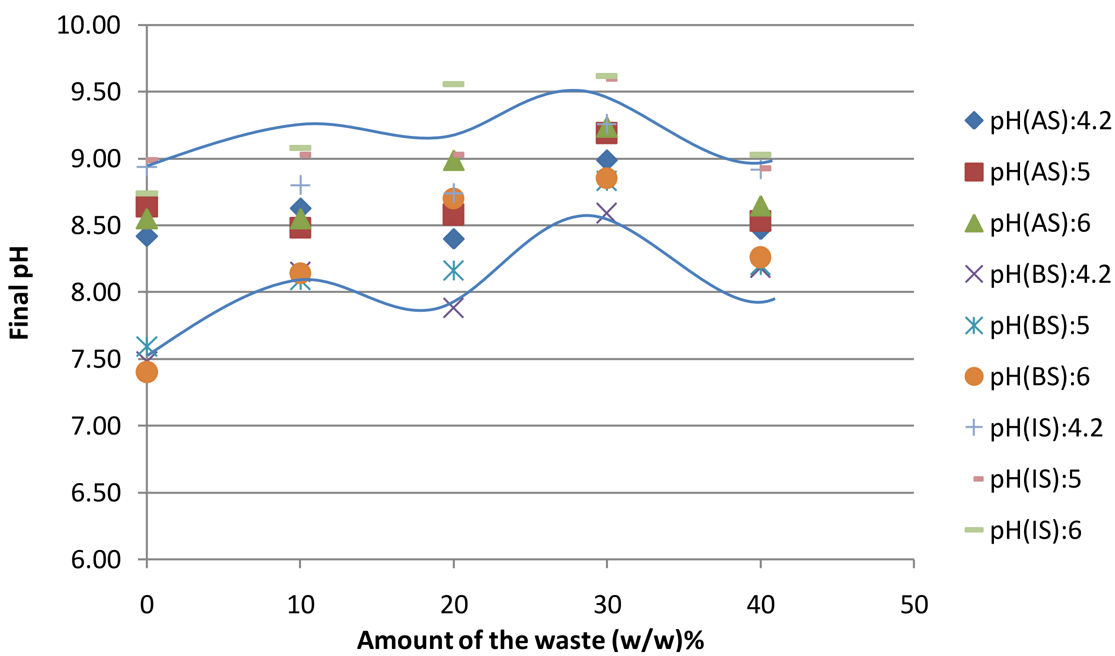

Figure 8. Final pH values of the leach extraction for travertine.

of both soil and waste leaching tests (Cooper, 1991; Sauve´ et al., 1997). Final pH of the leach medium increased due to OH^- ions of which the amount were two-fold higher than that of Ca^{2+} and Mg^{2+} ions formed after extracting of Ca(OH) and Mg(OH) complexes in the waste slurry (Delgado et al., 2006). Initial pH value (adjusted to 4.2, 5 and 6) increased up to 8 or 9 after the extraction (Figures 8 to 10). Mixing of each of the waste slurry materials with soil samples in a ratio of 10 and 20% of waste slurry increased the final pH of the leach medium. In case of 30% of waste slurry ratio, final pH of the medium increased up to a certain level for travertine while it decreased for Afyon sugar and Burdur Beige. Conversely, final pH of the medium showed an increase for Afyon sugar and Burdur Beige, while it decreased for travertine in case of 40% of waste slurry usage. When

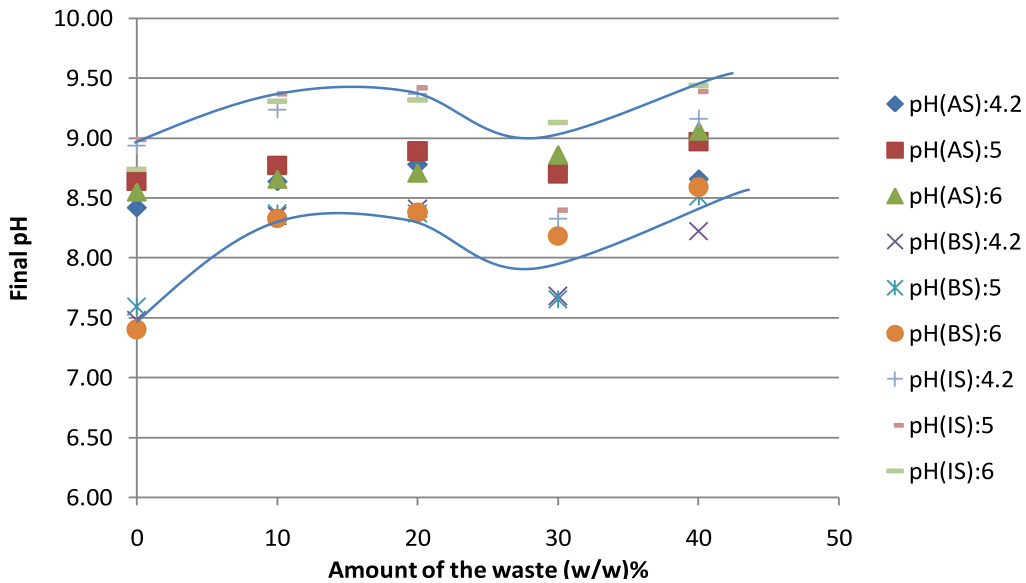

Figure 9. Final pH values of the leach extraction for Burdur Beige.

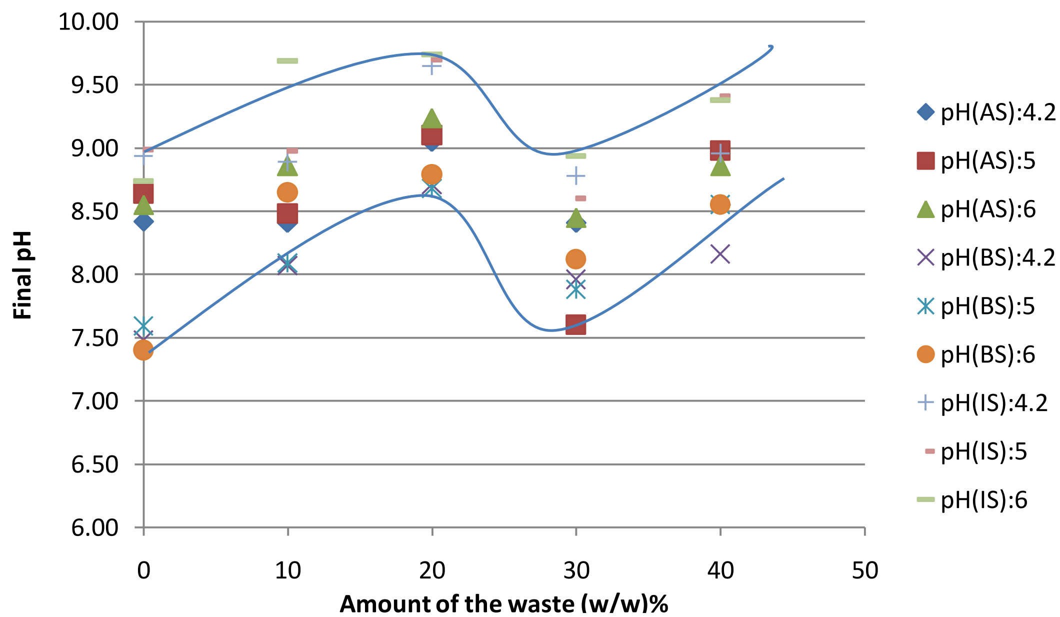

Figure 10. Final pH values of the leach extraction for Afyon sugar.

compared with the other materials, even though it may be thought of showing the final pH value of the leach medium, an increase for waste slurry of travertine resulted from the higher amount of fine particle size and the higher concentration of Ca^{2+} and Mg^{2+} ions together with OH^- ions in the nature of travertine, the main reason is thought to be the interactions taking place between the waste slurry material and soil sample.

Increasing of the final pH value for Afyon sugar and Burdur Beige and decreasing of it for travertine in case of 40% of waste slurry ratio also confirms this phenomenon. Similar to the effect of variety in pH and soil samples on the results for Ca^{2+} and Mg^{2+} concentrations, the variation had no remarkable effect on the result obtained for final pH values.

Risk assessment

The maximum concentration level of Ca^{2+} and Mg^{2+} of the mixtures did not exceed the value of the raw soil samples for travertine and Afyon sugar whereas it was found to be 30 to 40% of higher than that of the raw soil samples for Burdur Beige, which equals 29 and 10 mg/L for Ca^{2+} and Mg^{2+}, respectively. When these results are compared with the standards of TSE266 assessed for the quality of drinking waters by Turkish Standards Institute (Varol et al., 2008), it is understood that the obtained results for each sample are lower than the permissible value for Ca^{2+} by 75 mg/L and Mg^{2+} by 50 mg/L. This finding may also imply that the waste material has no risk of contamination of groundwaters in terms of Ca and Mg after it is extracted by leaching procedure before using as a road construction material. The maximum final pH value was recorded as 9.8. This value exceeds the limit value of 9.2 permitted by Turkish Standards Institute (Varol et al., 2008). It is seen that sample of Isparta soil which is classified as a volcanic and sandy material further exceeded the limit value.

Conclusions

In this study, short-term leach behavior of three different types of waste natural stone slurry mixed into soil in road and earth construction was investigated and the environmental impact of these materials that may arise from any road and earth construction was discussed. For this purpose, the change in Ca^{2+} and Mg^{2+} concentrations and the final pH values in the leach medium formed after each leach extraction study were examined as a function of different waste marble slurry ratios and initial pH values. The results obtained in the study imply that none of the waste marble slurries examined have environmental impact in terms of Ca^{2+} and Mg^{2+} concentrations after applying 'the synthetic precipitation leaching procedure' and that the maximum final pH value slightly exceeds the permitted value according to the Turkish regulations. From this point, it is clearly understood that waste marble slurry materials examined in this study can be used and utilized in road and earth construction without causing any environmental impact.

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