

UTILIZING PLASTIC WASTE MATERIALS IN GEOTECHNICAL ENGINEERING: A SUSTAINABLE SOLUTION FOR ENVIRONMENTAL CHALLENGES

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ABSTRACT. This work evaluates the chemical impact of polyethylene terephthalate (PET) as plastic waste material used in clayey soil improvement. The purpose of this work is to study the behaviour of PET added in different percentages in soil, and how it affects the chemical composition of soil by applying rapid ageing cycles. The thermal stability of the clay-PET mixtures using the thermogravimetric method was studied, and the impact of PET quantity on pH was measured. To detect the migration of PET degradation products to the soil gas chromatography with mass spectrometry (GC-MS) was used. The GC-MS analysis results indicate the presence of DIBP, the concentration increased proportionally with the amount of PET added to the clay. The thermogravimetric analysis shows a thermal degradation of the polymeric chain starting from 200 °C.

Keywords: soil, PET waste materials, thermal stability, GC-MS analysis, additives migration

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INTRODUCTION

Finding innovative and sustainable solutions to the world's problems of plastic waste pollution and soil degradation has become a top priority. Geotechnical engineering and soil improvement present unique opportunities to address both issues simultaneously. As the world faces an escalating environmental crisis caused by plastic waste pollution, finding long-term solutions to this problem has become critical. Soil is a natural resource, providing a foundation for agriculture, infrastructure, and ecosystems. The foundation of any land-based system is critical, and it must be strong for the system to function properly. Some soils cannot withstand heavy loads due to their low bearing capacity. The utilization of various plastic waste materials to improve soil properties has gained traction as an eco-friendly and sustainable approach. In order to fix this issue, the soil must be treated with various stabilizers such as cement, furnace slag, fly ash, limestone fines, bitumen, plastic waste, and so on. It is defined as the process of improving soil properties through physical or chemical treatments, as well as the blending and mixing of other materials with the soil [1]. Plastic waste that would otherwise take hundreds of years to degrade can be given a new purpose, thereby reducing its impact on ecosystems and mitigating its contribution to climate change. By diverting plastic waste from traditional disposal methods, engineers can contribute to a cleaner environment and lower greenhouse gas emissions associated with plastic degradation [2].

Various types of plastic waste, including high-density polyethylene (HDPE) and polypropylene (PP), can be transformed into geosynthetics, such as geogrids and geotextiles. These materials provide improved engineering properties, such as increased tensile strength, soil stabilization, and improved erosion control [3]. Geosynthetics reinforce the soil structure and are especially useful in infrastructure projects like roads, embankments, and retaining walls [4]. These materials, properly processed, and mixed with soil, can improve properties like shear strength, compressibility, and permeability, making it beneficial in various geotechnical applications. Plastic waste can be used as a lightweight fill material, helping to reduce the overall weight of a structure or project. This application is especially useful when there are weight restrictions or the need to relieve stress on weak soils, minimizing settlement and the risk of soil failure in construction projects [5].

Plastic waste acts as a geogrid, reinforcing the soil matrix and preventing erosion, making it valuable for slope protection and land reclamation projects [6]. When LDPE is mixed into soil, it acts as a water reservoir, reducing

evaporation and preventing moisture loss in arid areas. This feature is useful in agriculture, landscaping, and afforestation efforts because it promotes plant growth and aids in drought resistance [7].

One of the primary concerns with using plastic waste materials in geotechnical engineering is their long-term durability. Exposure to environmental factors like UV radiation and chemical degradation can lead to a reduction in their effectiveness over time. Research is ongoing to develop methods to enhance the durability of plastic waste-based geotechnical solutions [8]. Research about the strength properties of subgrade soil reinforced by recycled Polyethylene terephthalate fibers in combination with fly ash was carried out. The optimal properties of subgrade soil were acquired at 1.2 % recycled Polyethylene terephthalate fibers and 15 % fly ash [9]. The inclusion of plastic chips of waste bottles into the sand increased the shear strength of the sand. The optimum percentage of the plastic chip was found to be 0.75% [10]. The California bearing ratio (CBR) and the shear strength of sandy soil were improved by mixing recycled polyethylene terephthalate chips in three different ratios 2%, 4%, and 8 % [10].

It has been established that using plastic waste chips as a soil stabilizing material for subgrade soil in highway subbase construction can significantly reduce the base layer thickness of flexible pavement [11].

The influence of polyethylene waste admixture or cement on the properties of soil was studied by Nicoleta-Maria Ilies et al. (2017). Several tests were carried out using 2%, 4%, 6%, and 8% polyethylene material or cement. They concluded that using waste polyethylene material in soil stabilization is an environmentally friendly method. By incorporating 4% of polyethylene waste into the soil, significant results were observed; however, the improvement was not as substantial as the results of cement admixture, however, this method results in a larger carbon footprint, making it less environmentally friendly. [12,13].

Significant increases in the soil's shear strength (SS) parameters were obtained by mixing small strips of plastic waste in the soil in 0.5%, 1%, and 2% by weight of dry soil [14].

In this paper, we aim to evaluate, using appropriate and simple methods, the effect of recycling plastic materials as reinforcement material for expansive soil stabilization in order to improve and achieve the properties required for construction works. For instance, the chemical impact of using recycled plastics, in different proportions, on soil characteristics was evaluated. The experimental tests that were carried out and the results obtained are presented.

RESULTS AND DISCUSSION

The use of plastic waste materials in geotechnical engineering is a current topic, with several specialist studies in this direction. The improvement of soil by adding various plastic waste materials (including polyethylene terephthalate), in different forms, in order to increase its physical and mechanical characteristics has been studied.

Following the encouraging results of researchers on the improvement of soils with plastic waste materials, this paper studied the behaviour of PET in different percentages used in a clayey soil, respectively how it affects the analysis of its chemical composition by applying rapid aging cycles. In order to follow the chemical degradation of PET in the soil under normal atmospheric conditions, different samples were achieved. The percentages of PET used for improvement were 2%, 4% and 6%, reported to the dry weight of the clay. For a better identification of PET degradation products in the soil, depending on the freeze-thaw cycles (Table 1), the chemical composition of PET was taken into account. PET is a high-molecular-weight thermoplastic, semi-crystalline polymer, obtained from the reaction between terephthalic acid and ethylene glycol. The acetaldehyde, formaldehyde, 2,4-di-tert-butyl-phenol (2,4-dtBP), and bis(2-hydroxyethyl) terephthalate are the main compounds released from PET due to the UV sunlight and high temperature. The phthalates, namely benzyl butyl phthalate, bis(2-ethylhexyl) phthalate (DEHP), dibutyl phthalate, and di-isobutyl phthalate, as plasticizers, can be identified as degradation compounds [15].

Thus, 4 samples named Cl, Cl_{2PET}, Cl_{4PET}, and Cl_{6PET} were obtained. These were made in the laboratory, by mixing 2000 g of clay with 2%, 4%, 6% PET (reported to the dry weight of the clay). The mixtures have been brought to the optimum water content (OWC) of 18%. Cylindrical samples with a diameter of 10 cm and a height of 10 cm were collected from the Proctor apparatus. The samples have been introduced into the climatic cabinet and have been subjected to variable temperature and humidity conditions. The details of the composition, obtaining procedure and mechano-physical characteristics are given in Table 1.

In order to detect the migration of PET degradation products to the soil, gas chromatography coupled with a mass spectrometer was used. To identify potential migrants in plastic packaging materials, solvent extraction is commonly used, followed by chromatographic separations and mass spectrometric detections. Samples (plastics and clay, respectively clay mixed

with different percentages of PET) were kept in a paper bag, to exclude the contamination from the packing bag. Recycled PET materials were analyzed before and after mixing with clay. The plastic materials were chopped into small pieces (~0.5 x 0.5 cm). The extraction process is critical for determining plasticizers in polymers prior to analysis. Soxhlet extraction [16,17] and ultrasonic-assisted extraction [18] are two commonly described procedures for extracting these organic compounds from polymeric matrices. Microwave irradiation has recently been used in the field of sample preparation because it is a simple and quick methodology [19].

Table 1. Composition of clay-PET mixtures and conditions of applied rapid ageing cycles

Samples code	Composition	Freezing-thawing cycles	Temperature range
Cl	Clay + 18% water	-	-
Cl ₂ PET	Clay + 18% water + 2% PET	15	-10 °C ÷ +40 °C
Cl ₄ PET	Clay + 18% water + 4% PET	15	-10 °C ÷ +40 °C
Cl ₆ PET	Clay + 18% water + 6% PET	15	-10 °C ÷ +40 °C

PET sample (0.5 g) was weighed, placed in a 100 mL conical flask with a glass top, and extracted with dichloromethane (DCM) (2 × 10 mL) by ultrasonic assisted extraction (UAE) for 30 min at 25 °C. The extracts were combined and evaporated to dryness under 40 °C by a rotary evaporator. The concentrated mixture was redissolved in 1.5 mL DCM and subjected to GC-MS analysis. The extraction of each sample was carried out in triplicate.

Analysis of PET/ PET with clay samples by GC-MS: a volume of 2 µL of sample concentrated extract was injected in splitless mode into a gas chromatograph (GC-2010) from Shimadzu Japan, coupled with a mass spectrometer QP 2010 PLUS. The GC was equipped with a capillary column ZB5-MSplus (Phenomenex, 30 m length × 250 µm inner diameter × 0.25 µm film thickness). The carrier gas was He at 0.8 mL/min, injection temperature 220 °C. The oven temperature program began with an initial temperature of 50 °C for 10 min, and then the temperature was increased at a rate of 20°C/min to 310 °C and then maintained for 20 min. The mass spectrometer parameters used for the identification of chemical compounds were as follows: electron impact ionization; electron energy, 70 eV; with an acquisition range (m/z) from 35 to 800 in scan mode, at an acquisition rate of 500 ms, ion

source, and interface temperature 220 °C. The resultant compounds were identified by matching their mass spectra to the NIST (NIST 27, 147 libraries), WILEY library database (>90% match).

The results showed the presence of several chemical compounds in the PET samples studied, chemicals that are present in large quantities are 1,4-benzenedicarboxaldehyde, hexadecenoic acid, long alkyl chain (eicosane, tetracosane, heneicosane). Lubricants are a class of additives that are used to reduce friction between the equipment surfaces and the polymer, allowing the plastic to flow over and through the equipment and keeping it from sticking to the surfaces [20]. Carboxylic acids, their esters and amides, polyethylene waxes, paraffin, stearates, and silicones are the most common lubricants used in plastic packaging materials. Lubricants can generally be added to plastic resin or applied externally. Hexadecanoic acid and octadecanoic acid were found in all analyzed samples, indicating their widespread use as lubricant main components in the samples studied [21]. Carboxylic acids are generally recognized as safe (GRAS), and EU regulations allow them to be used as additives or polymer production aids.

Plasticizers are used to improve the flexibility, extensibility, and processability of plastic packaging materials [22]. One plasticizer was discovered in this study, 1,2-benzenedicarboxylic acid, bis(2-methylpropyl) ester (DIBP).

PET samples (PET1 and PET2) used in these tests were extracted by UAE and analyzed based on the method described above. The major compounds found were terephthalaldehyde, terephthalaldehydic acid, hexadecenoic and octanoic acid (Figure 1). In further experiments, we used the mixture obtained by mixing the two PET samples.

Analysis of starting clay-PET mixture in different percentages was measured (Figure 2). It can be observed that the quantity of used PET has no or has a small influence on the chemical composition of the clay. In the chromatogram of the sample with a 6% PET - clay mixture presents of lubricants can be observed, hexadecanoic acid, octadecanoic acid, and aromatic carboxylic acids.

After applying the aging cycles, in the chromatograms of samples can be observed a higher number of peaks (Figure 3), which can be attributed to degradation products from used PET plastic wastes, respectively different additives which have been migrated to the clay. The chromatograms of the three samples are similar, but differences can be observed in the concentration of the separated compounds. The higher concentration for DIBP (retention time 20.12 min) was found in sample 6% (Figure 4).

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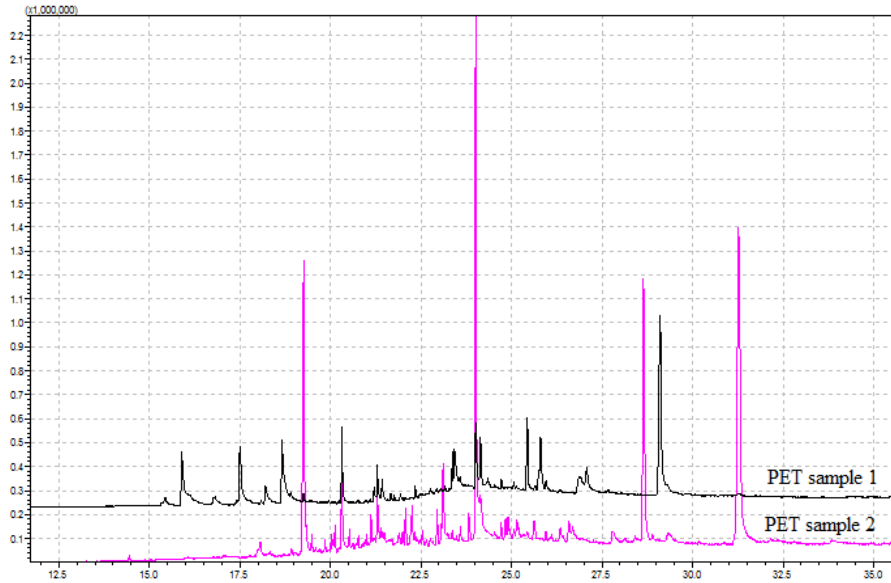


Figure 1. Gas chromatogram of used plastic waste PET (bottles used for water, soft drinks, and beer).

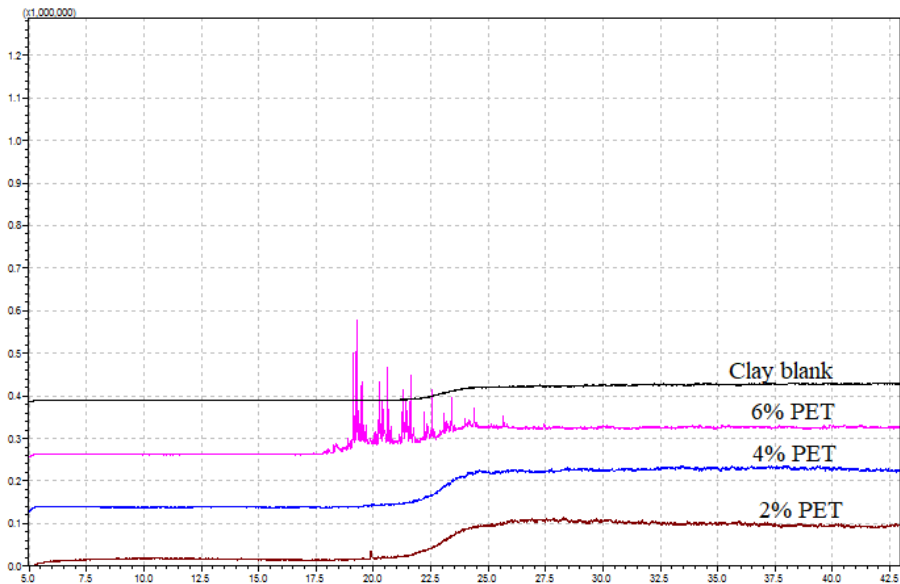


Figure 2. Gas chromatograms of clay samples with different percentages of chopped PET after mixing, before aging cycles.

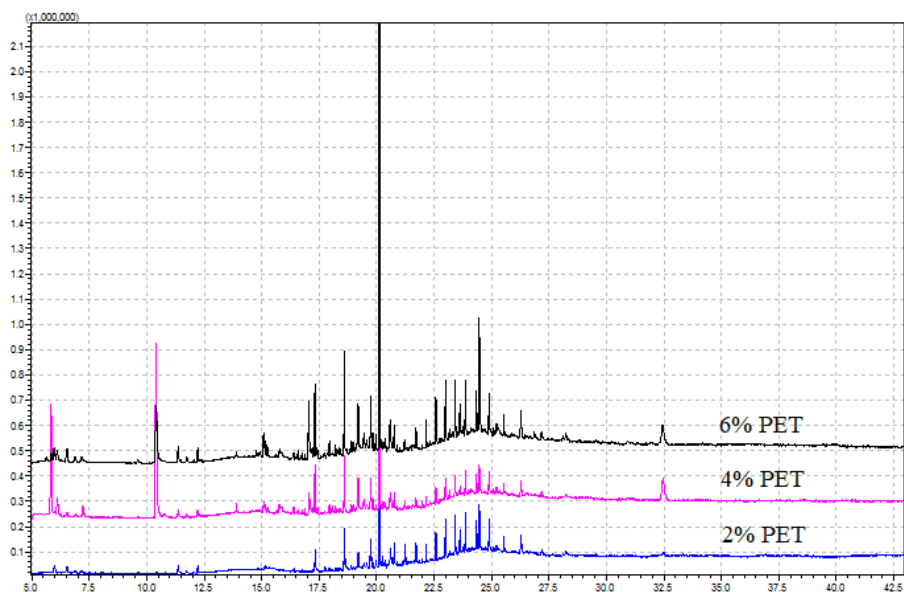


Figure 3. Gas chromatograms of the clay-PET mixtures after applying the aging cycle.

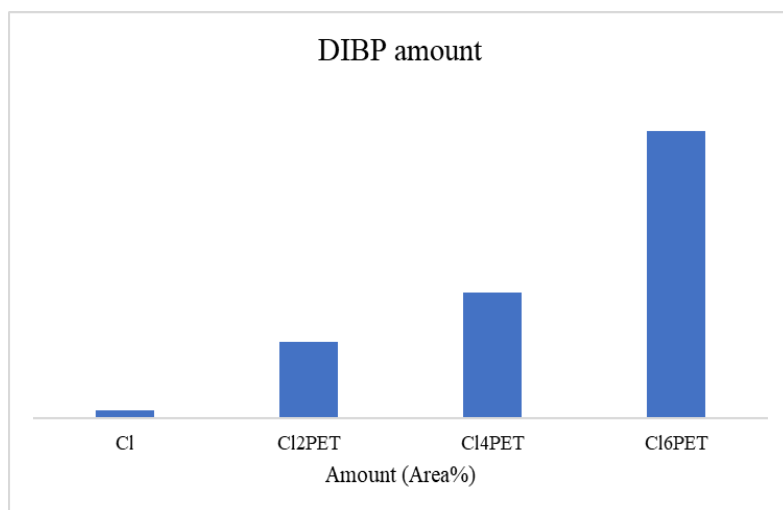


Figure 4. Graphic representation of the amount of DIBP in the aged samples, resulted from GC-MS analysis.

Also, diethylene glycol (10.39 min) can be found in higher concentrations in samples 4% and 6%. Long-chain alkanes are also present in all samples: tetradecane (17.31 min), pentadecane (18.60 min).

From a qualitative point of view, it can be mentioned that the chromatograms of the aged samples with different quantities of PET presented a higher number of peaks compared to the chromatograms of the starting clay-PET samples.

The next test determined the pH value of clayey samples. The test was conducted using the apparatus Mettler Toledo, FiveEasy20, 0 - 14 pH. The pH values can be observed in Table 2. A proportional increase in the pH value was observed, as the amount of PET added for improvement increased.

Table 2. The pH value for the clay and clay – PET samples, after applying the aging cycles

Samples code	Cl	Cl ₂ PET	Cl ₄ PET	Cl ₆ PET
pH value	6.80	6.82	7.28	7.56

To investigate the thermal stability of the PET - clay samples, thermogravimetric analysis was performed.

The TG weight loss curves and first derivative DTG curves for reference PET and the PET from PET-clay matrix samples are presented in Figure 5.

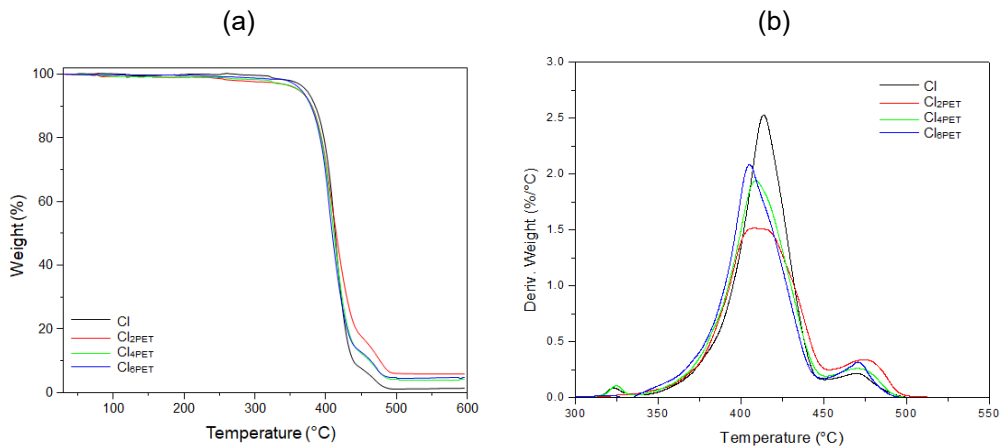


Figure 3. (a) Thermogravimetry (TG) and (b) its derivative (DTG) curves of PET, analysed from matrix clay-PET.

The curves present variations in the thermal stability among the samples. As seen from the figure the samples exhibit a two-step decomposition during TG analysis. The first step of decomposition for reference PET is due to the overlapping of two decomposition processes: the first process (30-430 °C) is mainly due to the degradation of the polymer chain through the end group-initiated mechanism and the second degradation process is due to the thermal degradation of the products formed during first decomposition process [23,24]. The second degradation step for reference PET (P3) in the temperature range 430-550 °C is due to the decomposition of thermally stable species formed during the first degradation step which could be cross-linked carbonaceous structures. In the case of PET-clay samples, the first step of decomposition took place similar to reference PET. The DTG curves revealed that complete thermal degradation occurs in two steps for all samples, with a small difference if compared to pure PET. The common step from 350 °C to 450 °C is attributed to the decomposition and carbonization of PET chains, while the second step for PET over 450 °C might be caused by the decomposition of thermolabile fragments produced in the first step. It can be stated that no significant difference in mass loss, ~ 5%, was observed between reference PET and PET analysed from 2, 4 and 6% PET-clay mixture. The TGA weight loss curves and first derivative TGA curves for reference clay and the clay analysed from the PET-clay mixture are presented in Figure 6.

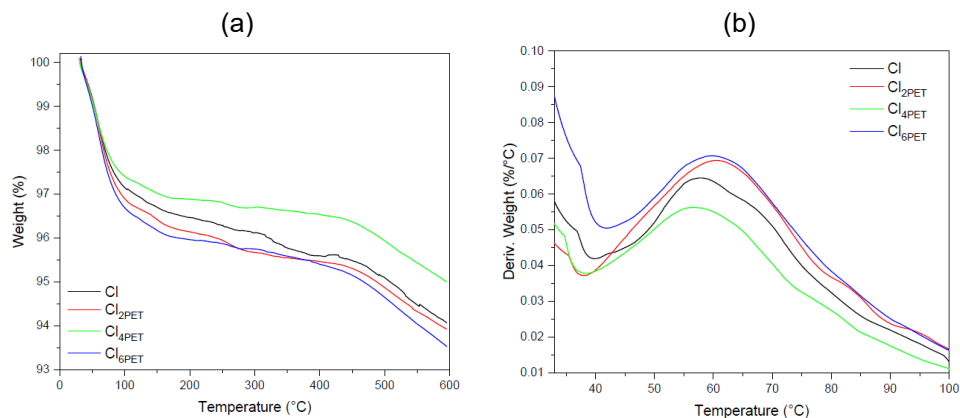


Figure 4. (a) Thermogravimetry (TG) and (b) its derivative (DTG) curves of clay, analysed from matrix clay-PET.

The mass loss of ~ 4 wt% corresponding to surface water can be observed in the temperature range 30-100 °C, and the chemical water mass loss (400-600 °C) was around ~ 2 wt%.

CONCLUSIONS

Improving soil with plastic waste materials is a topic that is increasingly capturing the attention of researchers. This method attempts to solve a part of the plastic waste pollution problem by reusing plastic waste materials in soil improvement.

This study investigated the migration of the chemical components from PET degradation in a clay improved with different percentages.

The clay - PET samples were subjected to variable humidity and temperature conditions in a climatic cabinet, similar to those in situ. GC-MS analysis was carried out on natural state clay and clay improved with 2%, 4%, and 6% PET. The results indicated the presence of DIBP in all four samples analysed. The presence of DIBP increased proportionally with the amount of PET added to the clay. As the percentage of PET added to the clay increased, the quantity of phthalate in the soil also increased, lubricants were also detected in the sample with higher PET quantity.

The thermogravimetric results highlight thermal stability of clay- PET mixture in the normal atmospheric conditions. The thermal degradation of polymeric chain was observed starting from 200 °C.

In conclusion, soil improved with plastic waste materials can be used, from a chemical point of view to increase the geotechnical properties of soil. This is a positive aspect that solves the problem of soils with unsuitable characteristics for foundation. Moreover, this improvement also solves a small part of the plastic waste pollution problem.

EXPERIMENTAL SECTION

The clayey-PET samples were made in the laboratory at the optimum water content (OWC) of 18%. Cylindrical samples with a diameter of 10 cm and a height of 10 cm were collected from the Proctor apparatus. These were introduced into the climatic cabinet (CONTROLS / 10-D1428/A) and subjected to 15 freezing-thawing cycles (-10 °C to +40 °C) in variable humidity conditions (up to 90%).

After the freezing-thawing cycles, approximately 10 grams of soil were collected from the unimproved clay and from each clay-PET sample. The amount of soil was ground and sieved to separate the PET from the soil and then weighed in Erlenmeyer glasses.

Over each sample was added 5 mL of dichloromethane. The samples were left in the ultrasound apparatus for 15 min. The organic solution was gravitationally filtered, and then evaporated in a rotary evaporator under vacuum at a temperature of 35 °C.

The samples were taken up in 200 µL of dichloromethane and were subjected to GC-MS analysis.

GC-MS analysis was carried out with a Shimadzu QP2010 Plus apparatus with an automatic injector AOC-20i, a ZB5MS Plus column, 30m x 0.25mm, 0.25µm, injection temperature – 220 °C, split injection mode, split ratio 20, injection volume 1 µL. The gas flow was 0.8 mL/min (He gas, 6.0), the initial temperature of the column was 50 °C. This temperature was maintained for 10 min, after which the column was heated to 310°C at 20°C/min. The column was left at this temperature for 20 minutes. The temperature of the ion source of the mass spectrometer was 220 °C and the interface temperature was also 220 °C. The analysis time was 43 minutes, the recording being made with the scan method (scanned between 35-800 m/z).

The thermogravimetric measurements were conducted on an SDT Q600 (USA) device from T.A. Instruments. Data on thermogravimetry (TG), and its derivative (DTG) curves, were simultaneously acquired under the following measurement conditions: heating from laboratory temperature to 600 °C, at a heating rate of 10 °C/min, under normal air atmosphere, using alumina crucibles.

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