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Research Article / Araştırma Makalesi

THE EFFECT OF AG NANOPARTICLES ON LEACHATE CHARACTERISTICS FROM AEROBIC LANDFILL BIOREACTORS

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ABSTRACT

Silver (Ag) nanoparticles are being used in many consumer products. Thus, it is likely that these products will last in municipal solid waste landfills at the end of their useful life. There are no adequate data in the literature related with behavior and effects of Ag nanoparticles on aerobic decomposition of municipal solid waste. Accordingly, the aim of this study is to investigate behavior of Ag nanoparticles and their effects on aerobic degradation in aerobic landfill bioreactors. Municipal solid waste with different amounts of Ag nanoparticles (10, 50, 100, 500 and 1000 mg/kg) were disposed in six lab-scale aerobic bioreactors. The experimental study was carried out about 106 days and the chemical and physical properties of leachate samples were analyzed. The results indicated that Ag nanoparticles have not shown a negative impact on the aerobic decomposition of solid waste in aerobic landfill bioreactors.
Keywords: Solid waste, Ag nanoparticles, aerobic landfill, leachate.

AEROBİK BİYOREAKTÖR DÜZENLİ DEPO SAHALARINDA SIZINTI SUYU KARAKTERİSTİĞİ ÜZERİNE AG NANOPARTİKÜLLERİN ETKİLERİ

ÖZ

Gümüş (Ag) nanopartikülleri birçok tüketici ürünlerinde kullanılmaktadır. Faydalı ömürlerinin tükenmesi ile birlikte bu ürünler kentsel katı atıklarla birlikte depolama sahalarında bertaraf edilmektedir. Ag nanopartiküllerinin aerobik depolama sahalarında ve kentsel katı atığın kompostlaştırılması işlemine olan etkileri hakkında literatürde yeterli çalışma bulunmamaktadır. Bu nedenle, bu çalışmada Ag nanopartiküllerinin aerobik depo sahası biyoreaktörlerinde gerçekleşen aerobik parçalanmaya etkileri araştırılmıştır. Kentsel katı atık ve farklı miktarlardaki Ag nanopartikülleri (10, 50, 100, 500 ve 1000 mg/kg) 6 adet laboratuar ölçekli aerobik biyoreaktöre ilave edilerek, çalışma süresince (106 gün) sızıntı suyunda gerçekleşen fiziksel ve kimyasal değişimler incelenmiştir. Sonuçlar, Ag nanopartiküllerinin aerobik depo sahalarında gerçekleşen aerobik parçalanmaya olumsuz bir etkisinin bulunmadığını göstermiştir.
Anahtar Sözcükler: Katı atık, Ag nanopartikül, aerobik depo sahası, sızıntı suyu.

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1. INTRODUCTION

Silver has been used in consumer products because of its antimicrobial effects for over 2000 years. [1]. Technological developments result with the development of engineered silver nanoparticles and the usage areas increased. Silver nanoparticles (AgNPs) have different physicochemical behaviors and characteristics than macro sized silver [1]. Particles in nano size have high surface area to volume ratio and this condition causes more reactivity and in some circumstances higher toxicity [1]. The limited knowledge about this situation raised concerns about potential environmental and human health effects of engineered nanoparticles [2].

Nano silver is used in wide range of industries such as textile, electronic and cosmetic industries. Worldwide production of silver nanoparticles reached to 400 tons per year. [3]. Because of their bactericial properties, near 30% of produced AgNPs are utilized for medical supplies and devices [3]. Nano silver particles released in water from consumer products may be discharged to the sewer system, accumulate in sludge and finally dumped in landfills [4]. It is estimated that 4.77 tons of AgNPs per year will be dumped into landfills [4].Furthermore, to calculate nanoparticles' environmental concentrations, researchers came-up with several analytical techniques, such as probabilistic material flow analysis considering the life-cycle of nanoparticle products [5]. Using various variables, such as production volume, location, particle release with their flow coefficient, researchers have improved a life-cycle perspective model for quantification of nano-silver and measured nano-silver concentration in rivers in the range of 0.002 ng/L- 1.5 ng/L [6]. Sun et al., 2014 [7] obtained the values of 0.66 ng/L in Europe and 0.45 ng/L Switzerland for AgNPs concentration in surface water. AgNPs concentration in surface water and soil in Germany were determined to be 145 ng/L and 27.33 µg/kg, respectively [8]. Approximately 0.04 tons/y of AgNPs are released to the wastewater and 0.01 tons/y of AgNPs in mineral waste with paint is landfilled [9]. This emerging problem should be currently addressed.

There are several concerns about the effects of nanomaterials in landfills. With the growing utilization of AgNPs, the microbial activity in landfills and compost systems may be influenced negatively. Recent studies show that 15% of the total silver in the forms of Ag^+ and/or AgNPs could be occurred from exposure of biocidal plastics and textiles [1]. Other studies indicate that between 34% and 80% of the Ag released from nano textiles are in the form of AgNPs (nanoAgCl particles), nanocomposites and elemental AgNPs[1]. Caballero-Guzman et al. [9] concluded that 0.31 tons/year nano-Ag are transferred to recycling system by consumer electronics. Following products as nano-Ag sources can be ordeded; meditech (0.04 tons/y), textiles (0.03 tons/y), metal coatings (0.02 tons/y) and paints (0.01 tons/y). As a result of these conditions, researchers focus on understanding fate and toxicity of AgNPs in environmental systems.

Landfilling is still the most widely used technology in solid waste management. Because of the advantages as higher degradation and stabilization rate, much better leachate quality aerobic bioreactor landfill applications increased in recent years. The bioreactor landfill provides control and process optimization, primarily through the addition of leachate or other liquid amendments, if necessary. Solid waste stabilization can be accelerated if the landfill is designed and operated as a bioreactor. Also, the bioreactor landfill operation may involve the addition of biosolids and other amendments, temperature control, and nutrient supplementation. Additionally; the concept of aerobic degradation by injecting air into a landfill introduces important advantages in waste management. There have been many studies related with lab, pilot and field-scale aerobic landfill bioreactors, recently [10][11][12][13].

The aim of this study was to evaluate the effects of AgNPs addition on aerobic degradation and leachate characterization in aerobic landfill bioreactors. For this purpose, different amounts of AgNPs were added to bioreactors and mixed with municipal solid waste (MSW). The conventional parameters of leachate samples were monitored during aerobic stabilization of

MSW in order to determine the concentration which can effect the aerobic degradation of municipal solid waste.

2. MATERIALS AND METHODS

In order to investigate the effects of AgNPs on aerobic stabilization process, six laboratory-scale aerobic landfill bioreactors made from 0.6 cm polypropylene were used in this study. Height and thickness of the reactors are 100 cm and 0.5 cm, respectively. The operational conditions of aerobic landfill bioreactors are given in Table 1. .Five reactors were operated with addition of Ag nanoparticles in different amounts, and one reactor was used as a control (AC) reactor. The reactors consist of leachate discharge valves to obtain leachate samples, leachate recirculation system and aeration system to accelerate stabilization, and landfill gas collection pipes to monitor gas components. Temperature probes were placed on the depth of 40 cm from the surface of the reactors. A perforated leachate collection pipe placed at the bottom of each reactor was used for leachate collection. Air was introduced from the bottom of the solid waste and passed through an upward direction by the help of the perforated aeration pipes. The O_2, CO_2 and CH_4 content of the generated gas was regularly measured in order to control the availibity of sufficient O_2 for aerobic degradation. A detailed schematic design of the experimental set-up is depicted in Figure 1.

Operational conditions of the reactors are summarized in Table 1. It can be seen from Table 1 that different amounts of AgNP were added to the reactors with approximately same amount of waste to investigate effect of concentration of AgNP on aerobic degradation. Several researchers [4], [5], [8], [9], [14] stated that AgNPs don't have any negative impact on methane production potential in anaerobic degradation of the solid waste. Considering literature data, higher AgNPs concentrations were studied.

AgNPs were purchased from Sigma-Aldrich (product code: 576832) at anatase form with <100 nm particle size. Disposal of about 30 kg municipal solid waste (MSW) in the bioreactors and addition of different amounts of AgNPs to the reactors were performed in three stages to provide homogenization. At each stage; 10 kg municipal solid waste (MSW) and one third of the determined amount of AgNPs were placed in the reactors and mixed by using a stick.

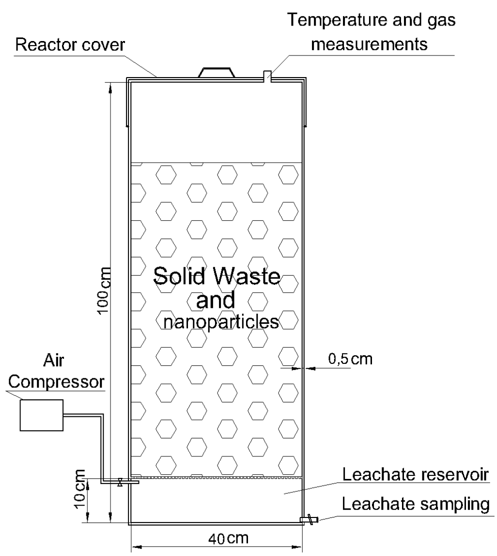

Figure 1. Schematic view of the aerobic bioreactors used in the study

Air compressors were used to provide air for aerobic decomposition of solid waste. Aeration was performed from the bottom of the reactors. Although a wide range of aeration rates for aerobic decomposition have been reported in the literature, the general consensus is that an airflow providing outlet CO_2 concentration about 15% is required. In this study, CH_4, CO_2 and O_2 concentrations within the effluent gas were monitored regularly to determine the air requirement for aerobic decomposition of the solid waste [15]. Gas monitoring analysis was carried out using GeoTech GA2000 Plus Model Landfill Gas Monitoring Device. It was observed that when O_2 content of the output gas decreased below 8%, CH_4 production starts. [16]. To prevent anaerobic conditions, O_2 ratio in the output gas in all bioreactors was kept between 8-14% by aeration.

Table 1. The operation conditions of aerobic landfill bioreactors

Reactors	Characteristics of the reactors	Amount of MSW (kg)	Amount of AgNPs (g)	mg AgNPs/ kg TS
AC	Aerobic control bioreactor	29.16	-	-
Ag10	Aerobic bioreactor +10 mg/kg AgNP	31.73	0.3173	26.33
Ag50	Aerobic bioreactor +50 mg/kg AgNP	30.43	1.5215	131.62
Ag100	Aerobic bioreactor+ 100 mg/kg AgNP	30.23	3.023	263.32
Ag500	Aerobic bioreactor+ 500 mg/kg AgNP	30	15	1315.79
Ag1000	Aerobic bioreactor +1000 mg/kg AgNP	31.37	31.37	2631.71

Solid waste samples were obtained from the entrance of the fermentation unit of Istanbul Compost and Recycling Plant in Istanbul, Turkey. The average composition of MSW generated in Istanbul is 44% organic matter, 8% paper, 6% glass, 6% metals, 5% plastic, 5% textile, 9% nylon, 8% diaper and 9% ash and others[17].

Leachate samples were taken weekly and stored at 4ºC. Leachate samples taken from all of the reactors were characterized for pH, alkalinity, chemical oxygen demand (COD) and chloride following Standard Methods for Examination of Water and Wastewater [18]. First 15 days after disposal no leachate was observed in Ag10 and Ag1000 reactors. Therefore, 1L distilled water was added to these two reactors for 2 times. This promoted leachate generation and accelerated aerobic stabilization.

3. RESULTS AND DISCUSSSON

3.1. Leachate Quality

The variations of pH values in all bioreactors can be seen in Figure 2. The initial pH values were about 7.5-8.8 and these ranges were fluctuated during 80 days in all bioreactors. It is common to encounter initial acidic conditions (pH=4-6) associated with the formation and dissolution of organic acids, whereas higher pH values (7-9) dominate later stages due to the degradation of these acids and the generation of ammonium (NH_4^+) and CO_2 [10], [12]. The pH values in all bioreactors were decreased to 7.5-8.5 range after 80th day and there were no significant changes until the end of operation. According to the results, the presence of Ag nanoparticles with different amounts of aerobic landfill sites did not affect the pH change over time, during aerobic degradation.

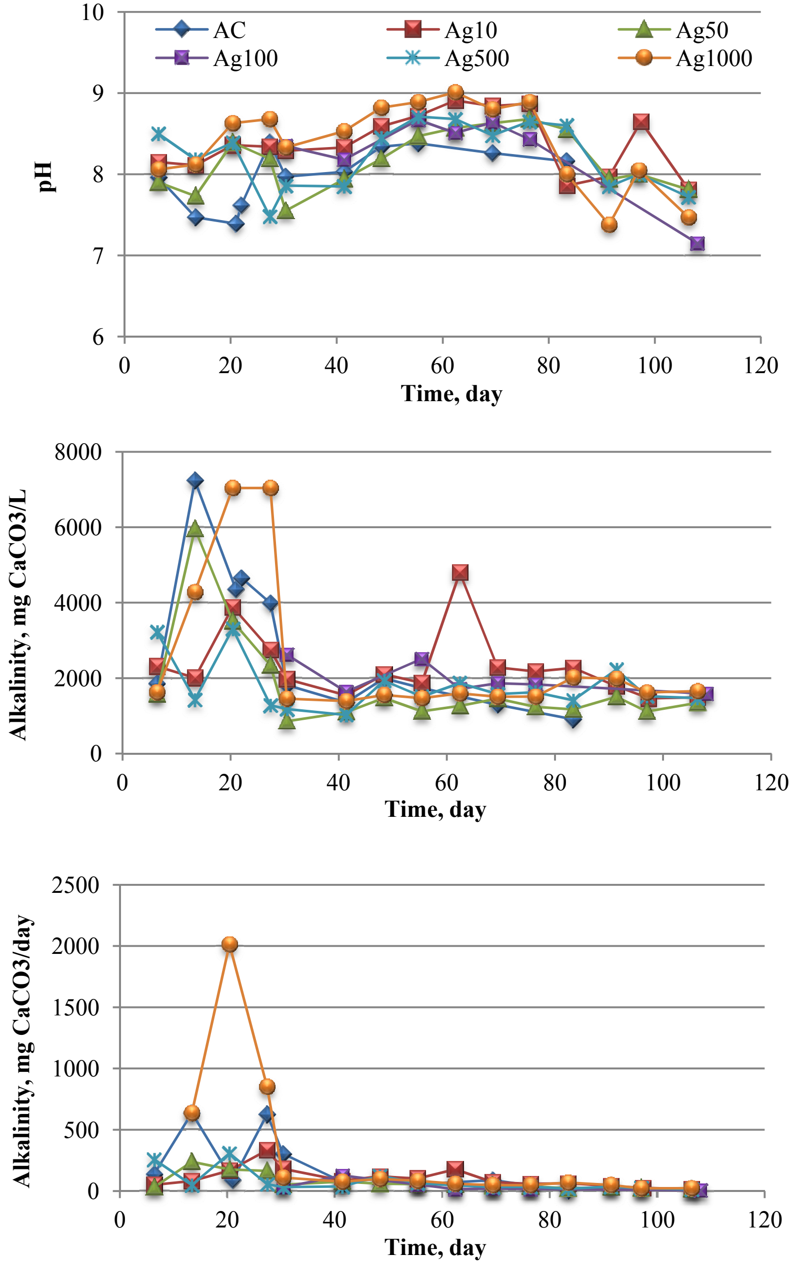

Figure 2. Variations of pH and alkalinity values of leachate in all bioreactors

The alkalinity of water is a parameter of acid neutralization capacity and is due primarily to the salts of weak acids. The changes of alkalinity concentrations in the leachate from bioreactors were given in Figure 2. The initial alkalinity concentrations were 1500-3500 mg $CaCO_3$/L ranges in all bioreactors and remained in this range during operation time. At the end of 106 days, the alkalinity concentrations were measured around 2000 mg $CaCO_3$/L. According to these results, it can be concluded that the amounts of AgNPs has no effect on the variations of alkalinity concentrations in leachate during aerobic degradation.

Chloride is a non-degradable conservative parameter and its concentration change is commonly used to assess the variation of leachate dilution. No observable difference was observed in chloride concentration between acidogenic and methanogenic phases [19], [10]. The chloride is washed out from the landfill via leachate recirculation. Leachate recirculation only, would lead to chloride removal less than that dissolved in the leachate, and thus, leachate chloride value increases. Aeration might also maintain a chloride concentration balance between dissolution and removal [20]. It is observed that as pH increases, the dissolution of chloride increases and thus, the chloride concentration in the leachate increases. This increase is consistant with the findings of the researchers; Manning and Robinson [21] and Bilgili et al. [10]; Sekman et al., [22].

As can be seen in Figure 3, at the beginning of the operation, Cl^-concentrations were determined to be in the range of 2000 – 5000 mg/L in all reactors. After 20 days of operation, Cl^-concentrations began to increase rapidly and reached to values of 4000 – 6500 mg/L after 60 days of operation in all reactors. Cl^-concentrations of AC, Ag10, Ag50, Ag100, Ag500 and Ag1000 reactors were determined to be 5300, 4000, 3500, 4700, 4700 and 3800 mg/L, respectively, after 106 days. After this increase, no considerable change was observed during the rest of the study.

COD is often used to determine the degree of degradation of solid waste. Leachate from the acidic phase at sanitary landfills is characterized by high values of organic pollutants; a large portion of the organic matter consisting of volatile acids.

Figure 4 shows the variations of COD in leachate generated in all bioreactors. As can be seen in Figure 4, the initial COD concentrations were determined to be in the range of 2500-3600 mg/L in all reactors. COD concentrations of the leachate increased to 6000-10500 mg/L values in the reactors, at day 20 and then began to decrease rapidly. At the end of the experimental period on day 106, COD values in the AC, Ag10, Ag50, Ag100, Ag500 and Ag1000 reactors were found to be 2800, 2200, 1600, 2900, 2600 and 1200 mg/L, respectively.

Bilgili et al.[16] concluded that COD values decreased to 6500 and 17,000 mg/L in two pilot-scale aerobic landfill reactors with (A1) and without (A2) leachate recirculation, respectively on day 120 and it was observed that COD values were 5000 and 8000 mg/L, respectively at the end of the test period on day 250. The findings in this study were found to be consistent with the results obtained by Bilgili et al. [16]. It can be said that different amounts of AgNPs in MSW have no effect on the rate of solid waste degradation in aerobic landfills.

Variations of COD/CODmax ratio showing the aerobic decomposition rate were determined to be similar in six bioreactors (Figure 5). As can be seen from Figure 5, the addition of AgNPs have no negative impact on aerobic decomposition in bioreactors. These results are consistent with the findings of Gitipour, et al. [1] who investigated the effects of AgNPs on composting process.

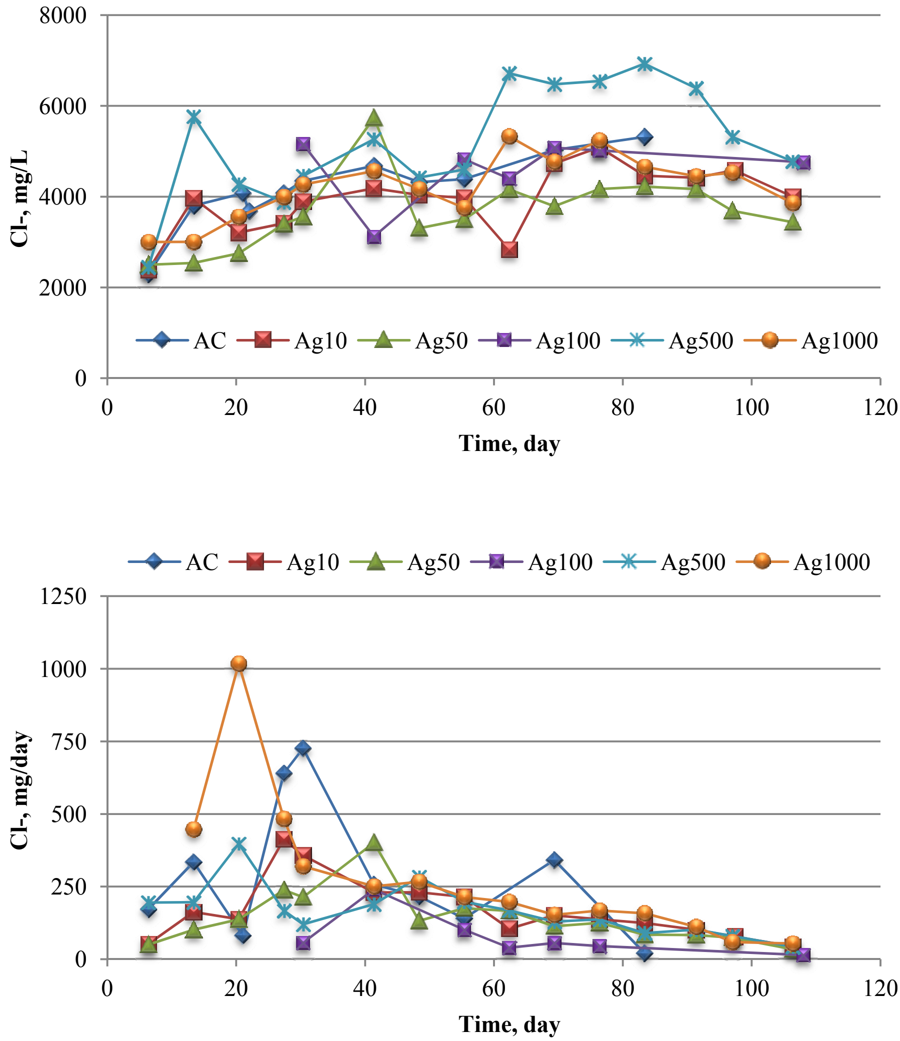

Figure 3. Variations of chloride values of leachate in all bioreactors

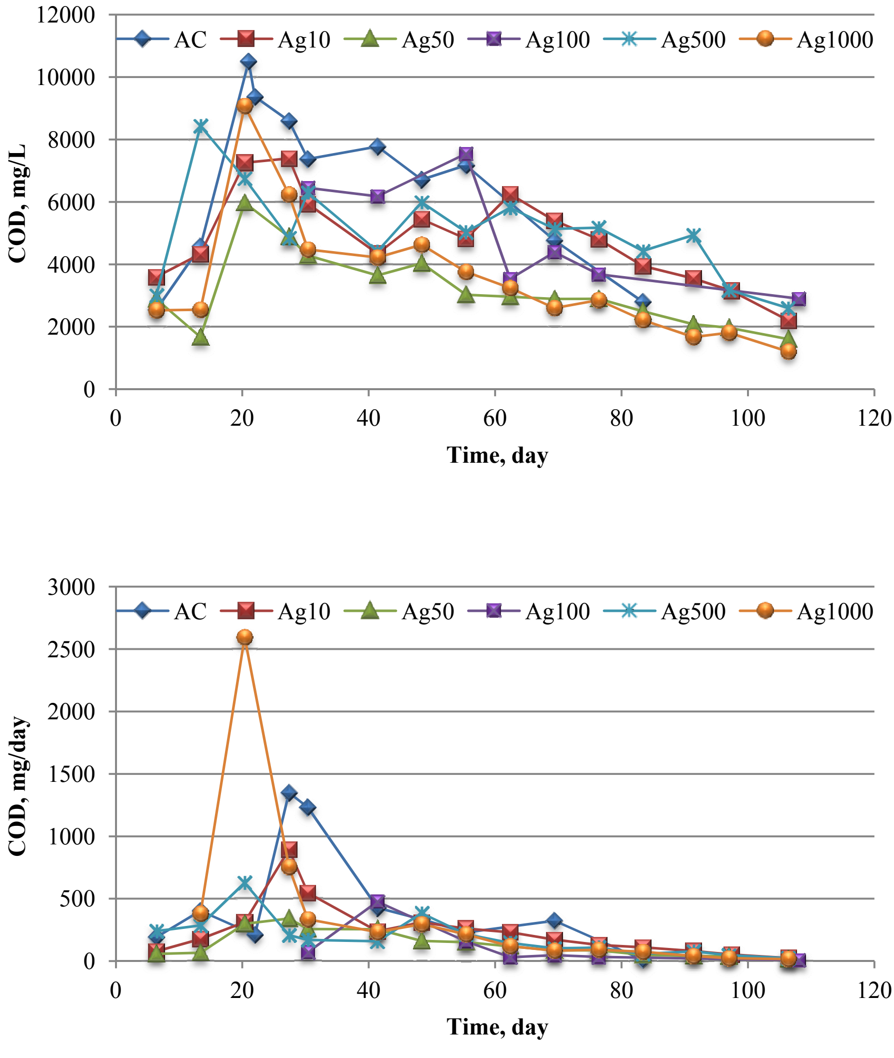

Figure 4. Variations of COD values of leachate in all bioreactors

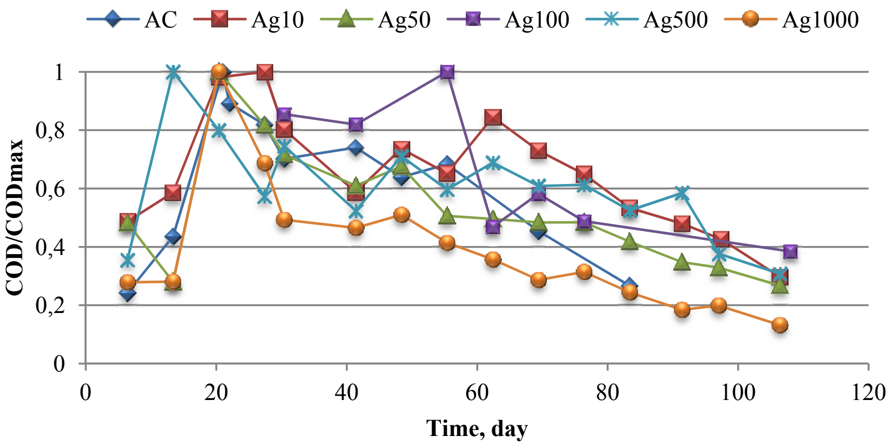

Figure 5. Variations of COD/COD_{max} values of leachate in all bioreactors

4. CONCLUSION

In this study, effect of AgNPs on leachate characteristics in aerobic landfill bioreactors was investigated. Results show that aeration and leachate recirculation not only increased the rate of biodegradation of municipal solid waste in landfills but also improved leachate quality. It was observed that AgNP have no significant effect on aerobic solid waste degradation.

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