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Research article

Greywater as a sustainable water source: A photocatalytic treatment technology under artificial and solar illumination

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ABSTRACT

Greywater considers being a highly reclaimable water source particularly important for water-stressed nations. In this work, heterogeneous photocatalysis using artificial and solar illumination has been applied for the mineralization of simulated light greywater (effluents from dishwashers and kitchen sinks were excluded from the study). The effects on the process' efficiency of TiO_2 P25 catalyst's concentration, initial concentration of H_2O_2 and Fe^{3+}, pH of the solution, as well as the type of radiation, were evaluated in a bench-scale Pyrex reactor and a pilot-scale slurry fountain photoreactor. The treatment efficiency has been followed through the evolution of the organic matter content expresses as dissolved organic carbon (DOC). Best results were obtained with the photo-Fenton-assisted TiO_2 photocatalytic process with 72% DOC removal after 210 min of bench scale treatment, while under the same photocatalytic conditions in the pilot reactor the DOC removal reached almost 64%. Moreover, the decrease in toxicity, phytotoxicity and biodegradability of the simulated wastewater has been observed after solar-induced photocatalytic treatment.

1. Introduction

Given the worldwide water scarcity and future demands, the debate on the utilization of recycled wastewater as an alternative water source is gaining increasing attention. One major source of highly reclaimable water, particularly important for water-stressed nations, is the urban wastewater generated from washing activities in households which is referred to as "greywater". Bearing in mind that greywater generally has a lower organic load and pathogen content than municipal wastewater which includes additionally the respective streams from toilets and kitchen sinks, it may be considered as an ideal candidate for decentralized treatment and reuse systems.

Greywater (GW) is defined as domestic wastewater originating from kitchen, bath and laundry excluding effluent from toilets, commonly referred to as "black water" (BW) (Friedler and Hadari, 2006; Gross, 2008; NSW, 2008; WHO, 2006). For wastewater deriving from showers, tubs, hand basins and clothes washing machines the term "light greywater" is often used, whereas GW including also effluents from dishwashers and kitchen sinks is referred to as "dark greywater" (Birks and Hills, 2007; Friedler and Hadari, 2006). The quantity and quality of greywater is determined by various factors; i.e. the social and cultural behavior, lifestyle, age distribution and living standards of the inhabitants, availability of water and its consumption as well as the technical framework by which people are surrounded. Greywater accounts for up to 75% of the wastewater volume produced by households (Hernandez-Leal et al., 2011).

The chemical constituents of GW are typically classified as inorganic and organic. Inorganic constituents include dissolved matter, nutrients, non-metallic elements, metals and gases (Table S1 Supplementary Material), whereas, organic constituents may be sub-classified as aggregate and individual (Takashi et al., 2007); aggregate constituents are used to characterize the bulk of the organic matter in wastewater and include parameters such as biochemical oxygen demand (BOD), chemical oxygen demand (COD), total organic carbon (TOC), total dissolved solids (TDS) and total suspended solids (TSS). On the other hand, the identification of individual constituents in greywater is limited by a significant

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knowledge gap in research (Ledin et al., 2006). About a decade earlier, Eriksson et al (Eriksson et al., 2002). identified as many as 900 xenobiotic organic compounds and compound groups (XOCs), which are commonly used in bathroom and laundry products and hence potentially present in greywater. Subsequent screening of bathroom greywater from a Danish apartment block confirmed the presence of almost 200 such XOCs, including surfactants, fragrances, preservatives, antioxidants, plasticizers, UV filters and solvents, many of which are endocrine disruptors (Eriksson et al., 2003). Many of these XOCs were still present after biological treatment which underlines the necessity for more advanced treatment options (Leal et al., 2010).

To understand the chemical characteristics of any type of wastewater it is important to bear knowledge concerning the sources of its chemical ingredients. Shower and hand basin GW contains soaps, shampoos, toothpaste, body care products, shaving waste, skin, hair, body fats, lint and traces of urine and feces; washing machine GW contains high concentrations of chemicals from detergent powders and emulsions (such as sodium, phosphorous, surfactants and nitrogen), bleaches, oils, paints, solvents and non-biodegradable fibers from clothing (Morel and Diener, 2006; Noah, 2002). Greywater originating from kitchen sinks and dishwashers is characterized by high pH values, salinity, turbidity, suspended solids, hot water, foam and odor, due to food residues, high amounts of oil and grease, dishwashing detergents and bacteria (Government, 2008; Morel and Diener, 2006; Noah, 2002). It can be easily understood that the characteristics of GW are highly influenced by lifestyle, social and cultural behavior of the residents, availability of water and its consumption. GW is therefore highly variable in its organic content (Jefferson et al., 2004).

Several remarkable attempts have been conducted on the treatment of GW with different technologies which vary in both complexity and performance (Ghaitidak and Yadav, 2013; Li et al., 2009), such as solar photocatalytic degradation with powdered activated carbon (Farre et al., 2008) or using TiO_2-coated textile fibers coupled flocculation with chitosan (Grčić et al., 2015), with Hydrogen Peroxide Plus (Gonzalez et al., 2008) and UVC/H_2O_2 (Chin et al., 2009).

Among the so-called Advanced Oxidation Processes (AOPs), photocatalytic oxidation has shown great promise in the treatment of wastewater, since it could be suitable to remove recalcitrant organic compounds (Gaya and Abdullah, 2008) and able to achieve the disinfection of wastewater (Paleologou et al., 2007). The photocatalytic decomposition of organic compounds of environmental concern (e.g. UV filters, detergents, fragrances, etc.) has been studied extensively during the last 25 years and it has been demonstrated that it can be an alternative to conventional methods for the removal of organic pollutants from water (Klavarioti et al., 2009; Malato et al., 2009; Petrovic et al., 2003; Tsoumachidou et al., 2016). Additionally, an advantage of the photocatalytic process is its mild operating conditions and the fact that it can be powered by sunlight, thus reducing significantly the electric power required and, therefore, operating costs.

Photocatalytic treatment with subsequent recycle and reuse constitutes a very attractive alternative from an environmentally sustainable perspective, as it offers the potential to substantially reduce domestic water demand in water scarce regions. The aim of this study was to evaluate the mineralization of simulated greywater (SGW) by TiO_2-induced heterogeneous photocatalytic process; bench-scale experiments were conducted under artificial UV-A illumination in order to assess the effect of various operating conditions on SGW treatment. Pilot-scale reactor was employed to assess the applicability of the proposed process under natural solar irradiation; the decrease in toxicity, phytotoxicity and biodegradability of the simulated wastewater has been observed after solar-induced photocatalytic treatment.

2. Experimental and analytical

2.1. Reagents

TiO_2 Evonik P25 catalyst (TiO_2 P25, 70% anatase and 30% rutile with BET surface area of 55 ± 15 m^2 g^{-1}) was employed in this study. Iron chloride ($FeCl_3 \cdot 6H_2O$, Alfa Aesar), hydrogen peroxide (H_2O_2, 30% w/v, Panreac Quimica), sodium peroxodisulfate ($Na_2S_2O_8$, Chem-Lab), titanium (IV) oxysulfate-sulfuric acid solution ($O_5STi \cdot xH_2SO_4$, 27–31% H_2SO_4 basis, Fluka) were used as received. NaOH and H_2SO_4 solutions were used for pH adjustment when necessary.

2.2. Greywater

The significant variance of both the qualitative and quantitative characteristics of greywater owed to regional, societal and also temporal profiles of the household end users in the primary reason that although greywater constitutes a highly reclaimable source of water, its reuse is nonetheless met with high skepticism (Eriksson et al., 2002; Jung et al., 2015; Li et al., 2009).

In the present study, the preparation strategy of a simulated greywater effluent was based on (i) the qualitative characteristics of real greywater discharges taken directly from Greek households, (ii) the identification and selection of individual commercial products found in supermarkets for average use in laundries, showers and sinks, and (iii) the quantitative characteristics of greywater individual sources that have been reported in previous studies (Friedler, 2004; Jamrah et al., 2006, 2008; Jefferson et al., 2004; Kotut et al., 2011; Prathapar et al., 2005).

In this context, raw greywater (RGW) samples were collected during the summer period from different bathroom sources and households. The sampling campaign was performed in order to characterize the quality of various greywater streams discharged from different sources (shower, hand basin and washing machine as shown in Table S2 of Supplementary Material). Bathtub as a source was excluded from the study as it is highly uncommon in Mediterranean countries to fill it up and bathe in it; instead, the tub - when available - is used to shower. Immediately after sampling, the containers were transported to the laboratory in cooling bags and microbiological analysis was carried out within 1 h, the remaining samples were kept refrigerated (at 4 °C) or frozen (at −18 °C) for a maximum of one month depending on the parameter to be analyzed for. In total, 16 samples were analyzed in regards to various qualitative parameters: pH, electrical conductivity (EC), cations (i.e. sodium, ammonium, potassium, calcium, magnesium), anions (i.e. fluoride, chloride, nitrite, nitrate, bromide, phosphate, sulfate), total solids (TS), chemical oxygen demand (COD), biochemical oxygen demand (BOD), total organic carbon (TOC) and common microbiological quality parameters (i.e. Total coliforms, *Escherichia coli* and Faecal enterococci).

Because of the extremely high variability in RGW samples it was deemed necessary to prepare a reproducible effluent that would simulate the actual wastewater; in this context, representative commercial personal care products (PCPs) were selected and specifically a laundry detergent, fabric softener, shampoo, hair conditioner, shower gel, liquid hand wash and a face cleansing gel. Actual commercial products were used so that the SGW produced would be similar in complexity compared to actual effluents; besides the active ingredients (e.g. detergents) RGW effluents also contain a vast variety of auxiliary chemicals and constituents (e.g. emulsifiers, coloring agents, preservatives etc.) that affect the behavior of the effluent during its treatment. As a result, a mixture of all