Solar Concentration for Wastewaters Remediation

Subjects: Environmental Sciences | Water Resources
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As the effectiveness of conventional wastewater treatment processes is increasingly challenged by the growth of industrial activities, a demand for low-cost and low-impact treatments is emerging. A possible solution is represented by systems coupling solar concentration technology with advanced oxidation processes (AOP).

Keywords: solar treatment ; wastewater treatment ; photo-Fenton reaction ; photocatalysis ; sunlight concentration ; advanced oxidation processes ; solar collector ; water pollutants

1. Introduction

The demand for clean water sources has been rapidly increasing in recent decades, led by industrialization, the expansion of agriculture and, especially, population growth. Access to safe water supplies has thus become an issue of global significance [1]. Moreover, the health risk associated with polluted water resources is projected to become a major global issue within the next few decades [2]. Among the various practical strategies and solutions proposed for more sustainable water management, wastewater reuse and recycling stands out as the most economically-viable and environmentally-friendly [3].

Presently, the most common wastewater treatments are based upon a combination of mechanical, biological, physical and chemical processes such as filtration, flocculation, chemical or biological oxidation of organic pollutants.

A common problem for current technologies is poorly biodegradable organic pollutants, the so-called bio-recalcitrant organic compounds (BROCs). A class of treatments capable of tackling BROCs is known as advanced oxidation processes (AOPs), relying on the formation of highly-reactive transient (e.g., superoxide, peroxide, hydroxyl radical) chemical species that can convert BROCs into more biodegradable compounds or, ideally, into inorganic carbon [4]. Among the most efficient AOPs are those based on hydroxyl radicals $OH^•$, a powerful oxidant. These methods are generally based on the dissociation of hydrogen peroxide in water, either by direct absorption of ultraviolet (UV) photons or by mediation with metal ions (Fe and Co are among the most studied) through Fenton and photo-Fenton reactions [5][6][7]. In addition, and equally or even more important, is the possibility of generating $OH^•$ radicals by the interaction, in water, of artificial or natural light with semi-conductors. This process is commonly described as photocatalysis [8].

In recent decades, the photocatalytic degradation of several organic compounds has received growing attention as a water purification process. Irradiating semi-conductors, for example TiO_2, in the form of micro- or nano-sized particles, on fixed supports or in aqueous suspensions, creates a redox environment that is reactive towards most organic species [9]. A number of reports have shown that surfactants, pesticides and dyes can be effectively converted into less dangerous products like carbon dioxide and hydrochloric acid. In 1998, the United States Environmental Protecting Agency (EPA) published a detailed list of molecules capable of being degraded by AOPs [10].

In photocatalysis, photons can be seen as reactants and/or co-catalysts, hence playing a critical role in the process. On this basis, considerable research effort has been made since the 90s to employ solar radiation as an abundant, renewable and potentially zero-cost light source. In this approach, solar photons are collected and directed into a photoreactor where they power catalytic reactions. Traditionally, solar collector systems are broadly classified as concentrating or non-concentrating, according to their concentration factor or to the temperature which can be reached by the system [11].

Combining wide versatility, potentially very low cost and the capability of total conversion to non-toxic products, the use of solar-powered AOPs for the treatment of urban and industrial wastewaters is likely the most promising application of AOP technology [12][13].

The application of solar disinfection (SODIS) has also been recently demonstrated on real wastewater samples with the possibility of integrating SODIS technology to an urban wastewater treatment plant (WWTP) [14][15]. Nanofiltration in combination with tertiary processes, including solar photocatalysis, is also the object of considerable research interest; however, it has presented uncertain results [16][17]. Another more material-oriented approach, albeit one that is still at an early stage in terms of literature reports, suggests the use of a combination between a photocatalyst and adsorbents. It is termed "integrated photocatalyst adsorbent" (IPCA), and is based on an adsorbent which has the ability to degrade organic matter in the presence of sunlight [18].

2. Solar Collector Systems Employed for Wastewater Treatment

The starting point for the use of solar collectors in wastewater remediation can be traced back to parabolic systems, originally developed for thermal energy applications, and then adapted in 1989 in Albuquerque, NM, USA for water purification. Immediately afterwards, in 1990, a dedicated facility started operations at the *Plataforma Solar de Almeria*—Spain. Ten years later, dedicated research on wastewater treatments started to be effectively performed [19]. Solar collectors can be concentrating or non-concentrating systems. The former can be classified depending on the principle adopted for focusing sunlight and based on whether they use a fixed or moving receiver [20], while the latter generally consist of flat panels which can be fixed or movable, i.e., following the sun. While in thermal solar applications all wavelengths of the sunlight spectrum are concentrated onto an absorber to produce an increase in temperature, for a solar AOP, the most effective photons are those on the high-energy side of the spectrum, in the UV or near UV range (300–400 nm wavelength), due to the prevalent use of wide band-gap semiconductors as catalyst materials. Wavelengths up to 600 nm are, to date, effectively exploited only by photo-Fenton reactions or by emerging catalyst materials designed ad-hoc for visible light absorption [21]. This is therefore an opportunity and a challenge for the materials science field. The use of light at wavelengths higher than 400 nm would also allow the use of silvered mirrors, which are simple and robust, but which present poor UV reflectance.

In fact, given that, to date, the range between 300 and 400 nm has been considered to be of exceptional importance, conventional silver mirrors have not been considered suitable as reflectors. This is mainly due to their low average reflectance in this wavelength interval, with a minimum at around 320 nm related to an interband transition [22]. Furthermore, the glass covering commonly present on silver mirrors contains iron impurities, which further contribute to reductions in UV reflected radiation.

Mirrors based on aluminum are generally considered a better option when working with processes requiring UV photons [23]. In fact, their reflectance is high (around 93%) and almost constant in the 300–400 nm range. As proposed by Kutscher and Wendelin [24][25], the best reflective surface for solar AOPs should be: (i) efficient in the UV range, (ii) weather resistant, (iii) reasonably cheap. At present, the benchmark solution is based on anodized and electro-polished aluminum surfaces. An available alternative is using aluminum mirrors protected with an acrylic coating [26].

The design of a solar AOP system requires consideration of a number of factors: the mirror materials and shaping, the catalyst, wastewater loading method (batch or once-through), flow type and rates, pressure drops, eventual pretreatment, eventual oxidant loading method, pH control and the use of a tracking system to enhance the direct solar radiation [21]. These criteria are presented in Table 1.

Table 1. Design factors for a solar wastewater remediation AOP.

Component	Feature
Mirror design	Parabolic/Dish/Plane
Tracking system	Automatic/Manual/Fixed
Catalyst	Dissolved/Suspended/Supported
Reactor configuration	Single/Parallel/Series
Wastewater loading	Once-through/Batch
Flow rate	Volume per time/On-Off

Component	Feature
System pressure	Pumping employed
Pretreatment	Present/Absent
Oxidant loading method	Once/Periodically dosed
PH control	Acidic/Neutral/Basic

Most existing literature reports are based on different combinations of these factors, giving rise to a variety of configurations [13][19][21].

3. Advanced Oxidation Processes and Photocatalysis

AOPs are commonly defined by the chemistry and chemical engineering community as water treatments aimed at the removal of pollutants via oxidation by highly-reactive radicals, such as the hydroxyl ($OH^•$) or others (e.g., superoxide, peroxide, sulphate). As we are interested here in processes which can be activated by sunlight, we will limit this review to those involving photocatalysis. In wastewater remediation, the most common process employs hydrogen peroxide (H_2O_2) as the hydroxyl radicals source, activated by UV light. Although oxidation reactions by $OH^•$ have been known for more than a century, with the use of Fenton's reagent in analytical chemistry, the application to wastewater remediation was only considered when evidence of $OH^•$ generation in "sufficient quantity to carry out water purification" was given in the late 1980s [27]. Even though AOPs are of special interest for several applications, among which aromatics and pesticides degradation in water purification processes [28], oil derivatives, and volatile compounds [29], they have not yet been employed for large-scale commercial use. AOPs offer important advantages in water remediation, such as the possibility to effectively eliminate organic compounds in the aqueous phase, rather than collecting or transferring pollutants into another phase. Indeed, the contaminants can, in principle, be converted by complete oxidation into inorganic compounds, such as carbon dioxide; this process is called mineralization. Also, $OH^•$ can, in principle, react with almost every organic aqueous pollutant without discriminating, potentially targeting a wide variety of compounds. A notable exception is that of Perfluorinated compounds, which are not attacked by $OH^•$ radicals due to the stability of the C-F bond [30]. Some heavy metals can also be precipitated as $M(OH)_x$ [31]. AOPs currently have a number of serious drawbacks, affecting cost and limiting their large-scale application. For example, a constant input of reagents is usually necessary to keep an AOP operational, because the quantity of hydroxyl radicals in solution needs to be high enough to react with all the target pollutants at useful rates. Scavenging processes can occur in the presence of species which react with OH radicals without leading to degradation, for example, bicarbonate ions (HCO_3^-) [32] and chlorides [33] should be removed by a pre-treatment or the AOPs will be compromised. In particular, the interference role of bicarbonate, chloride and dissolved silica anions has been recently studied in depth [34][35][36]. Finally, most existing processes rely on TiO_2 as the catalyst material. Although it offers important advantages in terms of stability, robustness and catalytic activity, it also has the serious limitation of requiring activation by UV light [37]. As such, it is mostly employed with artificial UV-sources, which are expensive, have short operational lifetimes, and require a high energy input.

For these reasons, it is not economically reasonable to use only AOPs to treat large amounts of wastewater, but AOPs can be conveniently integrated with conventional treatments as a final or intermediate step [38]. In this context, the use of solar radiation to promote AOPs could contribute to greatly reducing costs by relying on a free and renewable source of photons, even if the cost problems mentioned above must be mitigated in order to have an efficient and competitive water purification technology. This possibility, along with increasing efforts towards the implementation of water reuse worldwide, are currently accelerating research towards the implementation of large-scale AOPs [4].

4. Wastewaters

Since investigations dedicated to water purification with the use of sunlight began, several types of pollutants have been investigated. These have generally been model compounds, while real wastewaters, due to their greater complexity, are still less explored.

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