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Effect of water matrices on the photochemical degradation of orange G in aqueous solution

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Abstract

In this study, we assessed the effect of aqueous matrices nature in the degradation of an azo dye Orange G (OG), using a UV Process at a wavelength of 254 nm. First of all, the effect of initial dye concentration was investigated and found to follow pseudo-first order kinetic. The apparent reaction rate constant (k_{app}) was decreased from 0.622 to 0.034 min^{-1} with increasing initial OG concentration from 2 to 50 mg/L respectively. The implication of hydroxyl radical ($HO^{\bullet}$) in the UV-C system was demonstrated using tert-butyl alcohol as a hydroxyl radical scavenger. Then, the effects of bicarbonate (HCO_3^-) carbonate (CO_3^{2-}) and bromide (Br^-) ions on the efficiency of the photodegradation of OG (50 mg/L) were investigated. It was found that the presence of these ions, especially at high concentration ($[HCO_3^-]$ = 1000 mg/L, $[CO_3^{2-}]$ = 1000 mg/L and $[Br^-]$ = 3000 mg/L) inhibited the degradation process through consuming $HO^{\bullet}$, despite the formation of other radicals (carbonate and bromide radicals) which are probably less reactive than $HO^{\bullet}$ towards OG. Finally, five types of water: ultra-pure water, tap water, mineral bottled water, Mediterranean Sea water and Dead Sea water, were used as target matrices for the photodegradation process and compared with that used distilled water, the removal efficiency was as follow: 99.8%, 84.5%, 64.4%, 57.4% and 17% respectively.

Keywords: Azo dye; Orange G; carbonate; bromide; advanced oxidation process.

1. Introduction

The decrease of fresh water and the deteriorating water quality caused by the wastewater released from different industries are two of the most current environmental issues in global scale [1]. Indeed, the biological or chemical water pollution has been an alarming increasing in the last 50 years due to the intensification and diversification of industries [2]. Heightened concerns over public health and associated environmental hazards due to the presence of toxic organic compounds such as dyes [3] which during the dyeing processes, a portion of them remains unfixed and gets washed out [4]. Due to their synthetic nature and structure mainly aromatic, these unfixed dyes are non-biodegradable, having carcinogenic action or causing allergies and skin irritation [5]. Orange G (OG) is a typical example of azo dyes, widely used in textile, pulp and paper industries [6]. It is reported to show some chromosomal damage and clastogenic activity as special toxic effects [7]. Thus, its widespread utilization in different domains can cause a serious environmental problem due to its obvious and latent danger for humans and ecosystem [8]. The removal of colouring materials like pigments and dyes from the industrial waste water is a prime concern that attracted many researchers [9]. Many methods including chemical oxidation, coagulation and adsorption process, biological process are used to reduce the toxic dye effluents from wastewater, but the main drawback of these techniques is formation of secondary waste product that required further treatment, or they cannot be individually enough to remove completely dye from wastewater [10, 11]. Thus, an ideal dye removal method should be able to efficiently remove large quantities of dye from wastewater in a short time span and without secondary pollution [12]. In this context, Advanced Oxidation Processes (AOPs) (i.e. photocatalysis [13], UV/H_2O_2 [14], UV/O_3 [15]) have emerged as a set of oxidative water treatment that is used to treat toxic effluents. The efficiency of these techniques resides in the generation of high concentration of hydroxyl radical ($HO^{\bullet}$)[16], which is the most powerful and unselective chemical oxidant so far known. However, a drawback resulting from such a high reactivity is that the hydroxyl radical can also react with “non-target” pollutants in solution which are referred to as radical “scavengers” [17]. The presence of bicarbonate (HCO_3^-) and carbonate (CO_3^{2-}) ions is very common in most aqueous coloured solutions as well as in natural water [18]. Although less often related in the literature, bromide ions (Br^-) is naturally present in raw water, groundwater and surface water, and with the rapid growth of industrial activities, more bromide is discharged to environment [19]. Since these ions are known to react with the HO$^•$ radical [20], their coexisting with the target contaminant expected to affect the efficiency of the treatment. In the literature, researches involving the effect of common ions on the OG removal by different AOPs are reported [21-26], but to the best of our knowledge, no work was involving the influence of bromide ions. Also, because the influence of water matrices on the photo-degradation of OG can be representative of azo dyes, which can be degraded by HO$^•$ radical- induced AOPs, it was of considerable practical interest to examine the photochemical treatment in different water matrices such as mineral water, tap water, ultrapure water and sea water. Hence, the aim of this work was to investigate the effect of different ions on the performance of the degradation of OG by UV-C photolysis as a model AOP treatment technology, in a laboratory-scale photoreactor. A number of assays were carried out to assess the extent of the target pollutant photolysis in water with addition of inorganic salts. The salts tested are as a forementioned, dyeing auxiliaries or are naturally present in water (carbonate, bicarbonate and bromide ions). The amount of different salts was varied over a wide range, in order to evaluate the effect of higher concentrations on the photodegradation process. Finally, the photochemical removal of the contaminant in several types of water was investigated.

2. Material and Method

2.1. Chemicals

Orange G (abbreviation: OG; chemical class: azoic; molecular formula: $C_{16}H_{10}N_2Na_2O_7S_2$; molecular weight: 452.37 g/mol was purchased from RAL Diagnostics. It is highly water soluble (50 g/L at 20°C), and was used without further purification. The molecular structure of OG is shown in Fig.1. Sodium carbonate (Na_2CO_3, purity 99%) was purchased from Sigma-Aldrich. Sodium bicarbonate ($NaHCO_3$, purity 99%) was procured from Prolabo. Potassium bromide (KBr, purity 99.5%) and Tert-butyl alcohol (C_4H_9OH) were purchased from Aldrich. All solutions were prepared with distilled water (DW). Some environmental waters were used as aqueous matrices in other experimental series. Ultra-pure water (UPW) was produced by Nature Company Ltd. Jordan. Bottled mineral water (BW) was purchased at a local supermarket. Tap water (TW) was obtained from the local drinking supply. Sea waters were collected from the Mediterranean Sea (MSW) (Algeria) and the Dead Sea (DSW) (Jordan). The composition of different matrices is shown in Table 1.

Table 1: The composition of the different types of matrices

Matrices	HCO_3^- (mg/L)	CO_3^{2-} (mg/L)	Br^- (mg/L)	Salinity (g/L)
DW	0	0	0	0
UPW	0	0	0	0
BW	265	< 0.01	0	2.078
TW	341.6	/	0.86	0.21
MSW	140	10	65	35
DSW	220	240	5000	270

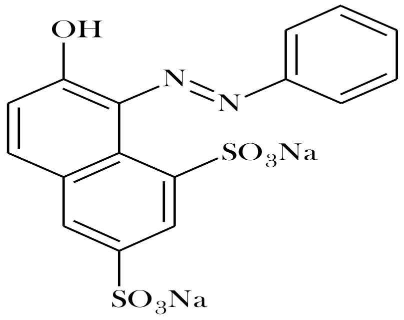

Figure 1. Structure of azo dye Orange G (OG)

2.2. Reactor

Experiments were conducted in a cylindrical water-jacketed glass reactor. A 25 W low-pressure mercury UV lamp (300 mm long with 18 mm diameter) with a maximum emission wavelength of 253.7 nm and a lower emission wavelength of 184.9 nm, was immersed in axial position inside the reactor (2/3 of the solution). The temperature of the solution was monitored using a thermocouple immersed in the reacting medium. To obtain homogeneous mixture, various aqueous solutions of OG were prepared by adding the specific amount of additives under continuous stirring using a magnetic stirrer. Samples of dye solution were removed from the vessel via a sample port. The set-up used is shown in Fig.2. The temperature inside the reactor was regulated at 20±2°C by circulating cooling water within the jacket surrounding the cell.

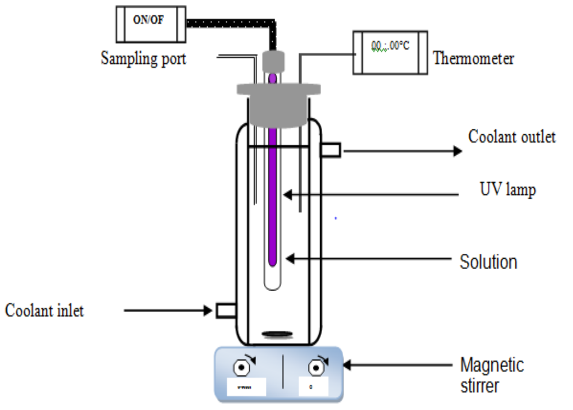

Figure 2. Experimental set-up for photochemical process

2.3. Procedure

The initial concentration of OG was 50 mg/L for all experiments, except for those carried out to examine the effect of initial dye concentration. Various aqueous solutions of OG (50 mg/L) were prepared by adding the required amount of additives under continuous stirring using a magnetic stirrer.

The decolourization of OG was carried out at 254 nm under different conditions using constant solution volume of 150 ml. Aqueous samples were withdrawn periodically and the concentrations of substrate were determined using a UV–vis spectrophotometer (SHIMADZU.UV-1605) at 478 nm. The percentage (%) dye removal is reported as:

$$\%\, dye\ removal = \frac{C_0 - C_t}{C_0} \times 100 \qquad (1)$$

Where C_0 and C_t represent dye concentration at the initial time and the time (t) respectively.

Experiments were conducted at natural pH without further adjustment.

3. Results

3.1. Effect of initial dye concentration

Degradation experiments in UV-C system at 254 nm were carried out at natural pH for 2, 10, 20, 30 and 50 mg/L of OG initial concentrations. Fig. 3. shows the normalized concentration profiles versus time. As can be observed, the removal efficiency of 100% was obtained for all the tested dye concentrations expected for 50 mg/L which was 99%. However, the time required for total elimination depends on the initial concentration. Thus, for a concentration of 2 mg/L a total elimination was reached after only 10 minutes of irradiation, while 40 minutes were necessary to remove 20 mg/L of OG. The plot of ln C/C_0 versus time (insert Fig.3.) shows a linear relationship, indicating that the OG degradation follows the first-order kinetic model below:

$$C = C_0 \exp(k_{app} t) \qquad (2)$$

Where C is the concentration of OG at time t ; C_0 is the initial concentration of OG; k_{app} the apparent reaction rate constant.

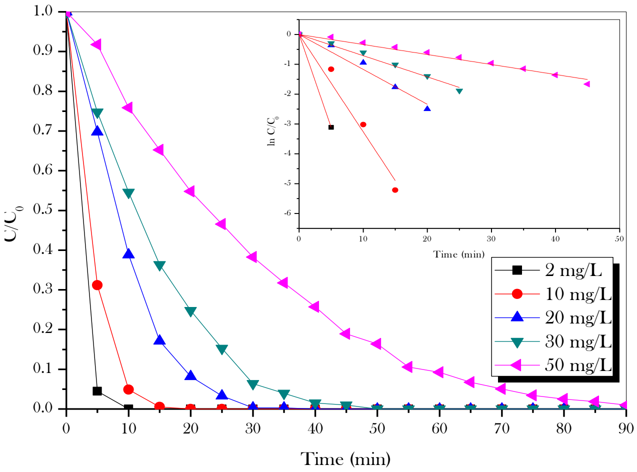

Figure 3. Effect of initial dye concentration on the color degradation of OG; (Fig.insert) Pseudo-first order model

The values of the apparent reaction rate constant (k_{app}) and the dye degradation half-time ($t_{1/2}$) are recapitulated in Table 2.

Table 2: The apparent reaction rate constant and the degradation half-time

[OG] (mg/L)	k_{app} (min^{-1})	$t_{1/2}$ (min)	R^2
2	0.622	1.114	1
10	0.326	2.126	0.975
20	0.117	5.924	0.969
30	0.071	9.762	0.988
50	0.034	20.386	0.982

The apparent reaction rate constants (k_{app}) are calculated from the integrated first-order equation:

$$\ln \frac{C}{C_0} = -k_{app} t \qquad (3)$$

The relationship between the half-time ($t_{1/2}$) and k_{app} is given by:

$$t_{1/2} = \frac{\ln 2}{k_{app}} \qquad (4)$$

From Table 2, it was observed that the apparent reaction rate constants decreased with increasing the initial dye concentration from 2 mg/L to 50 mg/L, while the half-time increased.

The decrease in the degradation kinetics with the increase in dye initial concentration can be explained by an inner filter effect caused by the rise of turbidity. Other possible explanation is the formation of intermediates which compete with OG for hydroxyl radicals thus reducing the degradation efficiency.

3.2. Contribution of hydroxyl radical in the UV-C system

Photolysis, both direct and indirect, has been shown to cause the breakdown of many emerging contaminants. In direct photolysis, the contaminant itself absorbs photons and is degraded as shown below [27]:

$$\text{Contaminant} + h\nu \rightarrow \text{Contaminant}^{*} \quad (\lambda = 254\ \text{nm}) \qquad (5)$$

$$\text{Contaminant}^{*} + O_2 \rightarrow \text{Contaminant}^{\bullet +} + O_2^{\bullet -} \qquad (6)$$

$$\text{Contaminant}^{\bullet +} \rightarrow \text{Products} \qquad (7)$$

In indirect photolysis, degradation occurs via $HO^{\bullet}$radical-mediated pathway through water photolysis, in which water is homolyzed into hydrogen atom and hydroxyl radical according to the following scheme [28]:

$$H_2O + h\nu \rightarrow HO^{\bullet} + {}^{\bullet}H \quad (\lambda = 185\ \text{nm}) \qquad (8)$$

$$HO^{\bullet} + \text{Contaminant} \rightarrow \text{Products} \qquad (9)$$

In order to evaluate these two paths, in other words, to evaluate the contribution of hydroxyl radical to the photooxidation mechanism, the $HO^{\bullet}$ radical-mediated process should be suppressed using a known $HO^{\bullet}$ radical scavenger in the solution. Experiments were conducted using tert-butyl alcohol which is a common $HO^{\bullet}$ radical scavenger [20]. As depicted in Fig.4a, the degradation of OG was significantly affected by the addition of tert-butanol to the solution. Moreover, this inhibition is more important when the dose of the alcohol increases. The apparent reaction rate constant decreased from 0.034 min^{-1} to 0.004 min^{-1} when 3.33% (V/V) concentration was added to the solution (Fig.4b). This is demonstrating the involvement of $HO^{\bullet}$ in the photolytic process. Therefore, the degradation is dominated by $HO^{\bullet}$ pathway.

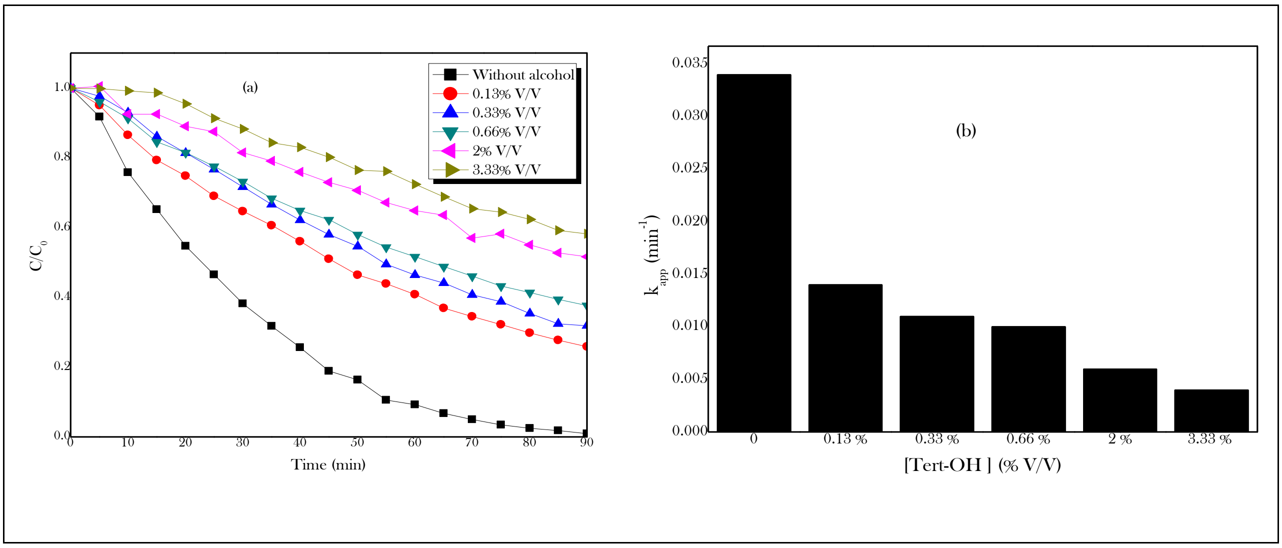

Figure 4. (a) Effect of tert-butyl alcohol on the color degradation of OG ; (b) Dependance of apparent reaction rate constant with alcohol concentration

3.3. Effect of bicarbonate ions on OG decolourization

Bicarbonate (HCO_3^-) is a major compound in aqueous environments, with concentrations varied from several dozen to several hundred mg/L [29, 30]. So, it should be an important scavenging factor for AOPs treatment.

For this purpose, decolourization of OG at initial concentration of 50 mg/L was studied in the presence of various concentrations of sodium bicarbonate ($NaHCO_3$) ranging from 30 mg/L to 1000 mg/L. According to the results shown in Fig.5a, low concentration of HCO_3^- (30 mg/L), had no significant effect on the removal efficiency. Indeed, 99% of decolourization was obtained in 90 min as it was in the absence of HCO_3^-. However, from 70 mg/L of HCO_3^- the inhibition effect became apparent. Fig.5b, shows the relationship between the apparent reaction rate constant (k_{app}) and the HCO_3^- concentration. When the concentration of HCO_3^- increased from1000 mg/L, k_{app} decreased from 0.034 min^{-1} to 0.016 min^{-1}.

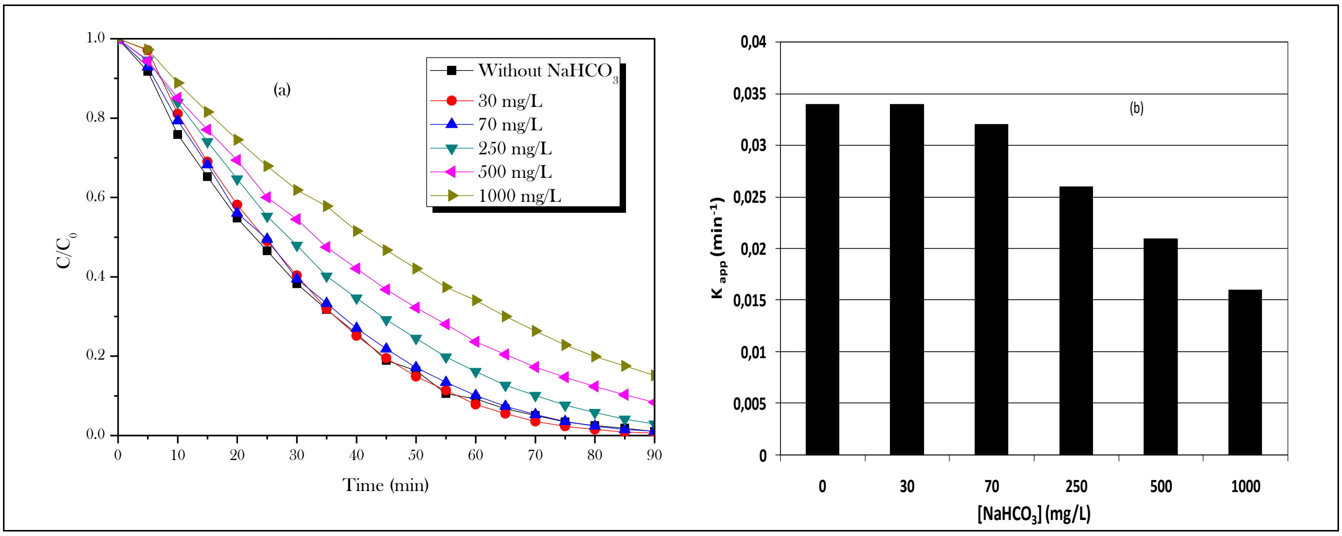

Figure 5. (a) Effect of bicarbonate on the color degradation of OG. (b) Dependence of apparent reaction rate constant with substrate concentration

Bicarbonate ionis known to react with hydroxyl radical to produce the carbonate radical ($CO_3^{\bullet-}$) [20, 31] as shown in Reaction 10:

$HCO_3^- + HO^\bullet \rightarrow CO_3^{\bullet-} + H_2O \quad k = 8.5x10^6\ M^{-1}s^{-1}$ (10)

As demonstrated in section 3.2 (contribution of hydroxyl radical in the UV-C system), OG decolourization is dominated by $HO^\bullet$ pathway. Thus, the fact that HCO_3^- compete with OG for the hydroxyl radical, probably explain the inhibition pattern found for the removal of OG by the UV system. This result is in conformity with others studies [17, 24] which attribute the negative effect found in their results to the hydroxyl radical scavenging effect.

3.4. Effect of carbonate ions on OG decolourization

Carbonate ions are common anions in water body, which have also important effect on the reactive species like hydroxyl radicals [32]. The effect of carbonate ions (CO_3^{2-}) was also studied for concentration ranging from 30 to 1000 mg/L. The result presented in Fig. 6a, shows that increasing in CO_3^{2-}concentration led to a decrease of OG decolourization. The relationship between the apparent reaction rate constant and the CO_3^{2-} concentration represented in Fig.6b, shows that the apparent reaction rate constant decreased to 0.015 min^{-1} when 1000 mg/L of CO_3^{2-} was added.

As scavenger, carbonate ions can also react with hydroxyl radicals resulting in the formation of carbonate radicals (Reaction 11) [20]:

$CO_3^{2-} + HO^\bullet \rightarrow OH^- + CO_3^{\bullet-} \quad k = 3.8x10^8 M^{-1}s^{-1}$ (11)

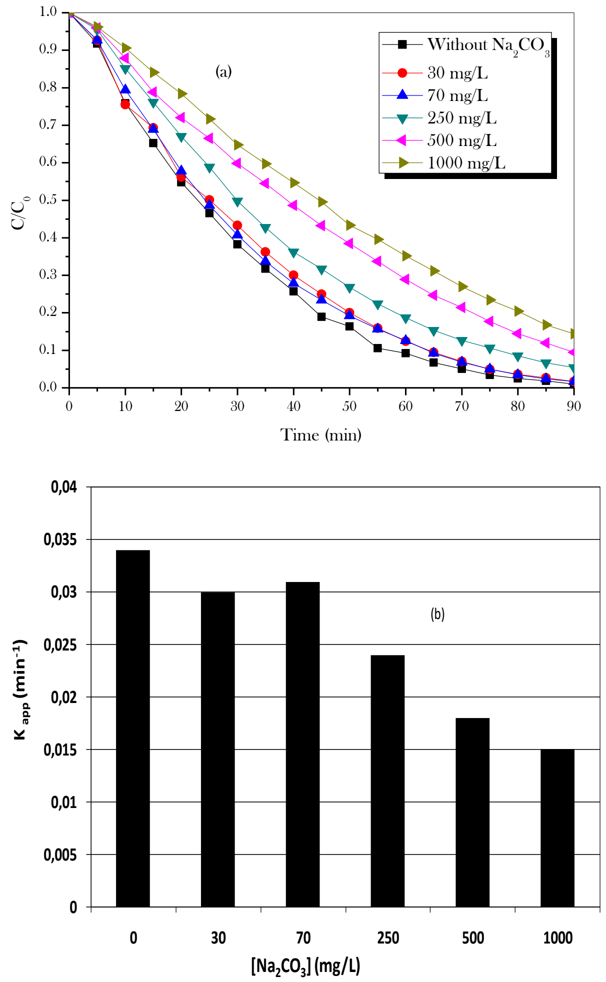

Figure 6. (a) Effect of carbonate on the color degradation of OG. (b) Dependence of apparent reaction rate constant with substrate concentration.

Comparing the effect of the two ions, we notice that the decreasing in the apparent reaction rate constant of CO_3^{2-} was faster than in case of HCO_3^- (Fig. 7), this is because the rate constants (Reactions10 & 11) show that CO_3^{2-} is 45 times more reactive with $HO^•$ than HCO_3^-, hence is more important retardant of the oxidation process [17].

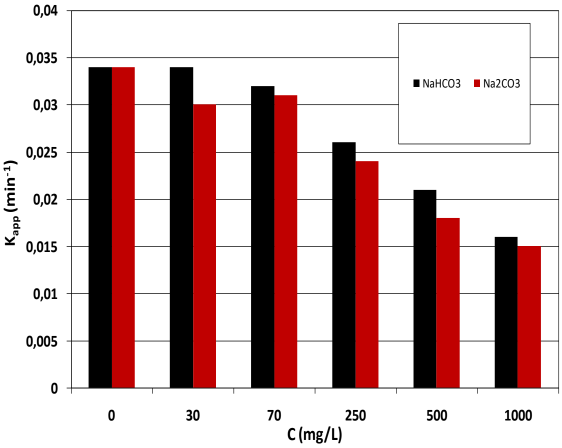

Figure 7. Dependence of apparent reaction rate constant with substrate concentration

The effect of bi/carbonate ions on the OG removal cannot be only attributed to the hydroxyl radical scavenging effect, but also includes the reactivity of the carbonate radical formed as a consequence to this scavenging. Indeed, in contrast to the hydroxyl radical, the carbonate radical is less reactive toward the degradation of organic compounds [33]. It is also very selective and attacks compounds trough electron transfer or hydrogen abstraction with rate constants in the range of 10^2- 10^9 M^{-1} s^{-1} [18]. This is why its involvement in the destruction of certain pollutants is favourable. Lai *et al.* [29] reported that the abundance of $CO_3^{•-}$ preferred moieties in methotrexate has enhanced the degradation when bicarbonate was added to the UV/TiO_2 process.

3.5. Effect of bromide ions on the OG decolourization

The bromide ions (Br^-) are present in natural water at different concentrations between 3 mg/L and 67 mg/L [34]. They are the most important scavengers of $HO^•$ in seawater, removing up to 93% , hence they are potentially of more significant concern for AOPs [35, 36].

The effect of bromide on the photodecolourization of OG was investigated. From Fig. 8 we can show that the removal efficiency of OG was significantly affected in the presence of bromide. When 10, 100, 1000 and 3000 mg/L of KBr were added to the solution, 98%, 87%, 38% and 28% color removal was obtained respectively and k_{app} decreased from 0.034 min^{-1} (without bromide) to 0.002 min^{-1} when 3000 mg/L of Br^- were present.

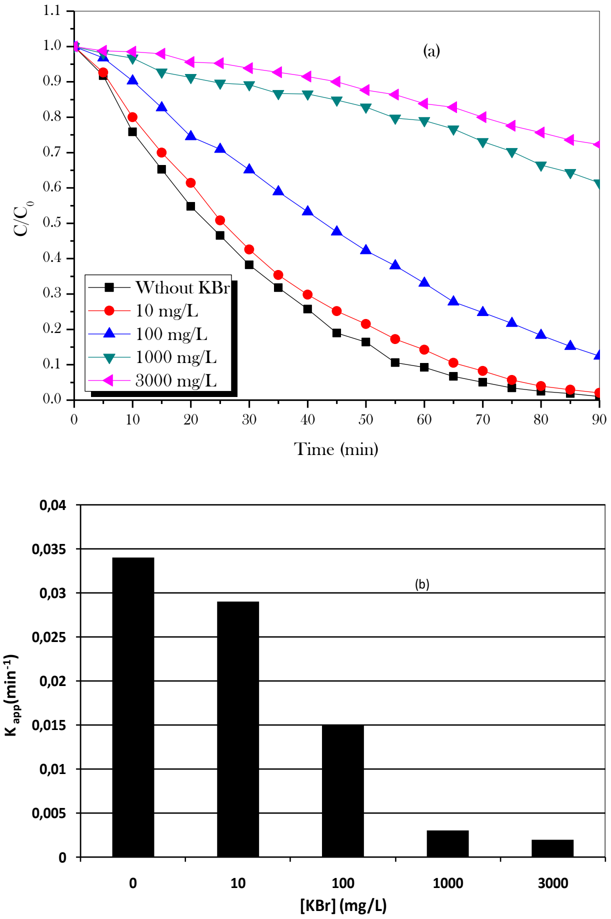

Figure 8. (a)Effect of bromide on the color degradation of OG. (b) Dependence of apparent reaction rate constant with substrate concentration

Bromide ion is able to react with hydroxyl radical and yield the dibromine radical $Br_2^{•-}$ as described below [37]:

$$2Br- + HO^• \rightarrow Br_2^{•-} + OH^- \qquad (12)$$

This radical is reactive but less than the hydroxyl radical toward organic and inorganic compounds [38]. As a consequence, the decrease of OG decolourization rate in the presence of bromide ions can be attributed to the scavenging of $HO^•$ and to the lower reactivity of $Br_2^{•-}$.

3.6. Effect of water matrices

Five different types of water (ultra-pure water, mineral water, tap water, Mediterranean and Dead Sea waters) were used as target matrices to investigate the photodecolourization of OG. Apart from the slight improvement obtained from the ultra pure water (99.8%), all the other matrices showed a negative effect compared with the distilled water (Fig. 9). The removal efficiency was

84.5%, 64.4% and 57.4% for tap water, mineral water and Meditteranean Sea water respectively. The worst result (17%) was obtained with Dead Sea water which is the most hypersaline waterbody on earth. These findings confirm the effect of different inorganic ions especially bi/carbonate and bromide ions on the photodegradation of OG. Besides, the effect of other water components may also contribute to the discrepancy [30].

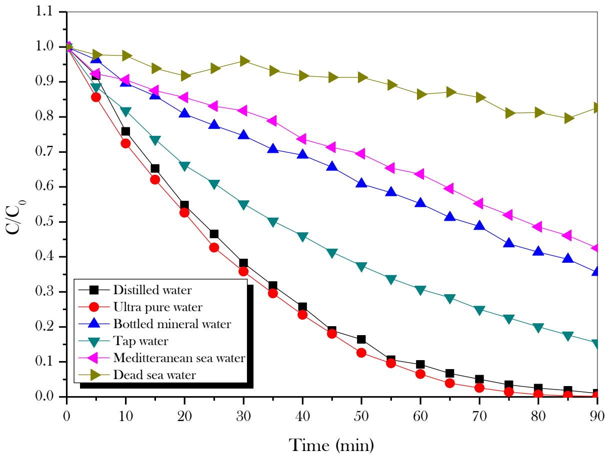

Figure 9.Effect of water matrices on the color degradation of OG.

4. Conclusion

In this study, it was illustrated that the photo-decolourization of OG at different concentrations was achieved after 90 min. However, the required time increased with increasing dye concentration. The process was adequately modeled using first order kinetics and pseudo-first reaction constants were obtained. The addition of some ions (bi/carbonate and bromide) at different concentrations reduces the reaction rate of OG (50 mg/L) photochemical decolourization. This inhibition is due to scavenging of hydroxyl radicals, despite the formation of other radicals which are probably less reactive than hydroxyl radicals towards OG. Different types of water (ultra-pure water, mineral water, tap water, Mediterranean and Dead Sea waters) were evaluated. Except for pure water, a decrease in removal efficiency was obtained with the tested matrices which has attributed to the effect of the presence of salts especially in the case of Dead Sea water. These results are helpful in understanding the fate of organic contaminants in real matrices.

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