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# New UV-LED frontal flow photocatalytic reactor for VOCs treatment: Compactness, intensification and optimization studies

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1           **Conception of new compact frontal flow photocatalytic reactor**  
2           **based on luminous textiles with high energy efficiency:**  
3           **Intensification and optimization studies toward gas**  
4           **pollutant elimination**

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6  
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11  
12    **Abstract**

13    A solution for the optimization of the photocatalytic treatment of toxic gases based on  
14    the use of TiO<sub>2</sub> media deposited on luminous textile, was studied and implemented.  
15    Cyclohexane was chosen as a reference for the type A gas filter tests. The  
16    photocatalytic media were firstly characterized by scanning electron microscopy  
17    (SEM). Then, the experiments carried out on a batch reactor showed that the TiO<sub>2</sub>  
18    coated optical fiber media gives better performances than conventional TiO<sub>2</sub>  
19    deposited on cellulosic fiber support. In order to take advantage of the new  
20    configuration based on optical fiber media, an intensification study was carried out by  
21    increasing the amount of TiO<sub>2</sub> in the media, and UV intensities of LED. The increase  
22    of these two parameters leads to a process degradation rate of about 4 times higher.  
23    The continuous treatment allowed the efficiency of the new configuration of the front  
24    flow reactors developed (PFR-LED) to be highlighted, in comparison with the

25 conventional configuration. This is interpreted in the increase of the specific  
26 degradation rate of the optimized PFR-LED by four times compared to the  
27 conventional reactor. The performance evaluation of the compact and optimized  
28 configuration of the frontal flow reactor (PFR-LED Optimized), aimed at highlighting  
29 the influence of the inlet concentration under different flow rates. In addition, the  
30 effect of the number of optical fibers support shows that the degradation rate and  
31 selectivity are enhanced. The main rates obtained were for a cyclohexane input  
32 concentration of  $1.19 \text{ mmol.m}^{-3}$  in optimal humid condition (38%) at an air flow rate of  
33  $18 \text{ L min}^{-1}$  with four photocatalytic media (4OF/4UV-LED), where the  $\text{CO}_2$  selectivity  
34 reached 31% for an abatement of 59%.

35 This global investigation, has mainly allowed the design of a new version of compact  
36 reactor. A version as economical as efficient in terms of elimination of gaseous  
37 pollutants which clearly meets the main goals of sustainable development.

## 38 **Keywords**

39 Air treatment; Front flow photocatalytic reactor; kinetic modelling; Compactness;  
40 Intensification; Optimization.

## 41 **1. Introduction**

42 Indoor and outdoor air pollution has become a real concern in today's world. Outside  
43 pollution is caused by many factors, sometimes natural such as volcanic air pollution  
44 [1]. But most often air pollution is of human origin, namely the pollution released by  
45 the large number of vehicles as well as by industries [2]. Inside, the air is also  
46 polluted in a specific way by potential sources (combustion devices, construction  
47 materials, decoration products) and also human activity (smoking, cleaning products,  
48 cooking,.. ) [3,4]. When this air, constituting a wide variety of volatile organic

49 compounds (VOCs), is breathed in by humans, it results in respiratory problems and  
50 lung cancer [1][2].

51 On this context, photocatalysis can be considered as a promising solution for air  
52 purification [3–7]. This process allows the mineralization of many organic compounds  
53 at room temperature using a semiconductor, namely  $\text{TiO}_2$  (high photoactivity,  
54 chemical stability and low cost) and an ultraviolet source to decompose VOCs  
55 through consecutive oxidation reactions and produce  $\text{CO}_2$ ,  $\text{H}_2\text{O}$  and other by-  
56 products [4-8].

57 Continuous reactors, equipped by immobilized catalyst, are most frequently used in  
58 air purification [10,11][12][13]. However, as reported in the literature [14], many  
59 disadvantages come into play when a fixed photocatalyst system is implemented. On  
60 the one hand, it is necessary to illuminate a large surface of the photocatalyst. The  
61 loss of photons due to the absorption of the UV radiation by the support leads to a  
62 decrease efficiency of the catalyst. On the other hand, another crucial disadvantage  
63 of this process, is the use of a UV lamp as a light source. Due to the lamp size, the  
64 reactor volume , as well as the high energy consumption [15,16] became an obstacle  
65 in the reactor design. New reactor designs based on optical fibers as a support for  
66 the photocatalyst have been developed and tested to overcome these problems.  
67 These reactors lead to an improvement of the photocatalytic efficiency due to better  
68 contact between the catalyst, the light and the pollutant [14–18][21] [22]. However,  
69 less mass transfer is observed under parallel configuration. [23]. Faced with this  
70 disadvantage, frontal flow photocatalytic reactors present an efficient solution, as this  
71 configuration allows to play on the compactness of the reactor for the treatment of air  
72 at high flow rates by favoring the mass transfer [24][25][26].

73 The aim of our contribution concerns the design of a new frontal flow reactor for the  
74 treatment of polluted air by optical fibers textile developed by the company Brochier  
75 Technologies-France. A particularly innovative aspect of this illuminated textile is the  
76 simultaneous weaving of textile and optical fibers, allowing the frontal flow  
77 photocatalytic reactor to have a very compact design. Therefore, the illuminated  
78 textile is considered both as a support for the  $\text{TiO}_2$  and as a transmitter of light  
79 through the optical fibers from the source to the catalyst surface. Thus, it combines  
80 the maximization of the interactions between the pollutant and the catalyst surface  
81 with a very large illumination area of the catalyst surface (maximization of the  
82 interactions between the light and the catalyst surface) [23,27–29]. In addition, this  
83 configuration allows the reactor size to be reduced compared to conventional  
84 reactors (process intensification), with less energy consumed [23,30,31].

85 In this study, a comparison between new and conventional configurations was  
86 highlighted. Firstly, in a batch system where Langmuir-Hinshelwood model was  
87 applied to represent the kinetics and compare the potential of the light fabric to the  
88 conventional heterogeneous photocatalysis process, i.e. using  $\text{TiO}_2$  deposited on  
89 cellulose paper based on activation by an external UV lamp. Followed by a  
90 degradation efficiency comparison of the front-flow photocatalytic reactors  
91 configurations, considering energy consumption with special attention to the reactor  
92 compactness. In addition, the parameters of the new frontal flow photocatalytic  
93 reactor based on optical fiber media were optimized, in order to further enhance its  
94 photocatalytic efficiency and to confirm its ability to treat pollutants under the  
95 influence of the operating conditions, i.e. inlet concentration, flow rate and relative  
96 humidity as well as the number of photocatalytic medias.

97 It should be noted that the established literature review does not show any work on  
98 this new configuration based on the sustainable development goals, even on its  
99 intensification.

## 100 **2. Material and methods**

### 101 **2.1. Pollutant studied**

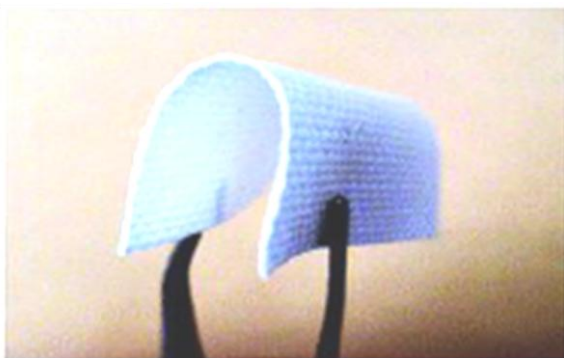
102 In this study, cyclohexane was used as a target pollutant. It is supplied by Sigma-  
103 Aldrich/Germany, in liquid form (purity > 98%), volatilized afterwards to obtain the  
104 desired gas concentrations.

### 105 **2.2. Photocatalytic support**

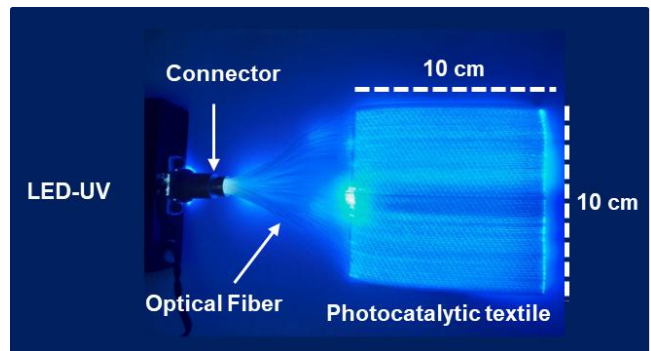
106 Two photocatalytic supports were used in this study (Fig. 1). The first one is a media  
107 provided by the Ahlström Paper Group, where the conditioning was achieved by an  
108 aqueous suspension of a mixture of TiO<sub>2</sub> (Millennium PC 500) and silica, deposited  
109 on a non-woven fibrous support of synthetic and natural cellulose. A winding press  
110 process was used to coat the fibers by fixing TiO<sub>2</sub> to different substrates. The  
111 colloidal silica used has a specific surface area of about 700 m<sup>2</sup>.g<sup>-1</sup>, with elementary  
112 particles of 20-30 nm in diameter. It acts as an inorganic binder, resistant to UV  
113 radiation and photocatalysis, transparent to UV and protecting the fibers. The  
114 titanium dioxide then forms agglomerates of varying size (0.1 to 1 μm). More details  
115 on this photocatalytic media have been presented in our previous publications  
116 [31][32][33].

117 The second is a new double-sided photocatalytic media, manufactured by Brochier  
118 Technologies (UVtex®). Based on the combination of plastic optical fibers (OF)  
119 (polymethylmethacrylate) around which textile fibers (polyester, Trevira CS TM fibers)  
120 are wound. The OFs have an average diameter of about 500 μm. Indeed, the core of

121 the OF is formed by a polymethyl methacrylate resin with an average diameter of 480  
122  $\mu\text{m}$  and covered with a 10  $\mu\text{m}$  thick fluoropolymer [29]. These optical fibers are  
123 treated to transmit and emit light laterally on their outer surface. For a textile sample,  
124 all OFs are collected at one end in an aluminum connector. The luminous textile is  
125 covered with a layer of silica that serves as a protection against degradation during  
126 the photocatalytic process. However, silica was chosen because it allows UV light  
127 transmittance, and being a non-organic compound, it is not degraded by  
128 photocatalysis [28]. The textile fiber is soaked for 60 min in a stirred suspension  
129 containing  $\text{TiO}_2$  Degussa P25 powder ( $50 \text{ g.L}^{-1}$ ), then dried for 2 h at  $70 \text{ C}^\circ$ . At the  
130 end of this process,  $12 \text{ g.m}^{-2}$  of  $\text{TiO}_2$  are uniformly distributed on the textile fiber.  
131 More information are mentioned in our previous publications [23,27].



**$\text{TiO}_2$  deposited on Cellulosic support**

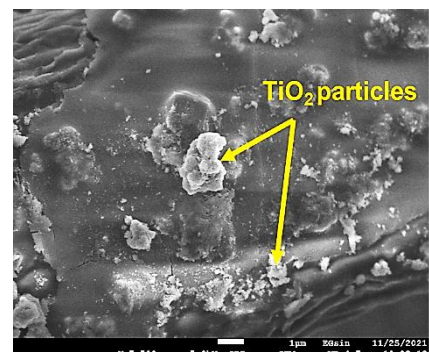
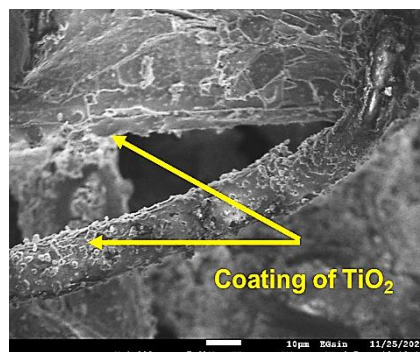
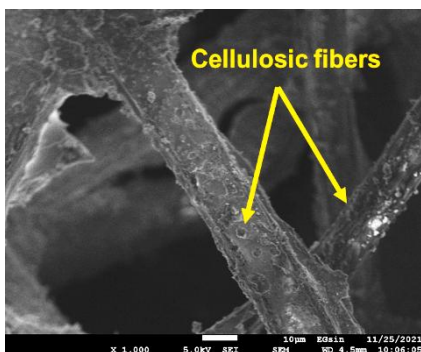


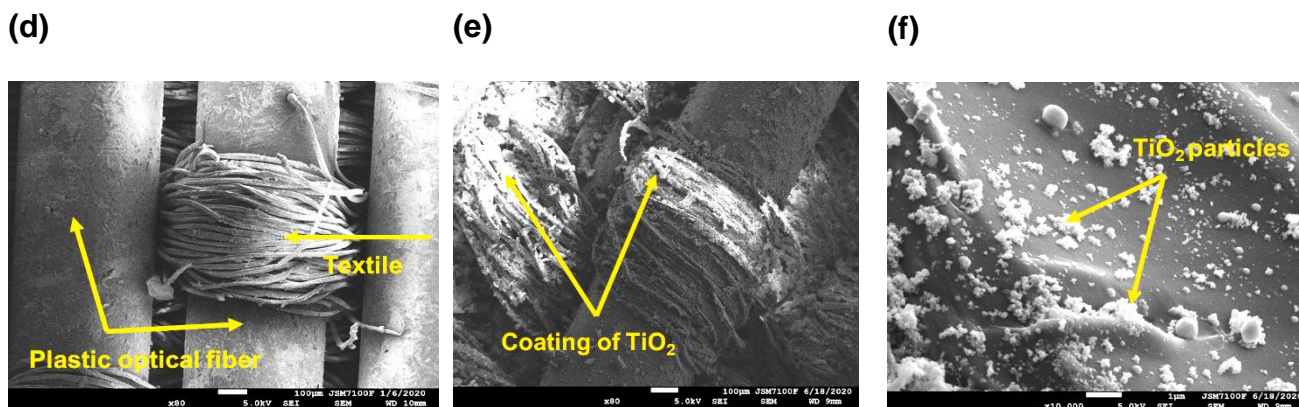
**$\text{TiO}_2$  coated on Optical Fibers**

(a)

(b)

(c)





**Figure 1:** SEM pictures of photocatalytic materials: (a, b and c) TiO<sub>2</sub> deposited on cellulosic support, (d, e and f) TiO<sub>2</sub> coated Optical fiber media.

132

133

134 Details of the surface texture characterization of the cellulosic and OF supports were  
 135 analyzed by scanning electron microscopy (SEM- JEOL JSM 7100F). Figure 1 (a, b  
 136 and c) shows the location of the TiO<sub>2</sub> particles deposited on the cellulosic fibers. As  
 137 described in our previous work [23,27], figure 1 (d, e and f) shows OFs on which TiO<sub>2</sub>  
 138 nanoparticles have been coated. The OF is knitted, according to the Jacquard  
 139 process, i.e. the interweaving of fibers are placed in the direction of the textile fibers  
 140 (chain), and of some fibers which are placed perpendicular to the fibers of the warp,  
 141 in the direction of the OF (Fig. 1 d, and e). As shown in Fig. 1.e, a coating layer of  
 142 TiO<sub>2</sub> is present on the textile fiber and the starting threads. Therefore, this allows a  
 143 large amount of TiO<sub>2</sub> to be activated, which will improve the photocatalytic efficiency.

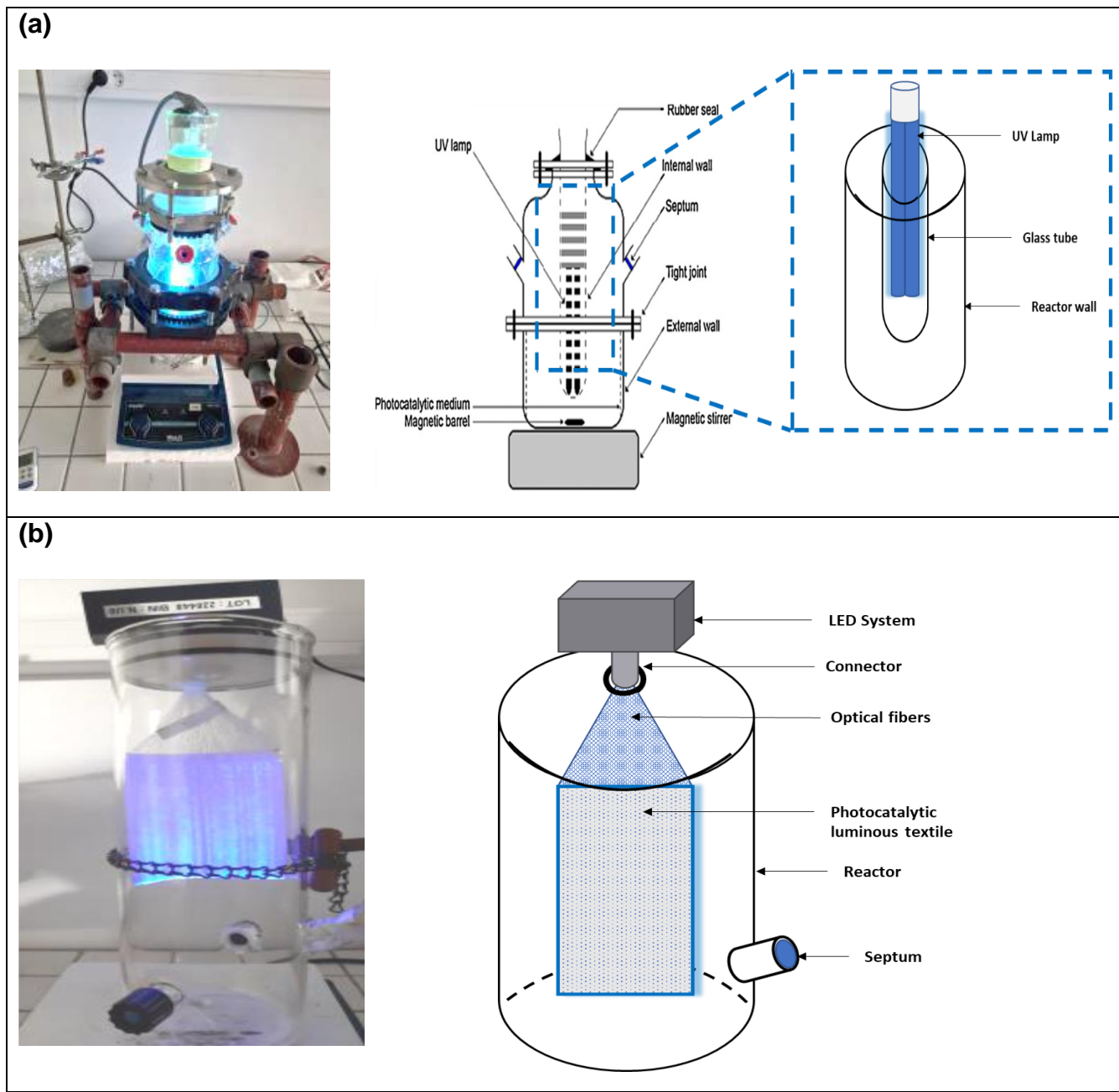
### 144 **2.3. Reactor and experimental set-up:**

#### 145 **2.3.1. Batch reactors**

146 In this study, two types of photoreactor were used. The first one is a conventional  
 147 reactor (Fig. 2a) which contains a central tube for the installation of the fluorescent



148 UV lamp (Philips PL-L 24W/10/4P) which allows the activation of the cellulose  
149 photocatalytic support deposited on the inner wall of the reactor. It is approximately  
150 40 cm high and has a volume of 1.5 L. The UV intensity of the lamp is measured by a  
151 VLX-3W radiometer equipped with a CX-365 cell. The second is a reactor with a  
152 luminous textile (Fig.2b) with a surface of 10\*10 cm containing a quantity of TiO<sub>2</sub>  
153 equal to 10 g.m<sup>-2</sup>. UVA-LED type (Bin N) connected to a dimmer supplied by Brochier  
154 technologies (Lightex) was used to ensure a good distribution of the radiation on the  
155 surface catalyst through the fiber filaments.



**Figure 2:** Photo and schema of batch reactors: (a) conventional configuration based on cellulosic media, (b) Optical fiber (luminous textile) configuration.

156

157 The major differences between the two reactors were the light intensity and reactor  
 158 volume used, which were greater with the conventional photocatalytic plant. This was  
 159 due to the use of external light source positioned vertically in the reactor to activate  
 160 the  $\text{TiO}_2$  deposited on the cellulose paper.

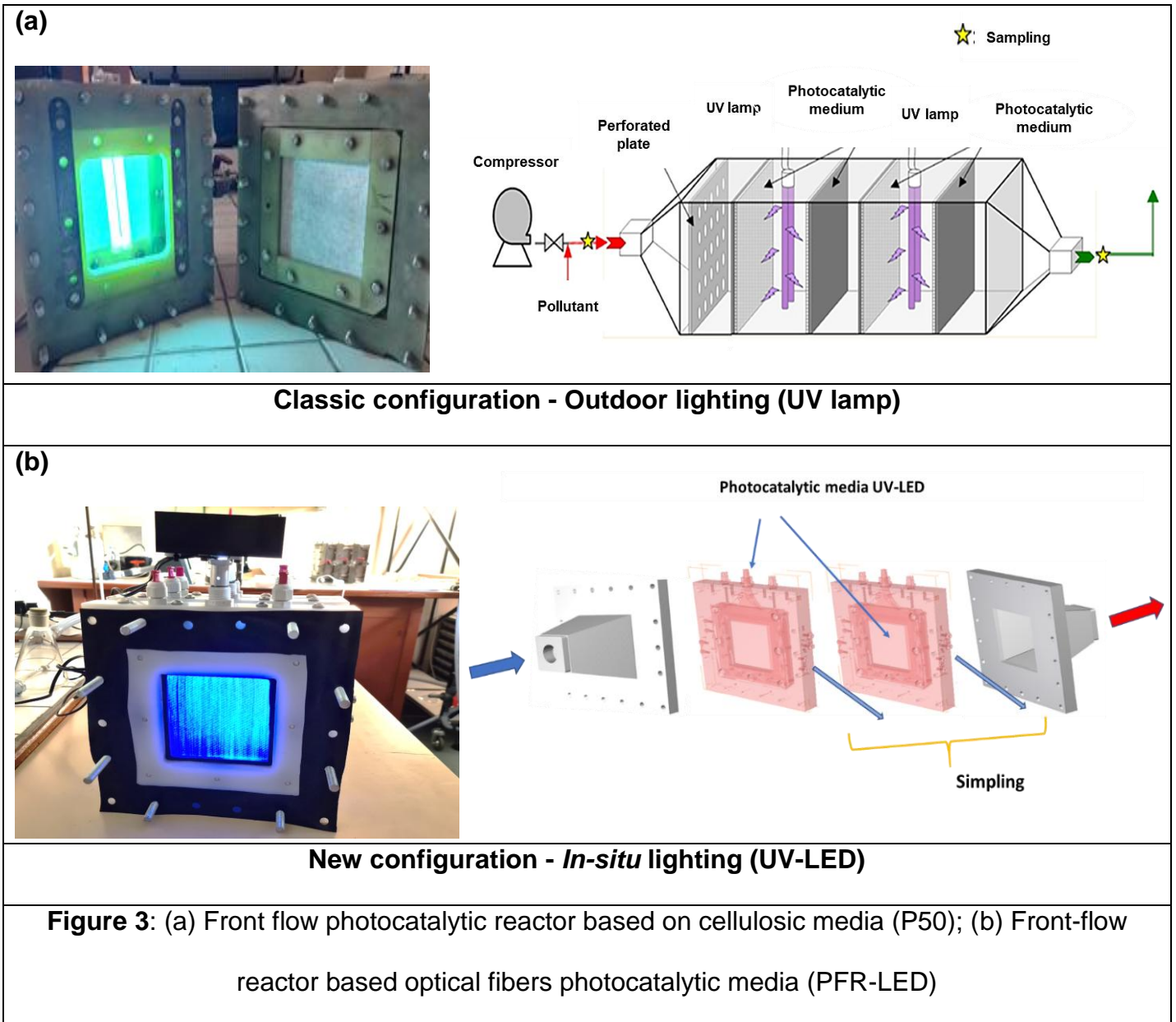
161 To generate pollution in the reactor, the pollutant is injected in the form of liquid drops  
162 which then evaporate. A magnetic stirrer is used to homogenize the gases.

### 163 **2.3.2. Continuous process**

164 The classic front flow photocatalytic reactor, namely (P50) is illustrated in Fig. 3a,  
165 with a passage section of  $0.01 \text{ m}^2$  and a total volume of about 4 L. It is formed by four  
166 mountable stainless-steel test chambers, containing four Cellulosic photocatalytic  
167 media ( $4 \times 0.01 \text{ m}^2$  of surface giving 0.64 g of  $\text{TiO}_2$ ) and two Philips UV-A lamps  
168 (model PLS 9W / 10). The intensity of the light incident at 365 nm on the surface of  
169 the catalyst is  $20 \text{ W/m}^2$  which is measured by the radiometer "VLX 3 W").

170 The second reactor represents a new configuration (Fig. 3b), recently developed and  
171 built to work in frontal flow. The reactor is composed of modules adapted for the  
172 installation of  $\text{TiO}_2$  on OF media, ensuring a passage section similar to that of the  
173 classic reactor (about  $10 \text{ cm}^2$ ). Made of Bakelite, which gives it a high mechanical  
174 resistance (hard, rigid), and good electrical insulation and temperature resistance  
175 ( $120 \text{ C}^\circ$  continuously). Bakelite is also not influenced by photocatalysis and UV light.  
176 The objective of this configuration is to optimize the photocatalytic treatment by  
177 reducing the residence time, while ensuring good degradation efficiency. This last  
178 configuration has been developed in two ways. The first one was based on OF media  
179 loaded with  $10 \text{ g.m}^{-2}$  of  $\text{TiO}_2$ , with an UV Intensity of  $1.5 \text{ W.m}^{-2}$ , namely Photocatalytic  
180 Filtrating Reactor-LED (PFR-LED). The second one, in order to optimize the  
181 photocatalytic treatment process, consists of an OF media containing  $26 \text{ g.m}^{-2}$  of  
182  $\text{TiO}_2$ , with an applied UV Intensity about  $2.6 \text{ W.m}^{-2}$  called PFR-LED Optimized.

183 It should be noted, that the electrical energy consumed and required for the  
184 photocatalytic process for each reactor, was measured using an energy cost meter  
185 (ENERGY LOGGER 4000F) of the manufacturer VOLT CRAFT®.



186

187

188

189 The main characteristics of the different reactors and configurations are presented in  
 190 Table 1.

Characteristics	reactors		
	P50	PFR-LED	PFR-LED Optimized
Volume (m <sup>3</sup> )	4.10 <sup>-3</sup>	1,2.10 <sup>-3</sup>	1,2.10 <sup>-3</sup>
Compactness (m <sup>2</sup> .m <sup>-3</sup> )	2.5	11.11	11.11
Masse TiO <sub>2</sub> (g.m <sup>-2</sup> )	64	3	78
UV intensity (W.m <sup>-2</sup> )	20	4.5	7.8
Energy consumed (watt)	31.1	14.5	15.7

**Table 1:** Characteristics comparison of frontal flow photocatalytic reactors

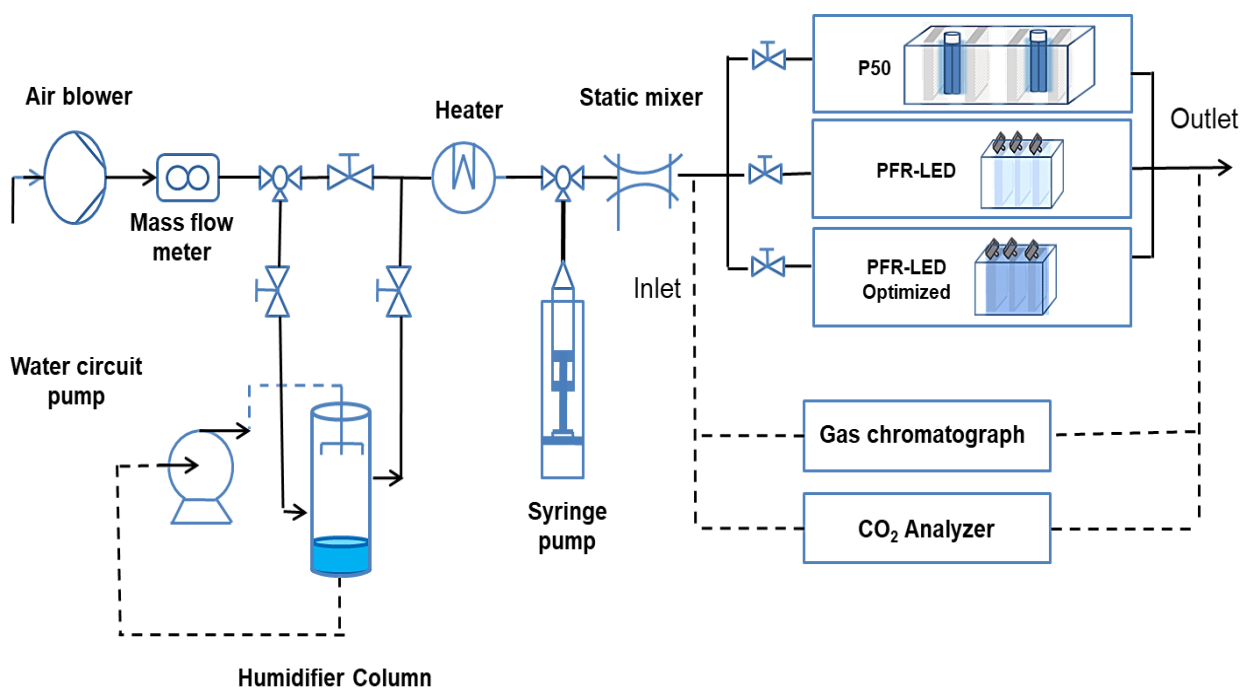
191

192 **2.3.2.1. Experimental setup:**

193 The appropriate reactor for the test is installed in each case. Dry air from the network  
 194 is sent as the carrier gas, controlled by a valve and a Gallus G4 gas meter of the  
 195 Itrón brand. The flow rates tested are equal to 13, 18 and 36 L.min<sup>-1</sup>. These flow  
 196 rates correspond to contact times of 18, 13 and 7 ms respectively. To humidify the  
 197 air, a bubbler was installed in the circuit containing water to achieve humidity ranges  
 198 of 5-80%. The pollutant is injected continuously in liquid form by a syringe / syringe  
 199 pump combination. A heating tape, is placed in the injection zone to facilitate the  
 200 volatilization of the pollutant and a static mixer makes it possible to homogenize the  
 201 effluent upstream of the photoreactor. Two septa downstream and upstream of the  
 202 photoreactor make it possible to sample the outlet and inlet gas with a syringe  
 203 (Fig.4).

204 Once the adsorption process reaches equilibrium (depending on the concentration),  
 205 as indicated by an identical input / output Volatile Organic Compound concentration,  
 206 the UV lighting is turned on. A transient period precedes a steady state phase which  
 207 is established when the concentration of pollutant at the outlet becomes constant.

208 The reactor is rinsed under UV light for 1 h in ambient air after each experimental  
 209 sequence. or it is useful to report that no deactivation has been observed.



**Figure 4:** Diagram of the continuous photocatalytic process treatment for the three reactors

210 **2.3.2.2. Analytic tools**

211 The concentration of cyclohexane was measured by a Thermo electron corporation  
 212 gas chromatography (Focus GC) using a flame ionization detector (FID) and an  
 213 FFAP column (length = 25 m and internal diameter = 0,32 mm). Nitrogen was used  
 214 as a carrier gas. The temperature conditions of the oven, the injection chamber and  
 215 the detector were, respectively, 50, 190 and 190 °C. The analysis was performed by

216 direct manual sampling with a 500 µl syringe and injection into the GC. The  
217 calibration was carried out by evaporating different quantities of cyclohexane on a  
218 closed bottle. The correlation of the pollutant with a peak area of GC-FID as a  
219 function of its concentration was carried out. Operation is done by Azur™ software.  
220 The CO<sub>2</sub> was analyzed by a Fourier transform infrared spectrophotometer (FTIR)  
221 from Environment SA (MIR 9000H).

### 222 3. Results and discussions

#### 223 3.1. Photocatalytic performance of catalysts

224 The objective of this study is to highlight the performance of the two studied  
225 photocatalytic media, cellulosic and OF. For a better evaluation, the efficiency of the  
226 media was regarded in terms of the photocatalytic degradation rate represented by  
227 the Langmuir-Hinshelwood model (Eq.1), which considers the contribution of the  
228 adsorption properties of the pollutant on the catalyst surface and the kinetics of the  
229 photocatalytic reactions [34][35][36]. Due to the complex mechanism of the reactions,  
230 the L-H model is only applied at the beginning of the treatment in order to avoid the  
231 influence of intermediate by-products on the photocatalytic removal of cyclohexane.  
232 As already reported in the literature, the reaction rate constant (k) and adsorption  
233 constant (K) can be determined using Eq.2.

$$234 \quad r_0 = \frac{kK C_0}{1+K C_0} \quad (\text{Eq. 1})$$

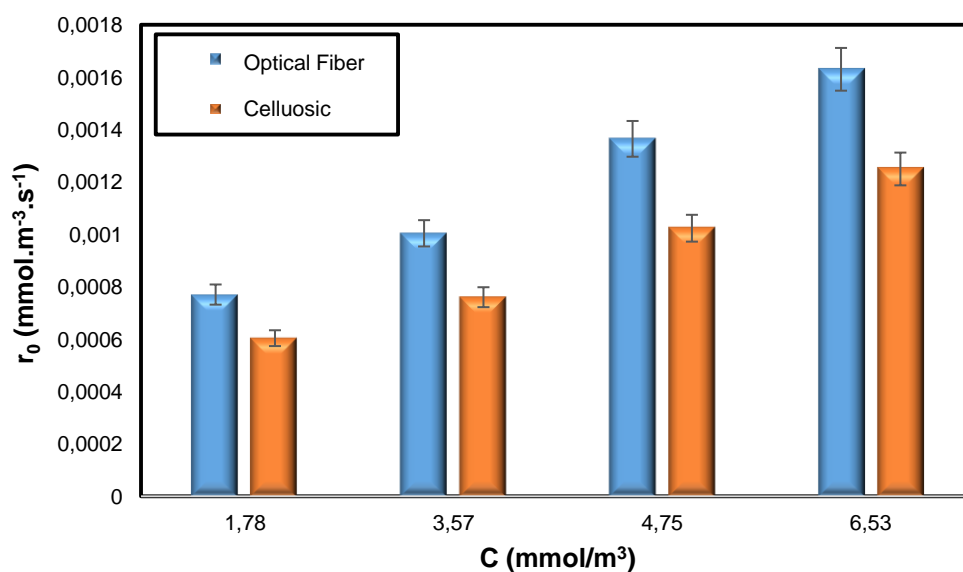
$$235 \quad \text{Or } \frac{1}{r_0} = \frac{1}{kK C_0} + \frac{1}{k} \quad (\text{Eq. 2})$$

236 where  $r_0$  is the initial reaction rate ( $\text{mmol.m}^{-3}.\text{s}^{-1}$ ),  $k$  the reaction rate constant  
237 ( $\text{mmol.m}^{-3}.\text{s}^{-1}$ ) and  $K$  is the adsorption constant ( $\text{m}^3.\text{mmol}^{-1}$ ).

238

### 240 3.1.1. Comparison of the photocatalytic media

241 The performance of the cellulosic and OF media was investigated in terms of initial  
 242 degradation rates ( $\text{mmol}\cdot\text{m}^{-3}\cdot\text{s}^{-1}$ ), at different pollutant concentrations ( $\text{mmol}\cdot\text{m}^{-3}$ ), with  
 243 the same amount of  $\text{TiO}_2$  deposited in each media under the same UV intensity  
 244 (Fig.5).



**Figure 5:** Initial degradation rate of Cyclohexane with Cellulosic and Optical fiber media at different concentrations (HR =  $40 \pm 2\%$ , T =  $20 \pm 2 \text{ C}^\circ$ , UV Intensity =  $1.5 \text{ W}\cdot\text{m}^{-2}$ ,  $m_{\text{TiO}_2} = 10 \text{ g}\cdot\text{m}^{-2}$ )

245 Before even deciding on the performance of the two media, it is useful to note that  
 246 the results (Fig. 5) show the same trends, where at low input concentration, the  
 247 degradation follows pseudo first order kinetics where the degradation is proportional  
 248 to the input concentration. This can be explained by the fact that not all active sites  
 249 are occupied and that an increase in concentration generates a greater surface  
 250 coverage which implies a better degradation rate [37].



251 We note that OF media gives a better degradation rate than Cellulosic one  
252 regardless of the initial concentration. Moreover, the improved efficiency of OF  
253 seems to be of the same order for all the initial concentrations tested.

254 Langmuir-Hinshelwood constants for the two medias were determined (table 2), to  
255 permit the comparison of the two media. Here, we note that experimental data fit well  
256 the model which means that the chemical reaction is the limiting step in the process  
257 [38][39].

Media	Cellulosic	Optical fiber
$k_{app} \text{ (mmol.m}^{-3}.\text{s}^{-1})$	0.0027	0.0032
$K \text{ (m}^3.\text{mmol}^{-1})$	0.128	0.144
$R^2 \text{ (%)}$	99.75	99.63

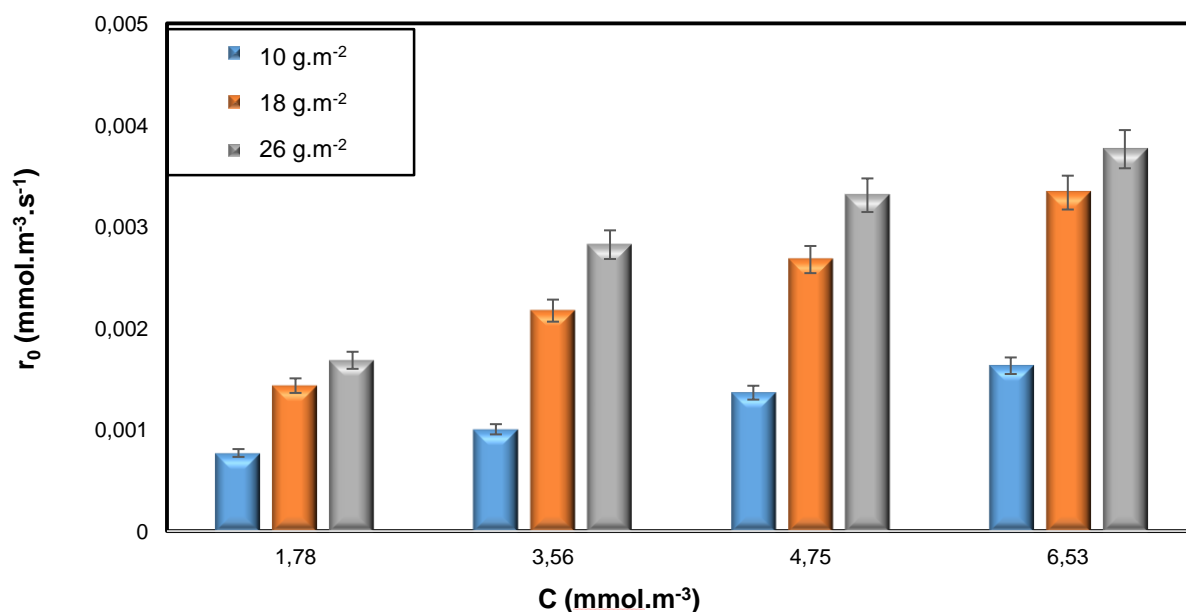
258 **Table 2:** Value of Langmuir-Hinshelwood reaction rate constants (k) and Langmuir  
259 adsorption constants (K)

260 The results confirm that the way to apply the light intensity is the key parameter,  
261 which influences the performance of the photocatalytic processes. The OF media  
262 better treatment performance in terms of mmol removed is also reflected by an  
263 increase in the apparent kinetic constant (k) from 0.0027 to 0.0032  $\text{mmol.m}^{-3}.\text{s}^{-1}$  for  
264 the cellulosic and OF media respectively, due to the *in-situ* illumination. In other  
265 words, for the same UV intensity, more active sites are created with the OF leading to  
266 more electrons and holes which explains the illumination performance translated by  
267 the percentage increase of the initial degradation rate compared to the conventional  
268 media which is about 23% [40].

### 269 **3.1.2. Photocatalytic capacity intensification of the optical fiber media**

#### 270 **3.1.2.a. Effect of mass of $\text{TiO}_2$**

271 In order to increase the photocatalytic efficiency of the OF media, the mass of  $\text{TiO}_2$   
272 deposited on the photocatalytic media was investigated. To this purpose, the  
273 degradation rate was studied for three OF media with respective  $\text{TiO}_2$  masses of the  
274 order of 10, 18 and 26  $\text{g.m}^{-2}$  by applying the same UV intensity ( $1.5 \text{ W.m}^{-2}$ ) at  
275 different concentrations.



**Figure 6:** Initial degradation rate with different mass of  $\text{TiO}_2$  on OF at different Cyclohexane concentrations. (HR =  $40 \pm 2\%$ , T =  $20 \pm 2 \text{ C}^\circ$ , UV Intensity =  $1.5 \text{ W.m}^{-2}$ )

276 The results illustrated in Figure 6 show the variation of the initial rate for the removal  
277 of cyclohexane, with different OF media containing different amounts of  $\text{TiO}_2$ , under  
278 the same UV irradiation. It is observed that the rate of the reaction increases with the  
279 increase in the mass of  $\text{TiO}_2$  deposited on the photocatalytic media, which can be  
280 explained by the fact, that the amount of catalyst increases the number of active sites  
281 on the photocatalyst surface, which in turn increases the number of hydroxyl and  
282 superoxide radicals [41–43]. Furthermore, it is found that the increase in reaction rate  
283 is not proportional to the increase in the amount of  $\text{TiO}_2$  deposited in the medium.  
284 This is shown in Table 3, where the apparent kinetic constants obtained go from

285 0.0032 mmol.m<sup>-3</sup>.s<sup>-1</sup>, 0.0072 mmol.m<sup>-3</sup>.s<sup>-1</sup> for 10 and 18 g.m<sup>-2</sup> which corresponds to  
 286 125% of increase on the other hand by passing from 18 to 26 g.m<sup>-2</sup> it is recorded an  
 287 increase in the initial degradation rate of 15% (0.0072 to 0.0083 mmol.m<sup>-3</sup>.s<sup>-1</sup>). It is  
 288 also noted that the adsorption constant (K) is approximately constant. This can be  
 289 interpreted by the fact that not all photocatalytic sites are activated at high amount of  
 290 TiO<sub>2</sub> [44,45]. In other words, the increase of TiO<sub>2</sub> in the media must be accompanied  
 291 by an enough UV intensity for the activation of all photocatalytic sites. In our case the  
 292 increase of the catalyst load lead to an increase of the degradation rate. This means  
 293 that all the catalyst is activated and there is no limit due to mass transfer. However, it  
 294 is possible that the optimum amount of the catalyst will be reached where the  
 295 efficiency of the degradation rate per unit mass will decrease.

296 This is in agreement with the literature, especially the work of Alonso-Tellez et al. [46]  
 297 and Wang et al. [47], and also that of Vezzoli et al [48], where it is shown that a  
 298 further increase in mass of the catalyst leads to recombination of charge carriers at a  
 299 relatively small distance from the solid-gas interface, resulting in a decrease in the  
 300 reaction rate.

Mass of TiO <sub>2</sub> (g.m <sup>-2</sup> )	10	18	26
<b>k<sub>app</sub> (mmol.m<sup>-3</sup>.s<sup>-1</sup>)</b>	0,0032	0,0072	0,0083
<b>K (m<sup>3</sup>.mmol<sup>-1</sup>)</b>	0,1441	0,1074	0,1170
<b>R<sup>2</sup> (%)</b>	99,63	97,92	98,3

301 **Table 3:** Value of Langmuir-Hinshelwood reaction rate constants (k) and Langmuir  
 302 adsorption constants (K) for the degradation of Cyclohexane with different optical  
 303 fiber media with different mass of TiO<sub>2</sub> at different concentrations. (HR = 40 ± 2%, T  
 304 = 20 ± 2 C°, UV Intensity = 1.5 W.m<sup>-2</sup>)

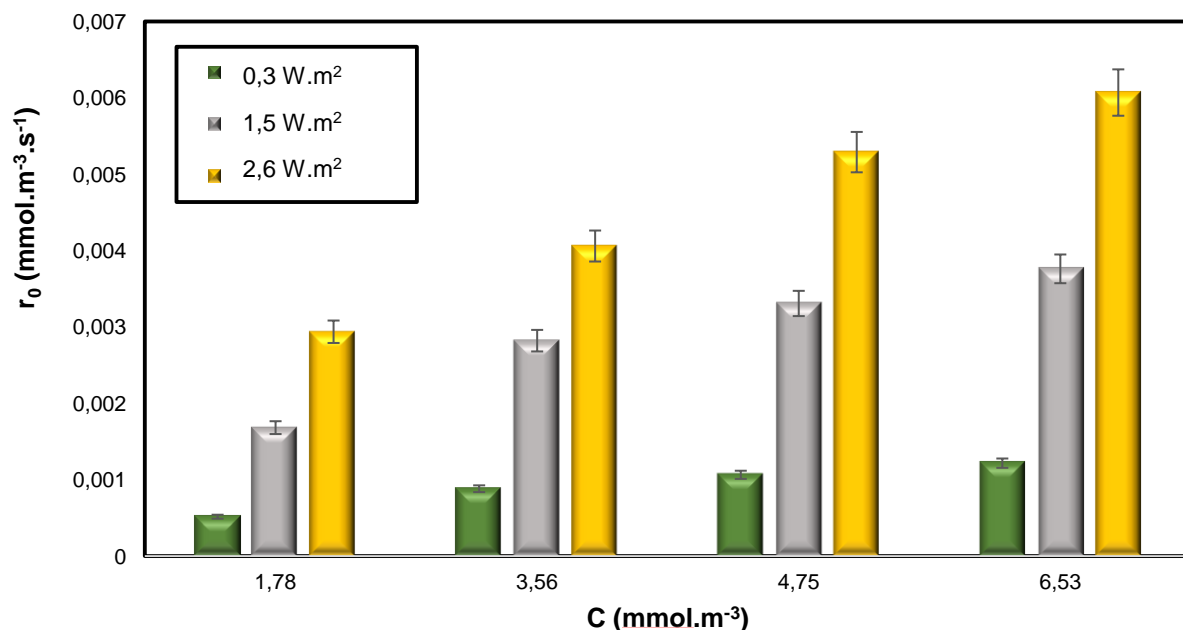
### 305 3.1.2.b. Effect of UV intensity

306 In order to boost the photocatalytic efficiency of the OF media, the light intensity was  
307 studied as a crucial factor in influencing the reaction rate. For this purpose, the  
308 degradation rate was studied for an OF media containing 26 mg.m<sup>-2</sup> of TiO<sub>2</sub>  
309 (maximum mass) under different UV intensities (0.3 W.m<sup>-2</sup>, 1.5 W.m<sup>-2</sup> and 2.6 W.m<sup>-2</sup>)  
310 at different concentrations.

311 The results illustrated in figure 7 show a similar behavior to what has been reported  
312 in the literature regarding the effect of UV intensity, where the photocatalytic  
313 oxidation performance of the OF media improves as the light intensity increases  
314 [44,49]. This can be explained by the fact that increasing the intensity of UV light  
315 creates more photons, which enhance the production of oxidizing species [50–52].

316 In this study, the results of the apparent reaction kinetic constants (Tab. 4) show that  
317 the increase in intensity has overcome the limitation of the photocatalytic oxidation  
318 efficiency caused by the loading of the photocatalytic media with TiO<sub>2</sub>. this is due to  
319 the efficiency of the illumination by *in-situ* UV LED lighting, overcoming the problems  
320 of shading and intensity loss [40].

321 According to the power relation law ( $k_{app} = k_0.I^\alpha$ ). It is also noted that the  
322 photocatalytic reaction rate follows a first order regime with applied irradiation  
323 intensities, where  $\alpha$  is about 0.64 with  $k_0 = 0.0064 \text{ mmol.m}^{-3}.\text{s}^{-1}$ , meaning that the  
324 photogenerated electron-hole pairs are consumed more rapidly by chemical reaction  
325 than by recombination [44][53]. The results reported by some authors [50]  
326 [55][56][33] give a value varying from 0.25 to 0.5 when an external lighting is used.  
327 So, the use of OF seems to be me more efficient [51][57].



**Figure 7:** Initial degradation rate of Cyclohexane with optical fiber media at different concentrations under different UV Intensity (HR = 40 ± 2%, T = 20 ± 2 C°, m<sub>TiO<sub>2</sub></sub> = 26 g.m<sup>-2</sup>)

UV Intensity (W.m <sup>-2</sup> )	0,3	1,5	2,6
k <sub>app</sub> (mmol.m <sup>-3</sup> .s <sup>-1</sup> )	0,0032	0,0083	0,0118
K (m <sup>3</sup> .mmol <sup>-1</sup> )	0,0903	0,1170	0,1284
R <sup>2</sup> (%)	98,36	98,3	97,48

328 **Table 4:** Value of Langmuir-Hinshelwood reaction rate constants (k) and Langmuir  
 329 adsorption constants (K) for the degradation of Cyclohexane under different UV  
 330 Intensity (HR = 40 ± 2%, T = 20 ± 2 C°, m<sub>TiO<sub>2</sub></sub> = 26 g.m<sup>-2</sup>)

331 It should be noted that a summary of the intensification part is highlighted, allowing  
 332 for a review of this study, the details of which are presented in Figure S1.

### 333 3.2. Continuous photoalytic study

334 In order to evaluate the performance of the different front flow photocatalytic reactors  
 335 presented below, many factors have been considered.

336 The overall rate of photocatalytic degradation is an important factor in evaluating  
337 VOC's removal performance. VOC's removal efficiency is defined as:

$$338 \quad RE (\%) = \frac{[VOCs]_{in} - [VOCs]_{out}}{[VOCs]_{in}} 100 \% \quad (\text{Eq. 3})$$

339 The overall rate of photocatalytic degradation of cyclohexane is calculated as:

$$340 \quad r = \left(\frac{Q}{S}\right) \left(\frac{[VOCs]_{in}}{100}\right) RE (\%) \quad (\text{Eq. 4})$$

341 Where  $[VOCs]_{in}$  and  $[VOCs]_{out}$  are respectively the inlet and outlet pollutant  
342 concentration ( $\text{mmol}/\text{m}^3$ ), Q is the volumetric flow rate ( $\text{L}\cdot\text{min}^{-1}$ ) and S the mean  
343 surface of the catalytic support ( $\text{m}^2$ ).

344 The overall selectivity of  $\text{CO}_2$  can be a useful parameter in evaluating the  
345 performance of the photocatalytic reactor with respect to the removal of VOCs. It  
346 makes it possible to estimate the rate of mineralization, that is to say the final  
347 reaction step of the process. The global selectivity of  $\text{CO}_2$  is expressed as follows

$$348 \quad SCO_2 (\%) = \frac{[CO_2]_{out} - [CO_2]_{in}}{N_c RE (\%) [VOCs]_{in}} 10^4 \quad (\text{Eq. 5})$$

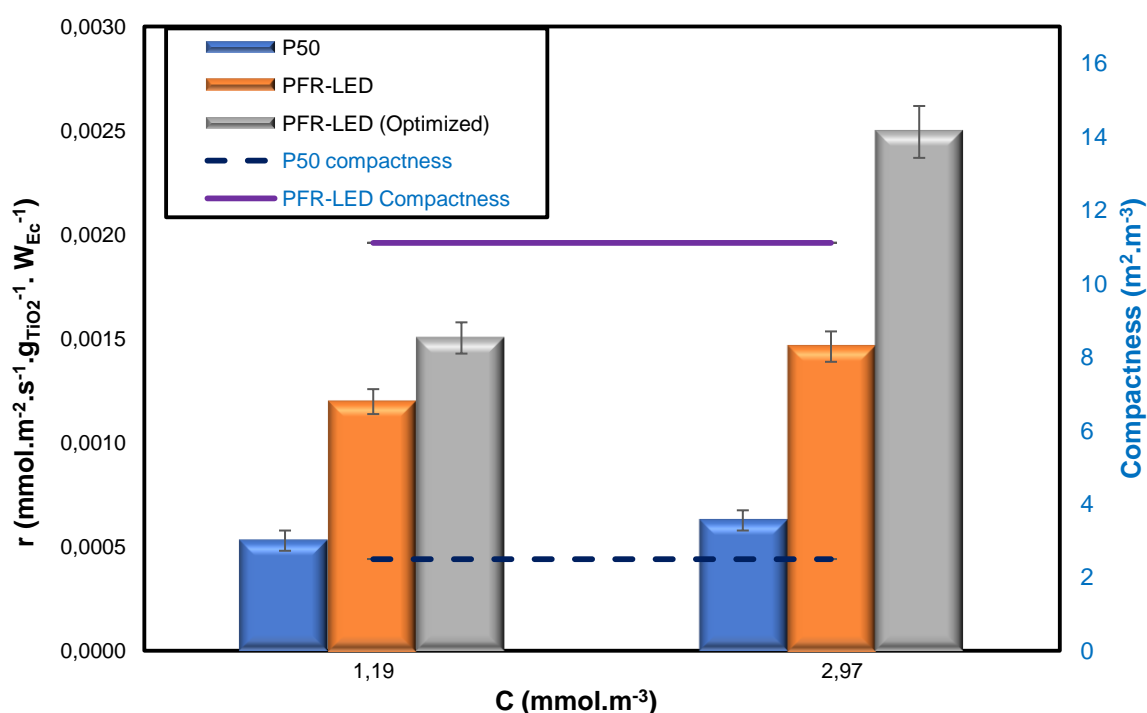
349 Where  $[CO_2]_{in}$  and  $[CO_2]_{out}$  refer respectively to the concentration of carbon dioxide at  
350 the air inlet and outlet ( $\text{mmol}/\text{m}^3$ ). The number  $N_c$  represents the stoichiometric  
351 coefficient of the overall degradation reaction (in our case, it is equal to 6).

352 Another criterion considered was the compactness of the reactor. This is an  
353 important engineering parameter, as it allows to highlight the efficiency of reactor  
354 designs in terms of compactness, which is expressed as the catalytic surface area  
355 per unit volume of the reactor (equation 6).

$$356 \quad Compactness = \frac{Catalytic\ area}{Reactor\ volume} \quad (\text{Eq. 6})$$

357 **3.2.1. Comparison of the photocatalytic reactor configurations**

358 This study focuses on the comparison of the cyclohexane removal efficiencies under  
 359 different inlet concentrations, with the different configurations of front flow  
 360 photocatalytic reactors (P50, PFR-LED and PFR-LED Optimized). Two key factors  
 361 were considered; the reactor compactness, as well as the specific degradation rate,  
 362 expressed in mmol removed per catalyst surface, per unit time, per gram of TiO<sub>2</sub> and  
 363 per unit of electrical power consumed (Ec) (Figure 8).



**Figure 8:** Specific degradation rate and Compactness for the elimination of cyclohexane with P50 ( $m_{\text{TiO}_2}$  0,64 g,  $I$  = 20  $\text{W}\cdot\text{m}^{-2}$ ,  $E_c$  = 31.1 W), PFR-LED ( $m_{\text{TiO}_2}$  = 0,3 g,  $I$  = 4,5  $\text{W}\cdot\text{m}^{-2}$ ,  $E_c$  = 14.5 W), PFR-LED (Optimized) ( $m_{\text{TiO}_2}$  = 0,78 g,  $I$  = 7,8  $\text{W}\cdot\text{m}^{-2}$ ,  $E_c$  = 15.7 W).  $Q$  = 18  $\text{L}\cdot\text{min}^{-1}$ ,  $\text{RH}$  =  $38 \pm 2$  %,  $T$  =  $20 \pm 2$  °C.

364 The results show first of all, that the use of lamps inside the reactor as a light source  
 365 for catalyst activation takes up a lot of space in the reactor. The compacity is 4.5  
 366 times higher than for LED reactors [14]. In addition, the system based on the OF

367 media was clearly more efficient in terms of cyclohexane removal per mass of TiO<sub>2</sub>  
368 and watt consumed. The performance of PFR-LEDs could be explained by the  
369 lighting efficiency as shown on part 3.1 [28][31][30].

370 The reactor based on optic fiber media (UV-LED) provides a better treatment  
371 capacity than a conventional system (P50), where at a certain UV intensity, the *in-situ*  
372 lighting allows a better propagation of the photons, thus allowing the activation of  
373 more sites on the surface of the catalyst [21][58]. OF media allows oxidation  
374 reactions to occur in a large area of the catalyst, thus increasing the reaction rate  
375 [21][59].

376 The results show that the use of a reactor based on OF media allows the interaction  
377 of key features for the photocatalytic reaction to further improve it. In particular the  
378 increase of TiO<sub>2</sub> concentration and UV intensity (PFR-LED Optimized) improve the  
379 photocatalytic capacity of the reactor for the removal of the pollutant. We note also  
380 that at high concentration, and contrary to the simple version of the PFR-LED, the  
381 optimized PFR-LED photocatalytic module allowed a better kinetic degradation of the  
382 pollutant thanks to its large photocatalytic active surface [47][50].

383 An energy gain has been proven in this study especially when passing from the  
384 conventional configuration to the new configuration based on FO media. Indeed, an  
385 improvement about 78% was found, this also highlights the interest of using such a  
386 lighting way of photocatalytic sites (*In-situ* lighting). It is also worth mentioning that a  
387 comparison in terms of consumed UV watt is developed in order to highlight the  
388 energy gain of the process intensification, illustrated in Figure S2.

389 Thus, the process performance of this last pilot will be studied below.

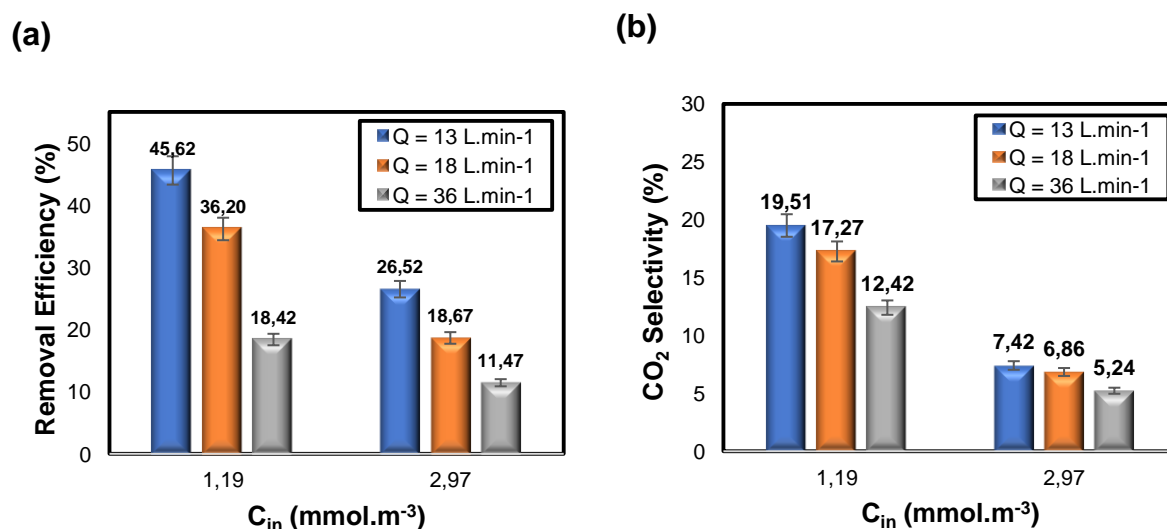
390



391 **3.2.2. Performance of the optimized photocatalytic reactor under simulated real**  
392 **conditions**

393 **3.2.2.a. Inlet concentration and flow rate effect**

394 In this study, two high concentrations were used (1.19 and 2.97 mmol.m<sup>-3</sup>) to  
395 understand their influence on the performance of the new photocatalytic reactor  
396 configuration (PFR-LED Optimized). The removal efficiency of cyclohexane as well  
397 as the dependence of the selectivity on different input concentrations under different  
398 flow rates are presented in Figure 9.



**Figure 9:** Inlet concentration and flow rate effect on (a) removal efficiency and (b) CO<sub>2</sub> selectivity of the PFR-LED (Optimized): m<sub>TiO<sub>2</sub></sub> = 0,78 g, l = 7,8 w/m<sup>2</sup>, HR = 5%, T = 20 ± 2 °C.

399 The results (Fig. 9a) show a similar behavior to what is reported in the literature  
400 [60,61], where at a given flow rate, the degradation rate tends to a limit at higher  
401 concentration of pollutants. This is related to the limitation by the chemical reaction  
402 step due to the unavailability of active sites. It is also recorded that the increase of  
403 the flow rate makes the removal efficiency of cyclohexane decrease, due to lowering  
404 contact time between the pollutant and the active sites [62][63][64][65].

405 Figure 9b, shows the dependence of the selectivity on the inlet concentration of  
406 Cyclohexane at different flow rates. Either the results agree with what was expected,  
407 the influence of the inlet concentration on the CO<sub>2</sub> selectivity is clearly shown. This  
408 decrease with increasing pollutant concentration, due to the fact that there are fewer  
409 active sites available on the surface of the photocatalyst [54] [63], which also  
410 confirms that the process is limited by the availability of active sites. Increasing the  
411 flow rate also leads to a decrease in the rate of mineralization. This is due to the fact  
412 that the contact time is insufficient for the degradation of the by-products. Adding to  
413 this, the competition of the by-products formed towards the active sites reduces the  
414 selectivity [66].

415 Thus, this compact configuration presents excellent performances in terms of  
416 pollutant removal with less energy consumption, compared to the classical  
417 configuration [33].

#### 418 **3.2.2.b. Relative humidity effect**

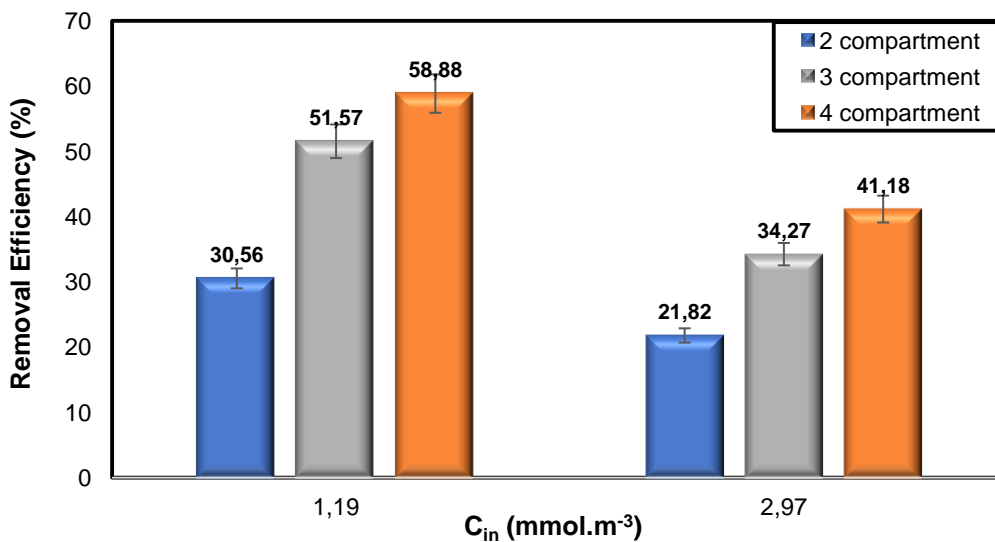
419 The effect of humidity on the photocatalytic performance of the new optimized PFR-  
420 LED reactor was investigated, under a relative humidity range of 5-80% where an  
421 optimum was recorded [67][68][69][70]. The description of this study and the results  
422 are detailed in the information support (Fig S3).

#### 423 **3.2.2.b. Effect of the number of compartments**

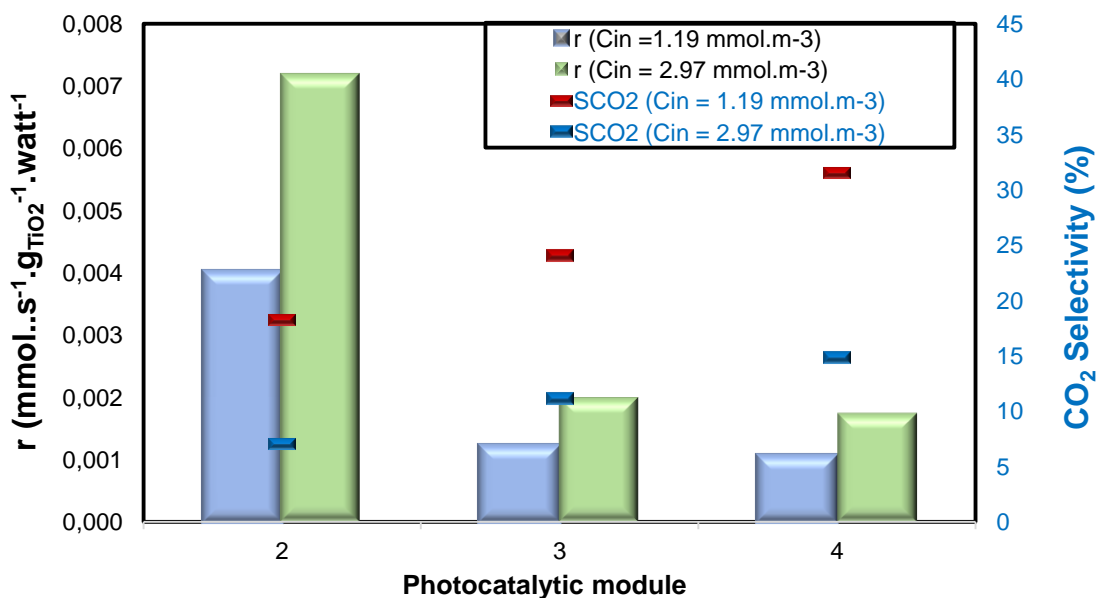
424 In order to further improve the design of the pilot, the effect of the number of  
425 photocatalytic media, i.e. the number of compartments was studied. For this purpose,  
426 several combinations using two to four pairs of optical fiber/UV-LED called: 2OF/UV-  
427 LED, 3OF/3UV-LED, and 4OF/4UV-LED) were installed. The characteristics of the  
428 new optimized configuration are  $m_{\text{TiO}_2} = 26 \text{ g.m}^{-2}$  and UV intensity =  $2.6 \text{ W.m}^{-2}$ . The

429 removal profile of Cyclohexane as a function of the number of photocatalytic media  
 430 (OF/UV-LED), at different input concentrations ( $1.19 \text{ mmol.m}^{-3}$  to  $2.97 \text{ mmol.m}^{-3}$ )  
 431 under the optimal relative humidity condition, as well as  $\text{CO}_2$  selectivity, are  
 432 presented in figure 10.

(a)



(b)



**Figure 10:** Effect of the number of compartments on (a) removal efficiency and (b)

Specific degradation rate and CO<sub>2</sub> selectivity of the PFR-LED (Optimized):  $m_{\text{TiO}_2}$

$$m_{\text{media}} = 26 \text{ g.m}^{-2}, I_{\text{media}} = 2,6 \text{ W.m}^{-2}, Q = 18 \text{ L.min}^{-1}, \text{RH} = 38 \pm 2 \%, T = 20 \pm 2 \text{ C}^\circ$$

433

434 The results in the Fig.10a showed that the oxidation efficiency of cyclohexane  
435 increases with the number of photocatalyst media. This behavior is due to the  
436 creation of more active sites with the increase of the photocatalyst surface. Also, to  
437 the increase of the residence time. The same behavior was previously highlighted by  
438 Abidi et al [23] during the treatment of chloroform by a photocatalytic planar reactor  
439 based on luminous textile.

440 The analysis of the specific degradation rate as well as the CO<sub>2</sub> selectivity rate,  
441 recorded after each addition of photocatalytic media is added, for the two  
442 concentrations studied as shown in Fig. 10b. The results show that the specific  
443 degradation rate decreases with the increase of the number of photocatalytic  
444 mediums. This is due to the fact that the added OF media receives less pollutant  
445 concentration and more by-products, and therefore, the efficiency of the added  
446 photocatalyst cannot be at its highest possible performance level [44]. However, the  
447 increase of OF media represents a real interest for by-products degradation and  
448 leading to better selectivity of the outlet effluent.

#### 449 **4. Conclusions**

450 In this study, an innovative TiO<sub>2</sub> coated OF light fabric was considered for the  
451 optimization of the photocatalytic treatment. Results showed that the light textile  
452 improves the contact between light, catalyst and pollutant and thus the process  
453 efficiency is improved. The intensification of the heterogeneous photocatalysis of  
454 TiO<sub>2</sub> under UV-LED irradiation was approved.

455 This new technology has allowed the development of a new design of frontal flow  
456 photocatalytic reactors, with a more compact configuration. The comparison of their  
457 performances with those of conventional photocatalytic reactors has shown that the  
458 optical fiber technology represents a gain in energy, compactness and photocatalytic  
459 efficiency for the removal of cyclohexane compared to conventional systems.

460 The performance of the optimized version of the front flow photocatalytic reactor  
461 (PFR-LED Optimized) was evaluated, where the effect of inlet concentration, flow  
462 rate, as well as the complex effect of humidity on the photocatalytic efficiency and  
463 mineralization of cyclohexane were shown. In addition, the effect of the number of  
464 photocatalytic compartments on the process was highlighted.

465 Through this study, a new front flow reactor design exhibiting a good energy  
466 efficiency and better compactness was developed. The data of this investigation  
467 respond perfectly with the requirements of the sustainable development policy,  
468 namely affordability and innovation to combat climate change, as stipulated in the  
469 sustainable development goals (GSDs) 7 and 9, which respectively target clean and  
470 affordable energy as well as industry, innovation and infrastructure.

471 In addition, this new compact and optimized version could be a real prospect for anti-  
472 gas filtration equipment, i.e. gas masks or on-board filtration systems.

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476

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