Effective decoloration of cationic dyes by silica gel prepared from Tunisian sands and TiO$_2$/silica gel composites: dual adsorption and photocatalytic processes

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Received 9 March 2019; Accepted 13 October 2019

ABSTRACT

Silica gels synthesized from Tunisian sands employed as alternative low-cost adsorbents, and silica gel/TiO$_2$ composites for combined absorption/photocatalytic decoloration of methylene blue (MB) dye solutions, were studied. The silica gel is characterized by a high specific surface area of up to $194 \text{ m}^2/\text{g}$ and is likely to increase in aqueous solution, according to the solid/liquid ratio which modulates the degree of hydration. This was determined at various synthesis pH values. For this silica gel, the maximum adsorption capacity (up to 91%, 125 mg/g) was obtained in acidic medium (pH 3). The adsorption mechanism fitted better using the Langmuir model, and the adsorption kinetics of the dye on these materials was well described by the second-order model. Silica gel/TiO$_2$ demonstrated an effective degradation of MB the first stage (30 min without UV-light exposure) and under UV. The kinetics of discoloration of MB followed a pseudo-first-order rate law. We can remark that 5 h of UV irradiation was enough to achieve 99% discoloration of the MB. The findings demonstrated the applicability of this silica gel/TiO$_2$ catalyst for the photocatalytic oxidation of MB.

Keywords: Silica gel; Titania; Cationic dye; Adsorption; Photocatalysis

1. Introduction

Currently, dyes play an important role in the industrial sector. These are used in the paper, cosmetics, food processing, pharmaceutical and especially in the textile industry. According to their chemical structure and method of application to the various substrates (textiles, paper, leather, plastics, etc.), there are different classes of dyes such as azo, reactive, acidic, basic, neutral, anthraquinone, disperse and direct. Azo and anthraquinone dyes are, however, the most used in the textile industry [1–3]. Azo dyes, in particular, are the most commonly used in the textile industry, such as methylene blue (MB = C$_{16}$H$_{18}$ClN$_3$S), which is a cationic dye. This dye poses a real danger when released into wastewater, since it may cause health issues such as burning eyes, and other harmful environmental effects such as blocking photosynthesis of waterborne flora [3–5]. The removal of this dye from wastewaters consists currently of distinct technological approaches, such as membrane filtration [6], coagulation–flocculation [7], chemical oxidation, electrochemical processes and adsorption [8,9]. The last one is a nondestructive process, by which the contaminants
can be transferred from wastewater to an adsorbent [10,11].
Currently, photocatalytic decoloration is one of the most promising technologies for the removal of organic pollutants
[12–14]. Titanium dioxide (TiO$_2$) is the common photocatalyst
used [15] since it possesses interesting features such as high
degradation efficiency, stability and low production cost
[16–18].

Instead of using suspended photocatalytic particles,
which must subsequently be removed/filtered from the
treated water, the use of supported TiO$_2$/systems is very
attractive. The use of silica gel as a substrate might assure
high surface area, while it is hydrophobic and relatively
cheap. Additionally, its high adsorption capacity might also
contribute to the retention/immobilization of dye molecules
[19,20]. Due to those properties, this material is widely used
in fillers [21], coatings [22], catalyst supports and hygroscopic
packaging for optical [23], electronic [24] and pharmaceutical

Several studies have investigated the photocatalytic deg-
radation of organic pollutants based on TiO$_2$ suspended in
wastewater [26–31]. In this work, we study the dual effects
(molar ratios at 1,060°C a mixture of sodium carbonate and silica sand
of sodium silicates, (ii) the dissolution, and (iii) the forma-
tion of organic pollutants based on TiO$_2$ products.

2. Materials and methods

2.1. Materials
The silica gel (GM) used in this study was prepared from
Tunisian sands, which have been characterized in our previ-
ous work [5]. These materials were collected from the Fortuna
formation in the North of Tunisia. The TiO$_2$ used was a
commercial titania (Aeroxide P25, Evonik, Aveiro, Portugal).

2.2. Silica gel and silica gel/TiO$_2$ preparation
Silica gels were produced by Tunisian sand using three
steps as proposed by Lazaar et al. [5]: (i) the preparation
of sodium silicates, (ii) the dissolution, and (iii) the forma-
tion. The intermediate compound was obtained by heating
at 1,060°C a mixture of sodium carbonate and silica sand
(molar ratios $n = \text{SiO}_2/\text{Na}_2\text{O}$ equal to 1.5) [32,33]. 10 g of this
prepared amorphous sodium silicate was dissolved in 500 mL
of distilled water (at 160°C) with stirring for 10 min to obtain
hydrated sodium silicate [32,34]. The final product (silica gel)
was extracted after the dropwise addition of 2 M hydrochlo-
ric acid solution (pH 3), with washing to eliminate Cl
ions
[33,35], and drying for 24 h at 60°C. In this work, silica gels
(GM) were prepared under different pH values (3, 6 and 10)
from silicates, which were denoted as GM3, GM6, and GM10,
respectively (Table 1).

<table>
<thead>
<tr>
<th>Samples</th>
<th>pH</th>
<th>Molar ratio SiO$_2$/Na$_2$O</th>
<th>$S_{\text{BET}}$ (m$^2$/g)</th>
<th>Pore volume (cm$^3$/g)</th>
<th>Pore diameter (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>GM3</td>
<td>3.0</td>
<td>1.5</td>
<td>194</td>
<td>0.53</td>
<td>111</td>
</tr>
<tr>
<td>GM6</td>
<td>6.0</td>
<td>1.5</td>
<td>91</td>
<td>0.23</td>
<td>79</td>
</tr>
<tr>
<td>GM10</td>
<td>10.0</td>
<td>1.5</td>
<td>33</td>
<td>0.12</td>
<td>129</td>
</tr>
</tbody>
</table>
where \( C_i \) is the initial concentration of MB (mg/L), \( C_e \) is the concentration of the MB at equilibrium (mg/L), \( q_e \) is the adsorbed quantity expressed as mg of solute per gram of adsorbent, \( V \) (ml) in the volume of the solute; \( W \) (g) is the mass of adsorbent.

The Langmuir and Freundlich models are often used for determining the maximum capacity of the binding of pollutants, and identification of the adsorption type. These two models allowed us to calculate the maximum adsorption capacity, as well as the parameters of adsorption [38].

The Langmuir equation may be expressed as follows:

\[
t/q_e = (1/q_m) + 1/(K_L q_m C_0)
\]

where \( q_m \) (mg/g) is the maximum amount adsorbed, \( q_e \) (mg/g) is the amount adsorbed at equilibrium, \( K_L \) (L/mg) is the Langmuir constant and \( 1/K_L \) is the slope of the plot. These two parameters \( (q_m, K_L) \) are evaluated, respectively, starting from the intersection with the ordinate at the beginning and the slope of the right-hand side \( C_e/q_e = f(C_e) \).

The Freundlich model is described by the following equation (4):

\[
\log q_e = \log K_f + (1/n) \log C_e
\]

where \( q_e \) is the amount of dye in mg absorbed per unit g of absorbent at equilibrium, \( C_e \) (mg/L) is the concentration of dye solution at equilibrium, \( K_f \) is the Freundlich constant and \( n \) is a parameter which represents the absence of linearity of the adsorbed quantity as a function of \( C_e \).

In the case of Langmuir models, to know if favorable or unfavorable adsorption is occurring, a factor of separation \( R_L \) is used [14]. This factor is defined by the following relation [39]:

\[
R_L = 1 + K_L C_0/n
\]

where \( K_L \) is the Langmuir constant (l mg\(^{-1}\)) and \( C_0 \) is the initial concentration of the dye (mg L\(^{-1}\)).

The adsorption kinetics was evaluated following pseudo-first and second-order equations.

The pseudo-first-order equation [40] is given by:

\[
\log (q_e - q_t) = \log q_e - (k_1/2.303)t
\]

where \( q_e \) (mg/g) is the capacity at adsorption equilibrium, \( q_t \) (mg/g) is the quantity of adsorbent at the time \( t \) (min) and \( k_1 \) is the equilibrium rate constant (min\(^{-1}\)).

The pseudo-second-order kinetics may be expressed as [40]:

\[
t/q_t = 1/k_2 q_e^2 + t/q_e
\]

where \( q_e \) (mg/g) is the capacity at adsorption equilibrium, and \( k_2 \) (g mol\(^{-1}\) min\(^{-1}\)) is the constant speed of adsorption.

2.5. Photocatalytic experiments

The photocatalytic effect of the TiO\(_2\)/silica gel composites was studied by testing the degradation of MB at room temperature. 0.005 g of photocatalyst was suspended in 400 mL of 10 mg/L MB solution in a beaker. In the preliminary experiments, reactants were premixed in the dark for 30 min to allow adsorption. After that, UV germicidal lamps (Philips PL-S 9W, NL) (Geobiotec, Geosciences Dept, University of Aveiro, Portugal) were switched on to initiate the photocatalytic decoloration reaction. UV irradiance was a 13 W m\(^{-2}\). At an interval of 1 h, 15 mL samples were drawn out of the reactor to determine the change of the MB concentration, by measuring the absorbance on a UV-Vis spectrometer (PerkinElmer lambda 25) (Department of Materials and Ceramic Engineering, University of Aveiro, Portugal). The photocatalytic decoloration efficiency \( \xi \) (%) was measured as:

\[
\xi \% = (1 - C_t/C_0) \times 100
\]

where \( C_t \) is the initial concentration for the calculation of the photocatalytic decoloration efficiency (mg/L) at 0 min and after a certain irradiation time \( t \) (min), respectively. The plot of ln \( (C_t/C) \) vs. the contact time \( t \) under UV exposure, gives a straight line, whose slope indicates the value of the pseudo-first-order apparent rate constant \( k'_{\text{app}} \) [14,41].

3. Results and discussion

3.1. Characterization of silica gels and silica gel/TiO\(_2\)

According to the X-ray diffraction (XRD) diagrams obtained (Fig. 1), the silica gel has a mineralogical composition virtually identical irrespective of pH of synthesis (Fig. 1a). They are characterized by a large amorphous hump centered at around 20 = 23\(^\circ\), indicating their accentuated amorphous nature, with crystalline peaks of quartz, and in the case of GM10, a very small amount of poorly crystalline feldspar. The XRD patterns of pure TiO\(_2\) are shown in Fig. 1b and the TiO\(_2\) P25 powder consists of mostly anatase, with some secondary rutile phase.

Fig. 2 shows SEM images of silica gel GM3 and the silica gel/titania composite (GM3)/TiO\(_2\) (20/80). The silica gels (Fig. 2a) present similar structures with large particle aggregates of 1.14 to 5.45 μm. Almost the same results were found by Brahim et al. [33] for the size of silica gel prepared from Algerian siliceous sands. The SEM image (Fig. 2b) of the silica gel/TiO\(_2\) composite showed that TiO\(_2\) nanoparticles were well immobilized on the adsorbent; some of them were agglomerated on the surface of the adsorbent.

The BET specific surface area (SSA) of the silica gels are reported in Table 1. The values of the SSA of products prepared at pH 3, pH 6, and pH 10 were 194, 91, and 33 m\(^2\)/g, respectively. Hence, silica gel was prepared at pH 3 (GM3) had a higher surface area compared with the silica gel were synthesized at pH 6 (GM6) and pH 10 (GM10). These results
Fig. 1. X-ray diffractogram of (a) silica gel: Qz, quartz and Fd, Feldspar; (b) TiO$_2$ P25.

Fig. 2. SEM images of (a) silica gel (GM3) and (b) silica gel (GM3)/TiO$_2$ (Ti80).
are a good agreement with previous results in the literature [42]. The nitrogen adsorption–desorption isotherms of the silica gels are shown in Fig. 3, and they have a type II adsorption isotherm. They present pore volumes varying from 0.12 to 0.53 cm$^3$/g (Table 1) and pore diameters ranging from 79 to 129 Å, the largest values being for GM3, this indicates that our silica gel is a mesoporous material, with the pore diameter exceeding 20 Å [43].

3.2. Adsorption experiments

3.2.1. MB adsorption performance

The effect of pH on the adsorption of MB onto silica gels (pH 3, 6, and 10) is shown in Fig. 4. The results obtained show that the time required for equilibrium is reached after 240 min reaction time, but that the adsorption begins to plateau after 120 min. The removal efficiency for GM3 exceeded 81% after 240 min, compared to 66% and 49% for GM6 and GM10. Therefore, the retention of MB by the silica gel is favorable when synthesized at acid pH (pH 3), and decreases when synthesized at pH 6 and 10. As GM3 showed superior MB adsorption, this silica gel was used for subsequent experiments.

The increased surface area of GM3 will have an effect on this adsorption. However, the influence of synthesis pH on the adsorption of MB may also partly be due to the surface charge of the adsorbents [5,44,45].

Fig. 5 shows the effect of GM3 of varying the initial concentration of MB (5–25 mg/L) on the adsorption. The results show that the MB adsorption capacity increases with the decrease in the initial concentration of dye, as would be expected. The adsorption rate of the GM3 exceeded 91% even after 180 min reaction time using just 5 mg/L (Fig. 5). When this initial concentration dye was elevated to 25 mg/L, the adsorption rate of the adsorbent GM3 decreased to 41%, due to the saturation of the active sites in the adsorbent [5,46]. Even if the adsorption is supposed to follow a multilayer mechanism, at high concentrations of dye greater competition between the MB molecules occurs.

3.2.2. Equilibrium and kinetics of MB adsorption

The calculated Langmuir and Freundlich parameters are presented in Fig. 6 and Table 2. The experimental data fit better with the Langmuir adsorption isotherm model, as the values of coefficients of determination ($R^2$) are closer to 1.

Fig. 4. Adsorption kinetics: effect of pH of silica gel synthesis on subsequent adsorption of MB. Experimental conditions: $C_i$, 10 ppm; $T$, 25°C; pH, 3; 6 and 10; $m_{adsorbent}$, 0.01 g.

Fig. 5. Adsorption kinetics: effect of initial concentration of MB in dye solution on adsorption of MB. Experimental conditions: $T$, 25°C; synthesis pH, 3; $m_{adsorbent}$, 0.01 g; time of equilibrium, 180 min.
(Fig. 6a), indicating that the adsorption process corresponds to monolayer coverage of MB molecules over the surface of the samples analyzed. The results indicated that higher adsorption capacity \( (q_m) \) was achieved by GM3 (125 mg/g) (Table 2) compared to those obtained by GM6 (91 mg/g) and GM10 (74 mg/g). GM3 also having the higher SSA. Our results indicated that \( R_L \) ranged between 0 and 1 for the adsorption of MB on GM3, indicating favorable adsorption.

For the Freundlich model, the measured parameters are summarised in Table 2 and Fig. 6b. A lower affinity for interaction between the adsorbate–adsorbent was revealed by the smaller \( R^2 \) regression coefficient. The above fact shows a monolayer adsorption with a heterogeneous energetic distribution of active sites, accompanied by interactions between the adsorbed molecules.

The parameters calculated for the various kinetic models (pseudo-first-order and pseudo-second-order) are presented in Figs. 6c and d. A much better fit \( (R^2 = 0.99) \) was described by the pseudo-second-order model. Moreover, the calculated adsorption capacity at equilibrium \( (q_{e,cal}) \) is very close

Fig. 6. Fitting with the linear Langmuir (a) and Freundlich (b) models for MB adsorption on GM3. Experimental conditions: \( C_i \), 10 ppm; \( T \), 25°C; synthesis pH, 3; \( m_{adsorbent} \), 0.01 g; time, 180 min. Kinetics by the pseudo-first-order (c) and pseudo-second-order (d) models for MB adsorption on GM3.

<table>
<thead>
<tr>
<th>Material</th>
<th>( q_m ) (mg/g)</th>
<th>( K_L ) (L/mg)</th>
<th>( R_L )</th>
<th>( R^2 )</th>
<th>( K_f ) (L/g)</th>
<th>( n )</th>
<th>( R^2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>GM3</td>
<td>125</td>
<td>1,000</td>
<td>0.00091</td>
<td>0.993</td>
<td>112.72</td>
<td>1.03</td>
<td>0.988</td>
</tr>
</tbody>
</table>

Table 2
Langmuir and Freundlich parameters for MB adsorption

<table>
<thead>
<tr>
<th>Material</th>
<th>( q_e ) (mg/g)</th>
<th>( k_1 ) (min(^{-1}))</th>
<th>( R^2 )</th>
<th>( q_e ) (exp)</th>
<th>( k_2 ) (g/mg min)</th>
<th>( q_e ) (mg/g)</th>
<th>( R^2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>GM3</td>
<td>–0.018</td>
<td>142.4</td>
<td>0.858</td>
<td>239.6</td>
<td>0.00038</td>
<td>250.0</td>
<td>0.990</td>
</tr>
</tbody>
</table>

Table 3
Pseudo-first and second-order parameters for MB adsorption
to the value obtained experimentally \( (q_{ex,cal}) \) (Table 3). Thus, the adsorption of MB on silica gel might be explained by the pseudo-second-order kinetic model. These results suggest a chemisorption mechanism, involving covalent forces through the sharing or exchange of electrons between sorbent and sorbate.

### 3.3. Photocatalytic decoloration of MB

#### 3.3.1. Photocatalytic decoloration efficiency

The silica gel/titania P25 can have the added photocatalytic properties from titania P25, the high stability from silica gel and extra properties coming from chemical bonds between two composites. Table 4, Figs. 7 and 8 show the results of photo-induced removal of cationic MB dye solution (10 mg/L) by GM3, GM6 and GM10 silica gel/titania P25 composites, having different proportions of TiO\(_2\). In darkness, the adsorption of MB on the silica gel/TiO\(_2\) increased with increasing the proportion of silica gel (Fig. 7). This explains the lowest photodecolouration correlation coefficient \( R^2 \) observed in Ti20 samples (Table 2). Under UV irradiation (after 5 h of reaction), the MB decolouration of pure TiO\(_2\) (99%) is slightly superior to that of the silica gel/TiO\(_2\) composites (Ti80 (93%), Ti50 (91%), and Ti20 (88%)) (Fig. 7), as they have less titania present, although they have broadly similar. However, one advantage of dispersion of TiO\(_2\) nanoparticles onto the adsorbent gel is that it gives a good adherence between the support and the catalyst over a sustained period, without adversely affecting its photocatalytic [15,47].

The results indicate that the synthesized photocatalyst can effectively degrade the MB. The photocatalytic activity of silica gel samples was increased by the increase in TiO\(_2\) loading. Fig. 7 also indicates that the photocatalytic activity of pure TiO\(_2\) P25 is higher than that of the silica gel/TiO\(_2\) samples. This result could be due to the poor accessibility to the TiO\(_2\) P25 surface. However, the silica gel/TiO\(_2\) P25 samples can be readily separated from the suspension after the reaction because they sediment in minutes when the stirring was stopped, while the TiO\(_2\) sample could not sediment easily, and lead to potential difficulty in downstream separation.

### Table 4

Photocatalytic decoloration efficiency (ξ), and correlation coefficient \( R^2 \) of the pseudo-first-order apparent constant of the photocatalytic reaction (Ti = 100% TiO\(_2\))

<table>
<thead>
<tr>
<th>Material</th>
<th>( R^2 )</th>
<th>Ti</th>
<th>Ti80</th>
<th>Ti50</th>
<th>Ti20</th>
<th>Ti</th>
<th>Ti80</th>
<th>Ti50</th>
<th>Ti20</th>
</tr>
</thead>
<tbody>
<tr>
<td>GM3</td>
<td>0.97</td>
<td>0.93</td>
<td>0.87</td>
<td>0.74</td>
<td>99</td>
<td>93</td>
<td>91</td>
<td>88</td>
<td></td>
</tr>
<tr>
<td>GM6</td>
<td>0.97</td>
<td>0.90</td>
<td>0.85</td>
<td>0.71</td>
<td>99</td>
<td>83</td>
<td>79</td>
<td>73</td>
<td></td>
</tr>
<tr>
<td>GM10</td>
<td>0.97</td>
<td>0.89</td>
<td>0.83</td>
<td>0.67</td>
<td>99</td>
<td>76</td>
<td>72</td>
<td>67</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 8. Photo-induced decoloration of dye solution (MB) by the silica gel/titania P25 composites as a function of time.
3.4. Mechanism of MB decoloration process

For MB adsorption, it was clear that silica gel GM3 shows the highest adsorption capacity (81%) due to the larger surface area. The adsorption capacity of these silica gel materials increased to 83% when reducing the concentration MB to 5 ppm. The percentage of decoloration was influenced by the change in pH. The discoloration of the dye was upgraded to 49% for the GM at pH 10. The isotherms of the adsorption of the MB dye on silica gel by the Langmuir isotherm model parameters were very favorable ($R^2$ up to 0.99), and this reveals that the adsorption process corresponds to monolayer coverage of MB molecules over the surface of the silica gel samples.

With the combined effects of adsorption and photocatalytic decoloration, very satisfactory decoloring efficiency was presented with mixtures, even those containing low TiO$_2$ quantity. Silica gel/TiO$_2$ composites at basic pH show lower decoloring performance (ξ equal to 67%), and this could be related to the inferior SSA of the silica gel.

### 3.3.2. Kinetics of MB decoloration

Adsorption tests in the dark were carried out to evaluate the adsorbed quantities of MB on the photocatalyst surface. Fig. 7 indicates that the adsorbed quantities do not exceed 73% of the initial quantity in the solution. The adsorbed quantities of MB decrease with the augmentation of TiO$_2$ content. This result can be explained by the availability of more adsorption sites at the surface of the composite silica gel/TiO$_2$.

The plot of ln($C_t$/C$_0$) vs. $t$ with different amounts of silica gel (GM3)/TiO$_2$ is shown in Table 5 and Fig. 9. All the curves showed a good linear correlation ($R^2$ > 0.99 for silica gel/TiO$_2$), suggesting that the decoloration of MB by silica gel-TiO$_2$/UV followed a first-order kinetic.

Under the same experimental conditions, the apparent rate constant ($k'_{app}$) of MB decoloration was 0.406, 0.269, 0.236, and 0.158 min$^{-1}$, while the percentage (%) of TiO$_2$ was 100, 80, 50, and 20, respectively. As the mass of TiO$_2$ (100%) increases, the apparent rate constant increases.

<table>
<thead>
<tr>
<th>TiO$_2$ (%)</th>
<th>$k'_{app}$ (min$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>0.406</td>
</tr>
<tr>
<td>80</td>
<td>0.269</td>
</tr>
<tr>
<td>50</td>
<td>0.236</td>
</tr>
<tr>
<td>20</td>
<td>0.158</td>
</tr>
</tbody>
</table>

4. Conclusions

In this work, it has been studied the used to a local natural Tunisian sand for the preparation of silica gels and their potential retention of a cationic dye (MB). Based on the results obtained, the following conclusions can be drawn:

- For MB adsorption, it was clear that silica gel GM3 shows the highest adsorption capacity (81%).
- The absorption efficiency of these silica gel materials increased to 83% when reducing the concentration MB to 5 mg/L.
- The isotherms of the adsorption of the MB dye on silica gel by the Langmuir isotherm model parameters were very favorable ($R^2$ up to 0.99).
- The pseudo-second-order expression ($R^2$ equals to 0.99) is better fitted to describe the adsorption kinetics of analyzed samples.
- The used of mixtures made by different reports of silica gel/P25 TiO$_2$ prepared to ensure photocatalytic properties. The high photocatalytic decoloration rate of MB of silica gel/TiO$_2$ composite (even with a mixture containing low TiO$_2$ contents) is attributed to the excellent adsorption effect in the dark and high-efficiency photocatalytic decoloration (99% removal after 300 min) in the presence of UV light irradiation at acidic pH.
- The results were found that these photocatalysts can effectively degrade the MB.
- The high photodegradation of MB by silica gel/TiO$_2$ is due to the anatase phase stability of the used photocatalyst.
- A kinetic study was performed and showed that the discoloration of dyes followed a pseudo-first-order rate law.
- This study demonstrated the applicability of silica gel/ TiO$_2$ P25 for catalytic treatment of MB.

Acknowledgments

This work was supported by FCT grant SFRH/BPD/72398/2010 and by UID/GE004035/2013 project. This study was supported by funding from MEDYNA: “FP7-Marie Curie Action funded under Grant Agreement PIRSES-GA-2013-612572”, and the Tunisian Belgium Wallonie-Bruxelles International WBI Research Project “Valorisation des Argiles tunisiennes”. Robert C. Pullar acknowledges the support...
of the Fundação para a Ciência e Tecnologia (FCT) grant IF/00681/2015, and this work was developed within the scope of the FCT project CICECO-Aveiro Institute of Materials (FCT UID/CTM/50011/2019), financed by national funds through the FCT/MCTES.

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