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CHARACTERISTICS OF CURCUMIN DYE USED AS A SENSITIZER IN DYE-SENSITIZED SOLAR CELLS

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Abstract. *Dye-sensitized solar cells are the closest mankind has come to replicating nature's photosynthesis. The type of a dye influences the efficiency of these cells. In this paper we studied curcumin dye as a sensitizer in dye-sensitized solar cells and compared it with most often used cyanidin. The results have shown that curcumin has higher efficiency and higher absorption in the visible part of the spectrum compared to cyanidin. Simulation models of dye molecules, curcumin and cyanidin, are deprotonated upon adsorption on the titanium dioxide surface. The energy levels obtained from the calculation indicate a higher probability of electron transition from molecule to titanium dioxide surface in case of curcumin than in case of cyanidin. Based on these results, we concluded that curcumin dye has better properties as sensitizer in dye-sensitized solar cells.*

Key words: *solar cells, curcumin, cyanidin, titanium dioxide, density functional theory, voltage-controlled resistance*

1. INTRODUCTION

A solar cell is a renewable source of energy that directly converts visible light into electricity [2-4]. When exposed to light, the solar cell becomes the source of direct current. Operation principle of all solar cells is based on photoelectric effect. There are first, second and third generations of solar cells. Dye-sensitized solar cells (DSSC) belong to the third generation. The major part of these cells is the nanoparticle anatase titanium dioxide coated with dye molecules. The type of the dye, the way it anchored to the TiO_2, directly affects the efficiency. Ruthenium polypyridyl complexes are known as the most efficient pigments, they achieved almost 12% efficiency [5]. However, these pigments contain a heavy metal which has undesired environmental impact. Cheaper alternative can be given by natural pigments, such as anthocyanins, betalains, chlorophyll, etc. Betalains are recorded as most efficient natural pigments achieving more than 2% [6]. Anthocyanins

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are very frequent in research papers that study natural pigments as sensitizers in DSSC [7]. They give different sensitizing performances from various plants, absorb light at the longest wavelength and have widespread availability [8]. Wongcharee et al. used extracts from rosella and blue pea flowers. Solar cells sensitized by rosella (delphinidin and cyanidin) have been reported to achieve efficiency up to 0.37%, whereas extract from blue pea (ternatin) can achieve up to 0.05% [9]. Tekerek et al. fabricated a solar cell also with rosella dye and compared it to black raspberry and black carrot dyes. They achieved efficiencies of 0.16%, 0.16% and 0.25%, respectively [10]. Curcumin can also be a sensitizer, but it has not attracted significant research attention. Kim et al. reported a dye-sensitized solar cell sensitized with curcumin dye, and showed 0.36% efficiency [11]. In this work we investigate two types of natural pigments: cyanidin extracted from raspberries and curcumin dye extracted from Curcuma longa.

The aim of this paper is to both experimentally and theoretically (simulation) confirm the thesis that curcumin is a better sensitizer in dye-sensitized solar cells than cyanidin. Firstly, we measured Current-Voltage characteristics and absorption spectrum. After that, to confirm the experimental results, we simulated the models of anatase $(TiO_2)_{16}$ cluster and cyanidin or curcumin molecule attached to it. Our calculation is based on density functional theory (DFT) and time-dependent density functional theory (TDDFT). Calculations were carried out with NWChem software [12].

2. Operation Principle of DSSCs

The main idea of dye-sensitized solar cells is to separate the light absorption process from charge collection process by using dye sensitizer with semiconductor. This process imitates the natural light harvesting procedure in photosynthesis [13]. That is why dye-sensitized solar cells are the closest mankind has ever come to replicate nature's photosynthesis. To separate these two processes we could use semiconductor with wide band gap such as titanium dioxide (TiO_2). A dye-sensitized solar cell is composed of photoactive electrode, electrolyte and counter electrode. Photoactive electrode is made of porous nanocrystalline anatase titanium dioxide deposited on FTO conducting glass (Fluorine doped Tin Oxide). FTO layer is 220 nm thick and it is deposited on the glass. It enables transport of photo-generated charge carriers to the electrode and it is also transparent so the light can penetrate into the solar cell. Dye is absorbed on TiO_2 layer to complete the photoactive electrode. Counter electrode is also FTO glass, but it is deposited with platinum to increase the conductivity. The space between electrodes is fulfilled with electrolyte which is based on iodide and triiodide ions (Fig. 1).

When sunlight passes through the photoactive electrode, molecules of the dye absorb the photons and electrons go from the HOMO (highest occupied molecular orbital) in the ground state to the LUMO (lowest unoccupied molecular orbital) in the excited state. Some of the excited electrons have enough energy to jump to the conduction band of titanium dioxide and then to diffuse to the electrode. Dye molecules that lost electrons are oxidized. Electrolyte gives electrons to replace the lost ones. After that, iodide molecules are oxidized. Electrons from photoactive electrode flow through an external load to counter electrode and recombine with electrolyte, thus completing the circuit. Hence, the operating mechanism of dye-sensitized solar cell generates electricity without irreversible

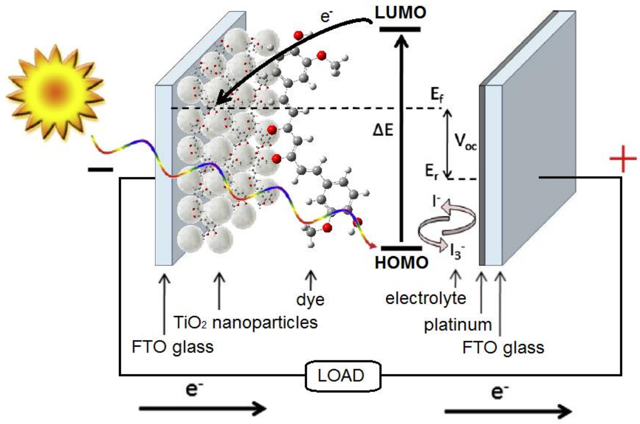


Fig. 1 Schematic structure and principle of operation of DSSC.

chemical changes in the cell. Dye molecules play a key role in producing electricity. They need to overcome small absorption of titanium dioxide by absorbing the photon and exciting the electron. Therefore, they are increasing the efficiency of solar cell. Thus, the greater absorption of the dye is, the more efficient the solar cell will be.

3. Dye Sensitizers

A dye sensitizer absorbs energy in dye-sensitized solar cell. When using natural pigments as a dye-sensitizer, a big problem is the degradation during prolonged exposure to sunlight due to UV radiation. Figure 2 shows optimized molecular structures of the cyanidin and curcumin.

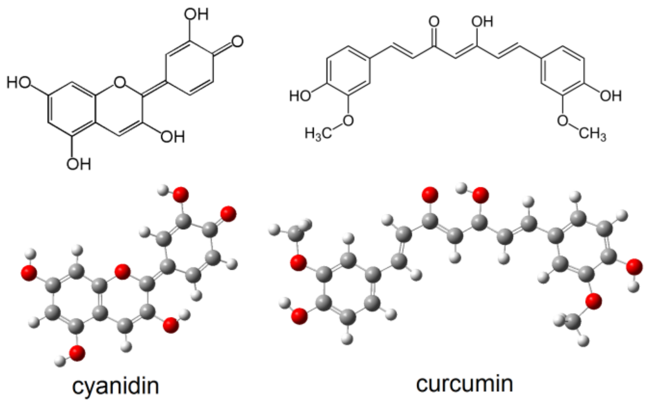


Fig. 2 Optimized molecular structures of the cyanidin and curcumin.

Anthocyanins are widespread water-soluble pigments that can be found in many flowers, fruits and leaves of angiosperms. They are responsible for different colours (red, blue and violet) depending on the pH value [14]. They have found new application in dye-sensitized solar cells because they have significant absorption in the visible part of the spectrum. Only organic dyes that contain several *=O* or *-OH* groups (for example cyanidin found in raspberry) capable of chelating to TiO_2 can be used as dye sensitizer.

Curcumin is an active ingredient of Turmeric (Curcuma longa). Turmeric is a rhizomatous herbaceous perennial plant of the ginger family. It is used for Indian spice, it has yellow color and is known as E100 (food additives). Curcumin can exist in two tautomeric forms (keto – solid and enol - solution). A molecule of curcumin has carbonyl and hydroxyl groups which can bind to TiO_2 surface.

4. Models and Computational Details

We used $(TiO_2)_{16}$ cluster, to model anatase TiO_2 slab. Cluster is obtained by correct "cutting" of anatase slab (Fig. 3). For proper cutting, three conditions must be fulfilled: all titanium atoms must be coordinated to at least four oxygen atoms, all oxygen atoms must be coordinated to at least two titanium atoms, and the ratio of the number of titanium and oxygen atoms in the cluster must be 1 : 2 [15]. After optimization band gap of the $(TiO_2)_{16}$ cluster was 4.52 eV.

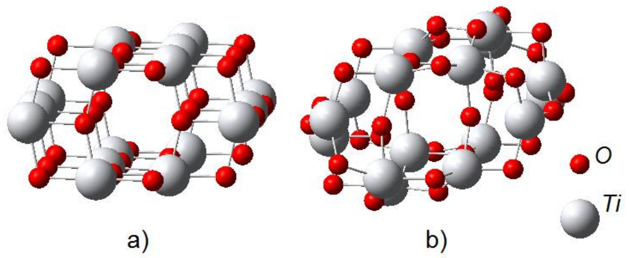


Fig. 3 Model of anatase $(TiO_2)_{16}$ cluster before and after optimization.

For all calculations a freely accessible software NWChem was used, performing density functional theory and time-dependent density functional theory for which we used B3LYP functional together with 6-31G basis set. Density functional theory is a powerful tool for solving multi-stage problems in quantum mechanics. It allows the complicated N-electron wave function and its associated Schrodinger equation to be replaced by much simpler single-electron equations in which the electron density is determined. We used DFT to calculate band gap, HOMOs and LUMOs for all structures and TDDFT to calculate absorption spectra of molecules.

5. Fabrication of DSSCs

Fabrication of dye-sensitized solar cell requires a preparation of titanium dioxide film, extraction of natural pigments, electrolyte preparation and solar cell assembly [16].

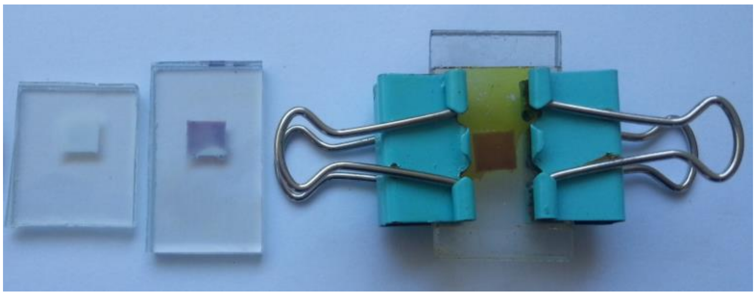
5.1. Preparation of TiO_2 film

A nanoparticle powder TiO_2 (P25 Degussa) was used to prepare the films. Water and acetic acid have been added due to the contribution to the mechanical properties of the films, i.e. good adhesion to the substrate and preventing the formation of cracks. Terpineol is added to prevent particle growth, ethyl cellulose to achieve porosity of the films due to decomposition during thermal annealing. The films were deposited with a doctor-blade technique on an FTO glass. Doctor-blade technique is process of paste deposition on some surface by a razor blade, while the scotch tape is used as a pattern which gives the shape to the deposited layer and uniform thickness of the film about 40 μm. Quadratic shapes were made with dimension 5×5mm with initial thickness 40 μm and final thickness 10-11 μm, after drying and thermal annealing. After the deposition, the films were left at room temperature for a few minutes, after which each film due to calcination was treated with the procedure: at 120°C/10 min, at 250°C/10 min, at 400°C/10 min, at 450°C/5 min and finally at 500°C/15 min, similar to the procedure presented elsewhere [17].

5.2. Natural dyes preparation and photoactive electrode formation

Anthocyanins are extracted from frozen raspberries. Raspberries are crushed in mortar and pestle until they became juicy. Curcumin was extracted from commercially purchased turmeric powder. The preparation process involved the dissolution of 5 grams of turmeric powder in ethanol. The prepared solutions were stored at room temperature and in a dark place to prevent their photodegradation.

Photoactive electrodes are made by soaking FTO glasses with TiO_2 layer in crushed raspberries or in solution of turmeric. They can stay in from several minutes to several hours, while dye molecules from the raspberries and turmeric naturally adsorb onto the titania particles. TiO_2 layer absorbs more dye molecules if it stays longer [18]. Films were pre-warmed to 80°C during staining to prevent unwanted binding of moisture from air to TiO_2. Figure 4 shows the look of a photoactive electrode after each procedure, chronologically.



a) b) c)

Fig. 4 FTO glass with TiO_2 layer deposition (a), finalized photoactive electrode stained with Raspberry (b) and finished solar cell stained with Curcuma longa (c).

5.3. Preparation of electrolyte

The electrolyte was prepared by dissolving 1.66 g of lithium iodide (approx. 60 mM LiI) and 0.254 g of iodine (approx. 0.5 mM I_2) in 20 ml of ethylene glycol at 50°C with stirring. The preparation of iodine-based electrolyte was chosen based on the reported procedures [9, 19].

5.4. Solar cell assembly

After photoactive electrode formation, the films were washed carefully with ethanol and distilled water. After drying with warm air, they were coupled with counter electrode and fastened with clips.

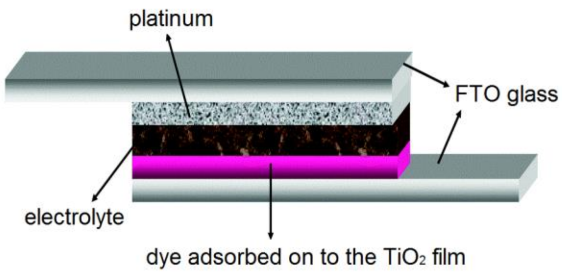


Fig. 5 Dye-sensitized solar cell assembly.

A platinum transparent electrode was prepared by a doctor-blade deposition of commercially available platinum paste (Platisol T/SP, Solaronix) on FTO glass. Furthermore, counter electrode was thermal annealed at 450°C for 30 minutes. Figure 5 shows a schematic representation of the cross-section of the solar cell.

After coupling the electrodes, the pressure of the clips is slightly reduced and the addition of the electrolyte between the electrodes by needle and syringe is applied, which completes the process of solar cell assembly (Fig. 4).

6. Measurement of Current-Voltage Characteristics

When measuring the Current-Voltage characteristics of a solar cell, it is necessary to measure the voltage of the cell and the current passing through the cell for different values of resistance in the circuit when it is exposed to solar radiation. Since dye-sensitized solar cell gives very weak current (microamperes or less), the current in the circuit was not measured directly by the ampere meter, because it would disturb the measurement. Instead, the current was determined indirectly, by measuring resistance and voltage in the circuit. This is done by using light-emitting diode and photo-resistor facing each other in a dark and closed system. Therefore, we used so-called voltage-controlled resistance, because different voltages on the light-emitting diode, give different resistances on the photo-resistor.

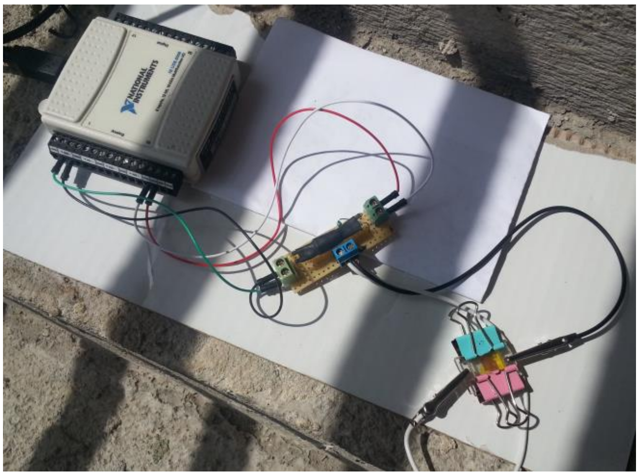


Fig. 6 Measuring equipment.

We used multifunctional system NI USB-6008 [20]. Voltage values of the LED were applied for 1136 known resistance values on the photo-resistor (range of 367-250000 Ω), and then, after 10 milliseconds the voltage of the solar cell (which was exposed to solar radiation) was measured (Fig. 6). Based on the known resistance and voltage in the circuit the current is calculated. After that the Current-Voltage characteristics are drawn.

7. Experimental Results

The analysis of TiO_2 films by scanning electron microscopy confirms the presence of a developed surface and a porous structure (Fig. 7).

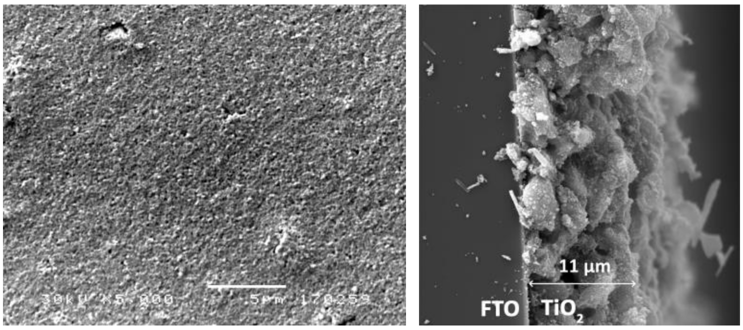


Fig. 7 SEM image of the TiO_2 on FTO glass surface
on the left and on the right its cross section.

Results for the Current-Voltage characteristics measured for dye-sensitized solar cell stained with Curcuma longa and Raspberry are shown in Figure 8. All measurements were recorded at a solar radiation intensity of 790 W/m^2.

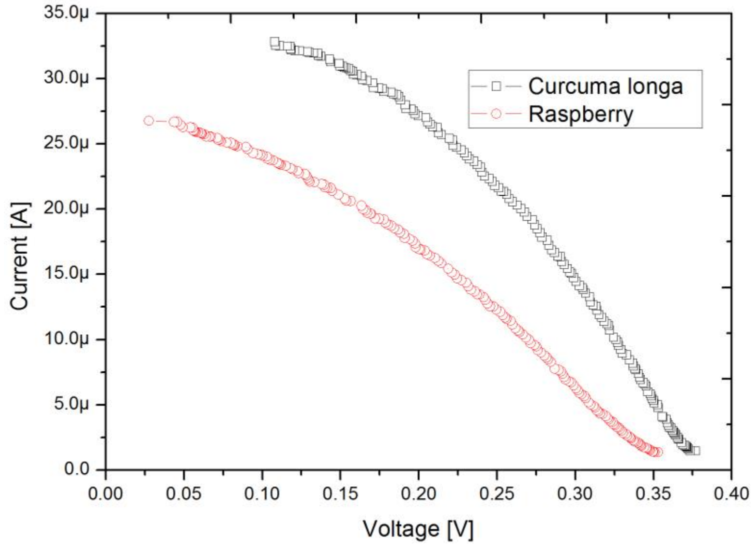


Fig. 8 Current-Voltage curve of DSSCs stained with Curcuma longa (black curve) and with Raspberry (red curve).

Dye-sensitized solar cell stained with Curcuma longa has efficiency of 0.028% and fill factor of 45%, while dye-sensitized solar cell stained with Raspberry has efficiency of 0.017% and fill factor of 36%. By comparing the Current-Voltage characteristics, we can conclude that the dye-sensitized solar cell stained with Curcuma longa is better than dye-sensitized solar cell stained with Raspberry. Graphic results can be explained by the absorption spectra of Curcuma longa and Raspberry (Fig. 9).

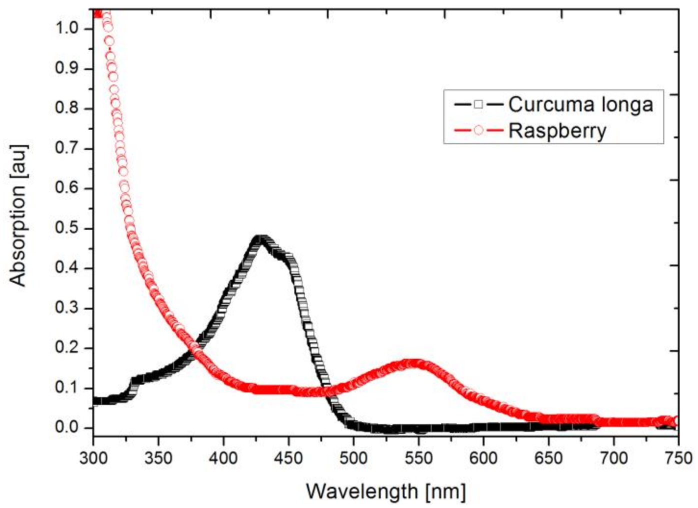


Fig. 9 Absorption spectra of Curcuma longa (black) and Raspberry (red).

The Curcuma longa is active in the visible region 400-500 nm and has a peak at 429.6 nm, while the Raspberry is active in the visible region 480-580 nm and has a peak at 544 nm, which is the characteristic of an anthocyanins [16]. The absorption spectra were recorded using the Perkin-Elmer Lambda 15 UV/Vis spectrophotometer. Samples did not have the same concentration of the solution, turmeric has a much higher absorption than shown. For our work the most important was to see the absorption peaks and compare them with simulation results.

8. SIMULATION RESULTS

Based on TDDFT, absorption spectra for cyanidin and curcumin were calculated (Fig. 10). Curcumin has an absorption peak at 420.8 nm, while the cyanidin has the highest peak at 477.3 nm, which differs from the experimental results. Considering that in experiment Raspberry dye contains more than one pigment that can absorb light, results obtained from simulation are in good agreement with the experimental values that has been previously explained (Fig. 9). Curcumin has higher absorption than cyanidin, which can explain the higher efficiency of the solar cell stained with Curcuma longa [21].

After optimization for models of the cyanidin and curcumin molecules, the HOMO-LUMO gap has value: for cyanidin 2.43 eV, which is in perfect agreement with reference work [13], and for curcumin 3.22 eV.

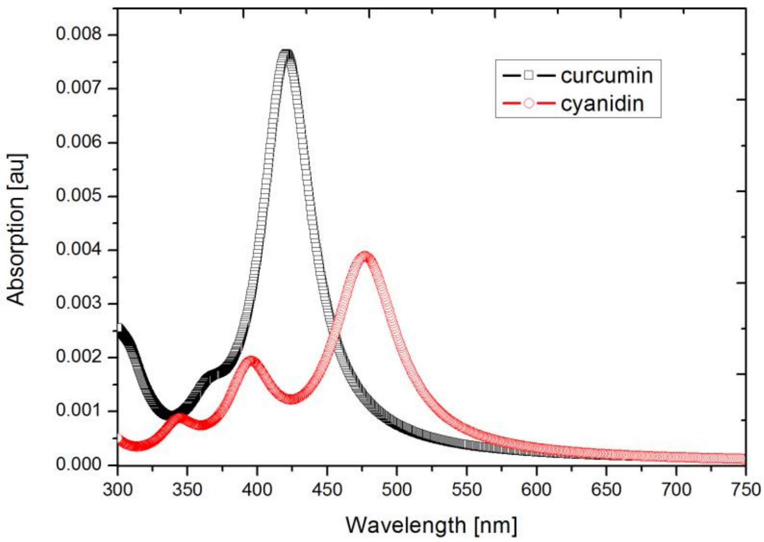


Fig. 10 Absorption spectra of curcumin (black) and cyanidin (red).

Dye molecule can be anchored on TiO_2 surface by the carbonyl (*=O*), hydroxyl (*-OH*) or carboxyl group (*-COOH*). Curcumin and cyanidin have only carbonyl and hydroxyl groups. A carboxyl group can be represented as a combination of a hydroxyl group and a carbonyl group. Adsorption modes can be bridged bidentate and monodentate modes. For simplicity, the adsorption modes are represented with a carboxyl group (Fig. 11) [22].

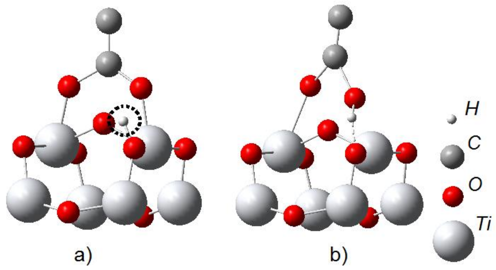


Fig. 11 Anchoring region for bridged bidentate (a) and monodentate (b) adsorption modes. The dotted circle denote the position of deprotonated atom (a).

When dye molecule binds to the titanium dioxide surface deprotonation process may occur. Deprotonation process happens when hydrogen atom of the dye molecule transfers to the titanium dioxide surface during anchoring. In the case of curcumin and cyanidin the *H* atom is transferred from the hydroxyl group to the TiO_2 structure. Deprotonation process lowers the energy of the system.

In Figure 11a, we can see that the dye molecule formed a bridged bidentate adsorption after the deprotonation was performed. Note that the hydrogen atom (dotted circle) is bound to oxygen from the cluster of titanium dioxide. However, the dye molecule can be adsorbed, as in Figure 11b, without deprotonation. In this case, hydrogen bond may occur. Of course, hydrogen atom can be also deprotonated which is lowering the energy of the system [13].

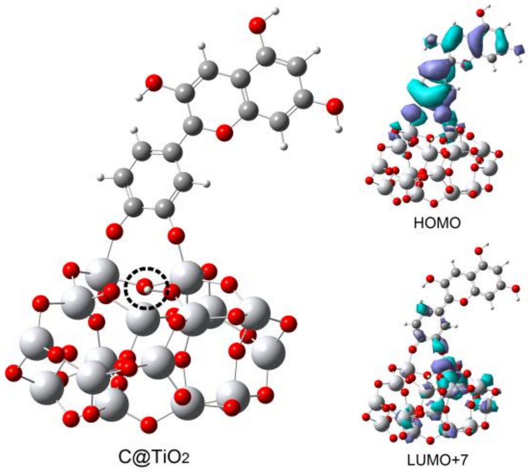


Fig. 12 Optimized geometries of the cyanidin adsorbed onto the $(TiO_2)_{16}$ model ($C@TiO_2$)[1], along with their HOMO and LUMO+7.

In our simulation, we observed three systems of molecule/cluster. Deprotonation was performed in each of them. Dotted circles denote the positions of protons that have been deprotonated from dye molecules to the $(TiO_2)_{16}$ cluster (Fig. 12, 13, 14). Figures also illustrate the HOMOs and LUMOs of molecule/cluster systems.

[1] $C@TiO_2$ label means cyanidin anchored onto the TiO_2.

In the case of C@TiO_2 and K2@TiO_2 the first level above the LUMO that is delocalized on the whole molecule/cluster is LUMO+7 (energy -2.859 eV for C@TiO_2 and -3.039 eV for K2@TiO_2). For K1@TiO_2 the first such level is LUMO+28, which is at higher energy (-2.597 eV). The absorption of electrons from the valence band to the LUMO+7 and LUMO+28 levels lead to direct electron injection [23] in the TiO_2, since the LUMO levels are delocalized along the whole system.

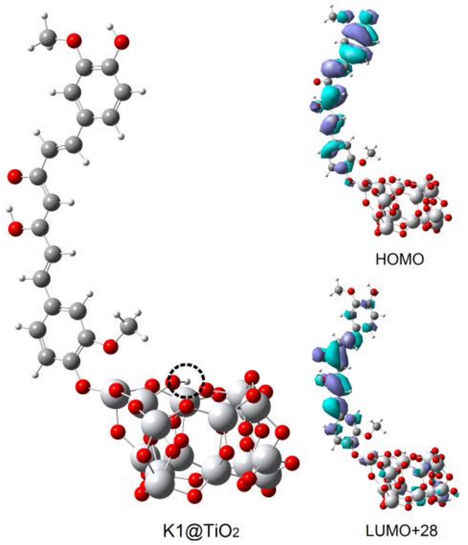


Fig. 13 Optimized geometries of the curcumin in monodentate anchoring adsorbed onto the $(TiO_2)_{16}$ model (K1@TiO_2)[2], along with their HOMO and LUMO+28.

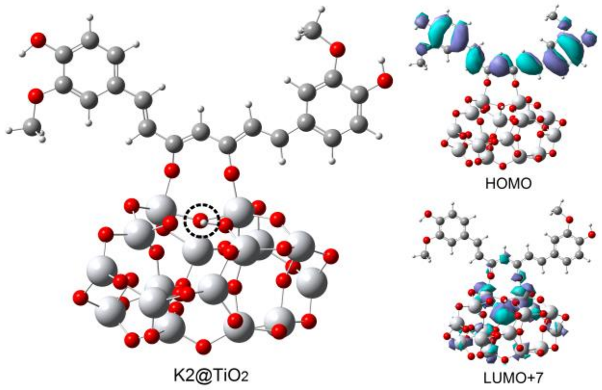


Fig. 14 Optimized geometries of the curcumin in bridged bidentate anchoring adsorbed onto the $(TiO_2)_{16}$ model (K2@TiO_2)[3], along with their HOMO and LUMO+7.

[2] K1@TiO_2 label means curcumin with one bond anchored onto the TiO_2.
[3] K2@TiO_2 label means curcumin with two bonds anchored onto the TiO_2.

After optimization was carried out for three molecule/cluster systems HOMO-LUMO gaps were calculated: for C@TiO_2, K1@TiO_2 and K2@TiO_2 in the order of 2.37 eV, 1.93 eV and 2.34 eV. We notice that HOMO-LUMO gaps have decreased after binding molecules onto the clusters. Also that curcumin has the smallest HOMO-LUMO gap when it is monodentate (K1@TiO_2) anchored onto the TiO_2. Based on these results, energy diagram of the cyanidin, curcumin, TiO_2 model and three molecule/cluster systems was made (Fig. 15).

Effective dye-sensitized solar cell requires the HOMO of the dye molecule to reside in the TiO_2 band gap and its LUMO to lie within the conduction band of the TiO_2 [13]. We noticed that the HOMO levels of all three molecule/cluster systems are in the band gap of the TiO_2, and that LUMO levels are below the CBM (conduction band minimum). The energy of the CBM is -3.835 eV. The nearest to the conduction band is LUMO level of K2@TiO_2 (-3.842 eV), then LUMO level of K1@TiO_2 (-3.925 eV) and at the end LUMO level of C@TiO_2 (-4.386 eV). In all systems, all other LUMO levels were found in the conduction band of TiO_2 cluster.

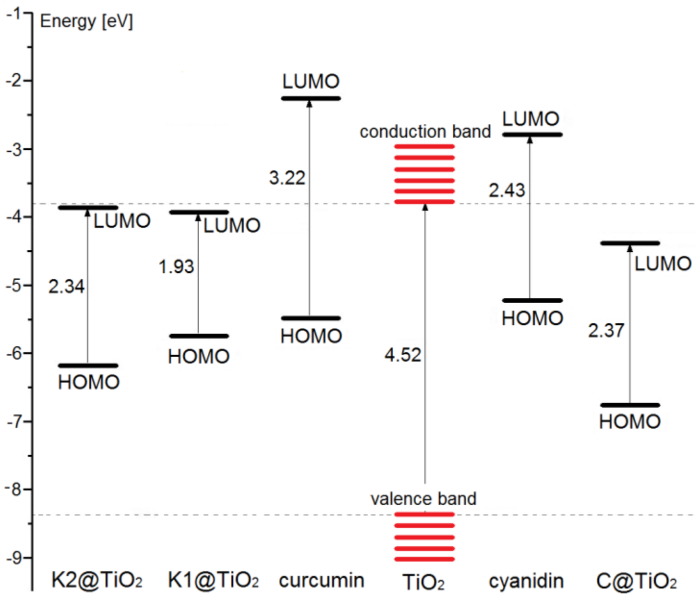


Fig. 15 Schematic energy diagram of the cyanidin, curcumin, TiO_2 model and three molecule/cluster systems.

The results confirm that electron has a higher probability to reach the conduction band in case of systems with the curcumin than in case of system with the cyanidin, which indicates another reason why the solar cell with curcumin has greater efficiency.

9. CONCLUSION

The experimental results showed that the dye-sensitized solar cell stained with Curcuma longa provides greater efficiency than the dye-sensitized solar cell stained with Raspberry. DFT calculations showed that a curcumin is closer to the conduction band minimum than a cyanidin, which indicates that electron from curcumin has a higher probability to reach the conduction band. We concluded that curcumin has better properties as a sensitizer than cyanidin for the needs of dye-sensitized solar cells, which is confirmed both by experimental and by simulation results. It is essential to find new dye sensitizers to improve efficiency of the dye-sensitized solar cells, one of the potential new dye sensitizer could be curcumin.

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