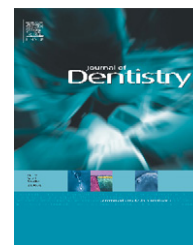


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# Core-shell quantum dots tailor the fluorescence of dental resin composites

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## ABSTRACT

**Objectives:** We characterized the optical properties, such as absorbance and fluorescence, of dental resins containing quantum dots (QD). We also determined the doping level needed to obtain a broad and nearly flat emission spectrum that provides the perception of white color.

**Methods:** The samples studied were resin composites from Charisma (Heraeus Kulzer) prepared with CdSe/ZnS core-shell QD (0.05–0.77 mass%).

**Results:** The results showed that the fluorescence of dental resin composites can be tailored by using CdSe/ZnS core-shell quantum dots.

**Conclusion:** QD core incorporation into dental resins allows the fabrication of restorative materials with fluorescence properties that closely match those of natural human teeth.

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## 1. Introduction

Due to the increased demand observed in recent years, cosmetic dental procedures have undergone continuous development.<sup>1–8</sup> The shape, shade, and arrangement of the teeth, as well as the relationship of teeth to facial features, are all important characteristics of a pleasing dental appearance.<sup>8</sup> Therefore, the search for new resin composites and techniques for teeth restoration is ongoing. The main objectives of the search are both the final quality of the restoration and the optical properties of the resin, which must be as similar as possible to the natural tooth. For example, the resin composites are applied by placing layers or increments to mimic the natural anatomy of teeth. The achievement of a restoration that is completely integrated into the tooth and that fulfills the functional and cosmetic requirements will depend on the dentist's knowledge of the dental patterns and anatomy, as well as on the restorative materials employed for the reproduction of these structures.

Properties of the resin composites such as the filler size and the type, content and shade of the material determine light absorption, scatter and reflection<sup>1,4,6,9</sup>. Optical properties, such as absorption and scattering phenomena within the dental material, are responsible for light attenuation, determining the material color and translucency. Therefore, the optical properties of the restorative material are responsible for success or failure in matching tooth color.<sup>6,10–12</sup> The visual appearances of natural teeth and the restoring materials depend on their optical properties and on the illumination applied. For example, natural teeth emit at 440 nm when exposed to ultraviolet (UV) light, which makes teeth whiter and brighter in daylight<sup>10,13–15</sup>; three emission peaks were observed at 350, 405 and 450 nm for human and bovine dental enamel.<sup>12,14</sup> Several commercial porcelains manufactured by Myerson, G-C Dental Company of Japan and Schwed present peak emission at 520, 400 and 435 nm, respectively.<sup>16</sup> Ideal restorative materials should have fluorescence similar to that of natural teeth. In the absence of fluorescence, as in the case

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of Lactona porcelain powder (Lactona Products, division of Warner Lambert Pharmaceutical Co., Morris Plains, NJ),<sup>16</sup> cosmetic qualities of the restoration are reduced, particularly under UV illumination.<sup>7</sup> The insertion of luminophores alone into the dental resin does not yield fluorescence intensity close to that of a natural tooth or they are dependent on the porcelain type.<sup>13</sup>

On the other hand, semiconductor nanoparticles or quantum dots (QD) are materials that hold potential for a variety of new applications, including their use in fluorescent labels for biomedical imaging, photonic devices, sensor materials, drug delivery and engineered tissues.<sup>17-19</sup>

This work is a proof of principle for the use of core-shell nanostructured quantum dots to tailor the fluorescence of dental resin composites. We applied the laser-induced fluorescence technique to study dental resins impregnated with different concentrations of CdSe/ZnS core-shell nanocrystals at room temperature.

## 2. Material and methods

In this work, A2 shade composite dental resin (Charisma, Heraeus Kulzer, São Paulo, Brazil), containing core-shell nanostructured CdSe/ZnS semiconductor nanocrystals, were studied by laser-induced fluorescence. Studies were performed for different concentrations of CdSe/ZnS core-shell nanocrystals embedded in the composite resins. The studied samples are shown in Table 1. The mean diameter of the nanocrystals used was 3.7 nm.<sup>20</sup> Those CdSe/ZnS QDs employed as resins dopant were encapsulated in PMMA and was supplied by the manufacturer, in powder form (Evident Technologies, Troy, NY, USA). Molecular weight was 74  $\mu\text{g}/\text{nmol}$ .<sup>21</sup> A Teflon matrix containing a hole (7 mm diameter  $\times$  3 mm deep) was used to standardize the QD-doped resin samples. After weighing an amount of resin sufficient to fill up the hole in the Teflon matrix and the desired amount of QD powder, the powder and the uncured resin were mixed and homogenized in a dim room with the aid of a spatula. The homogenized resin was then inserted into the cavity of the Teflon matrix in a single step and interposed between glass sheets to prevent bubbles and to obtain a smooth surface. The resin photo polymerization was accomplished by using a light emitting diode (LED) source (model UltraBlue, DMC, São Carlos, Brazil). The LED light, with emission centered at 465 nm (90 mW, 25 nm FWHM), was applied directly onto the

**Table 1 – Thickness (*L*) of the produced experimental samples and average values of the resin emission bands ( $\lambda_{em}$ ) versus (CdSe/ZnS)PMMA core-shell nanocrystal concentration.**

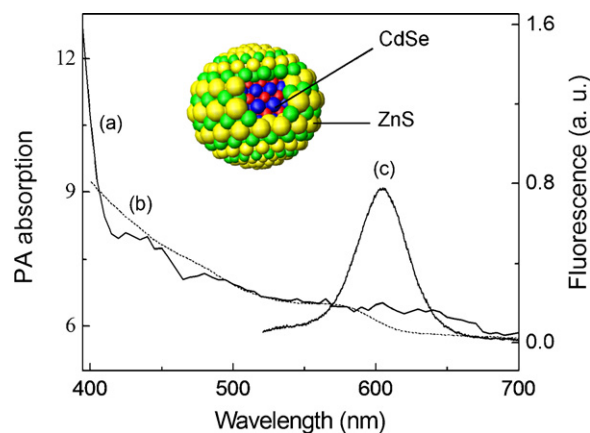
Sample	<i>L</i> (mm)	QD proportion in resin composite (in mass %)	$\langle\lambda_{em}\rangle$ (nm)
S1	2.5	0.048	601.0
S2	2.9	0.12	602.3
S3	3.2	0.26	604.0
S4	3.0	0.51	605.2
S5	3.0	0.77	608.4

glass sheet that covered the resin composite for a period of 40 s. Furthermore, a solution of core-shell colloidal nanoparticles was prepared by mixing PMMA-encapsulated CdSe/ZnS quantum dots in chloroform ( $\text{CHCl}_3$ ) solvent at 30 mg/mL, under constant magnetic stirring. In addition, three commercial samples with different core sizes of CdSe/ZnS core-shell QD suspended in toluene, were obtained in solution form from Evident Technologies. The different core sizes emitted green, yellow and red-orange light.<sup>21</sup>

The measurements of the absorption spectrum for the CdSe/ZnS quantum dots suspended in chloroform were performed with a spectrophotometer (model Carry 50, Varian, Palo Alto, CA, USA) at room temperature, with resolution of 1 nm. For the spectral absorption measurements of the dental resin samples, we employed the photoacoustic technique.<sup>22,23</sup> The fluorescence spectra were measured by exciting the samples with an argon ion laser at wavelength  $\lambda_e = 488$  nm and excitation power  $P_e \approx 15$  mW for resin samples; and at  $\lambda_e = 457$  nm and  $P_e \approx 2.1$  mW for core-shell solutions. The signal was collected by an optical fiber (superguide G fiber SFS 600/660 T, Fiberguide Industries, New Jersey, USA) with 600  $\mu\text{m}$  diameter, which delivered the collected light to a spectrometer (Model MS257, Oriel Instruments, Stratford, CT, USA) with a 300 lines/mm diffraction grating. Spectral acquisition was performed by a charge coupled device (CCD) connected to a computer.

## 3. Results and discussions

Fig. 1 shows the absorption and emission spectra ( $\lambda_e = 488$  nm) for the S5 resin sample containing QD (see Table 1) and the absorption spectrum for (CdSe/ZnS)PMMA core-shell nanocrystals suspended in chloroform. The inset in Fig. 1 shows the nanostructure of the QD, with the CdSe core surrounded by the ZnS shell. The core size of the nanocrystals in solutions was determined from the spectral absorption data.<sup>20,24</sup> The absorption band at  $\sim 575$  nm, correspondent to the transition of  $1S_{3/2}1S_e$ , is characteristic of a CdSe/ZnS core size of



**Fig. 1 – Absorbance [ $-\log(I/I_0)$ ] for (a) S5 (Table 1), and (b) (CdSe/ZnS)PMMA core-shell suspended in chloroform (5-mm glass cuvette). Emission for resin S5 is also presented (c). The inset shows the nanostructure of the QD, with the CdSe core surrounded by the ZnS shell.**

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