Supplementary Materials for

**Superdurable and fire-retardant structural coloration of carbon nanotubes**

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Supplementary Texts

Text S1. The primary mechanism for the structural coloration of TiO$_2$-coated CNTFs based on thin-film interference

To reveal the influence of surface roughness of TiO$_2$ layers on the structural colors of TiO$_2$-coated CNTFs, their surface roughness was characterized by atomic force microscopy (AFM). As shown in Fig. S16, the surface roughness of TiO$_2$ layers did not change significantly with the increase of thickness, which could also be confirmed by the SEM images in Fig. 1 and fig. S4. Although the surface roughness of TiO$_2$ layers with different thickness was similar, but the apparent color and the position of reflection peaks of TiO$_2$-coated CNTFs showed a strong dependence on the thickness of TiO$_2$ layers. Therefore, the thickness of TiO$_2$ layers is the primary factor determining the structural coloration of TiO$_2$-coated CNTFs.

Fig. S16. AFM images of TiO$_2$-coated CNTFs with 500, 1000 and 2000 ALD cycles. (A) AFM images of TiO$_2$-coated CNTFs with 500 ALD cycles. (B) AFM image of TiO$_2$-coated CNTFs with 1000 ALD cycles. (C) AFM image of TiO$_2$-coated CNTFs with 2000 ALD cycles.

As shown in Fig. 1, dividing the fiber surface into several micro regions, each region can be seen as a plane surface coated with a thin TiO$_2$ layer, suggesting that the structural colors of colorful CNTFs are generated by the thin-film interference of the TiO$_2$ layers.

To further reveal the relationship between the TiO$_2$ thickness and the structural colors, three different models were established using a finite element method (FEM) on COMSOL Multiphysics 5.5, which were planar surface interference model (Fig. S17A), stacked surface reflectivity model (Fig. S17B), and single fiber model (Fig. S17C), respectively. Using the above three models, the calculated reflection spectra of TiO$_2$-coated CNTFs were obtained (Fig. S17D-F). It is obvious that
the reflection spectra from the approximated plane surface reflectivity model (fig. S17D) matches much better with the experimental results (shown in Fig. 3D in the main text) than the reflection spectra from the other two models (fig. S17E and F), clearly confirming that the thin-film interference based on the approximated plane surface reflectivity model (fig. S17A) can well explain the mechanism for the structural coloration of TiO$_2$-coated CNTFs.

Fig. S17. Schematic diagram and calculated reflection spectra of TiO$_2$-coated CNTFs based on three different models. (A) and (D) Schematic diagram and calculated reflection spectra of TiO$_2$-coated CNTFs based on the approximated plane surface interference model. (B) and (E) Schematic diagram and calculated reflection spectra of TiO$_2$-coated CNTFs based on stacked film interference model. (C) and (F) Schematic diagram and calculated reflection spectra of TiO$_2$-coated CNTFs based on a single fiber model using FEM on COMSOL Multiphysics 5.5.

Text S2. The influence of surface roughness on the structural colors of TiO$_2$-coated CNTFs

From fig. S17D, we can see that the calculated reflection spectra did not match well with the experimental measurement. The main reason roots in the surface roughness of both TiO$_2$ layers and CNTFs. As shown in Fig. 1C-F and fig. S2-S5, it is obvious that CNTFs were formed by the intertwisting of countless single CNTs and CNT bundles, which were randomly distributed in CNTFs, exhibiting hierarchical surfaces with obvious roughness. Besides, the coated TiO$_2$ layer on CNTFs also exhibited a rough surface with many nanoscale wrinkles and particle-like humps.
The rough surfaces of both TiO$_2$ layers and CNTFs resulted in the fact that the reflection spectra based on simplified model (fig. S17A and D) could not fully match the measured results (Fig. 3D). To further prove the above analysis, we also utilized the same method to prepare colorful TiO$_2$-coated silicon wafers with highly smooth surfaces as control samples using the same ALD process (fig. S18A). As shown in fig. S18, the calculated reflection spectra of TiO$_2$-coated silicon wafers (fig. S18C) from the approximated plane surface reflectivity model using FEM on COMSOL Multiphysics 5.5 matched well with the experimental data (fig. S18B). Therefore, the surface roughness is another key factor affecting the reflection spectra of TiO$_2$-coated CNTFs because of the thin-film interference between the TiO$_2$ layers and the surface of CNTFs, as well as the combination effect of the reflection, refraction, and scattering from the wrinkles and humps on the surfaces of TiO$_2$ layers. The apparent color of TiO$_2$-coated CNTFs are actually generated by the synergistic effect of the above factors. Therefore, the dependence of peak positions on these factors did not exhibit a simple linear relationship with the thickness of TiO$_2$ layers, but a complicated one.
**Fig. S18. Colorful TiO$_2$-coated silicon wafers (SWs) and their reflection spectra.** (A) Photos of colorful TiO$_2$-coated SWs. (B) Measured reflection spectra at the normal incidence for TiO$_2$-coated SWs. (C) Calculated reflective spectra of TiO$_2$-coated SWs based on planar surface interference model using FEM on COMSOL Multiphysics 5.5.

**Text S3. The correlation between the surface roughness and the structural color of TiO$_2$-coated CNTFs**

In order to precisely describe the structural color of TiO$_2$-coated CNTFs using the reflection spectra and predict the color of TiO$_2$-coated CNTFs with any TiO$_2$ thickness, we first need to correct the reflection spectra shown in fig. S17D by defining one or more roughness parameters and establish a relationship between the experimentally measured reflection spectra and the calculated reflection spectra based on approximated plane surface reflectivity model by FEM on COMSOL Multiphysics 5.5. Two-dimensional models with a size of 30*30 μm were built in this
work. The shape of air-TiO$_2$ interface (interface 1) and TiO$_2$-CNTF interface (interface 2) were modified by generating a random curve using the following expression:

$$f(x) = H \sum_{m=0}^{N} \left[ g_1(m) \cos(2\pi m x + u_1(m)) \right]$$

, where $g_1$ and $u_1$ are 1D random functions. Here, we define two important parameters $H$ and $R$, among which $H$ represents the average height of the surface and $R$, the spectral exponent, represents the roughness of the surface. Besides, $N$ refers to the spatial frequency resolution, and we fix it as a certain value of 1000 to simplify the model. Replacing the constant function that represents the smooth interfaces in the planar thin film interference model mentioned before with the above function, and assigning different values to $H$ and $R$, the model matches better with the real situation. Fig. S19 shows some typical situations used in the modified model. It should be noted that according to the actual situation, the average height and roughness of interface 2 is larger than that of interface 1, which means $H_2 > H_1$ and $R_2 < R_1$. When $H=0$, the surface becomes smooth, which is the same as the planar surface interference model mentioned above. The results of the optimized model are shown as fig. S20, from which we can see that, by using the roughness parameters $H$ and $R$, the corrected reflection spectra can well match with the experimentally measured curves.

**Fig. S19. Different conditions for different surface roughness.** The corresponding roughness parameters $H$ and $R$ are indicated in the upper right corner, which are (A) $H_1=H_2=0$; (B) $H_1=0$, $H_2=10$, $R_2=0.9$; (C) $H_1=10$, $H_2=20$, $R_1=10$, $R_2=0.9$. 
Fig. S20. The correction of calculated reflection spectra using roughness parameters. (A) Comparison between the calculated reflection spectra and the corrected one using roughness parameters. All these curves are calculated based on approximated plane surface reflectivity model by FEM on COMSOL Multiphysics 5.5. (B) Comparison between the measured reflection spectra and the calculated spectra after correction using roughness parameters.

Text S4. Calculation of estimated UV aging time of TiO$_2$-assisted structural colors

For the estimated UV aging time, it should be mentioned that the accurate calculation is difficult because of the frequent changes in outdoor light conditions. So far, there are no unified standards or research papers that point out the formula for accelerated aging test speed in laboratories. The main reason is that there are too many changing factors under natural conditions, such as light time, light intensity, temperature and humidity, rainfall, and even pollutants, which will affect the aging process. In addition, outdoor lighting is not only related to seasons and regions but also closely related to temperature. Different places have different altitudes, and different seasons have different sunlight. Therefore, no testing time was given in all test standards. Generally, the aging time is the result of multi-party negotiation and estimate roughly.

For China, the total annual solar radiation throughout the country is 3340-8400 MJ/m$^2$-year.
For Beijing, the annual solar radiation hours are 2200~3000 h with a median value of 2600 h. The total amount of solar radiation is 5016~5852 MJ/m$^2$•year (1393~1625 kWh/m$^2$•year) with a median value of 5434 MJ/m$^2$(1509 kWh/m$^2$•year). Meanwhile, UV light takes up about 5% of total solar radiation.

(Data was obtained from https://news.solarbe.com/202006/29/326417.html).

In this work, we used a total annual solar radiation of 5434 MJ/m$^2$ (1509 kWh/m$^2$•year) and an annual solar radiation hour of 2600 as the reference data. Therefore, the total annual UV light radiation was calculated to be 271.7 MJ/m$^2$ (which equals to 75.45 kWh/m$^2$•year)

Thus, the annual average hourly solar radiation per unit area was calculated to be

75450 Wh/m$^2$•year÷2600h/year≈29.0 W/m$^2$;

The total radiation intensity under test conditions was 407.5 W/m$^2$. Thus we obtained

407.5 W/m$^2$÷29 W/m$^2$=14.05

Therefore, we estimated that the simulated irradiation was about 14.05 times higher than that of outdoor UV irradiation. However, UV aging is also closely related to temperature. The annual average temperature in Beijing is 11.6 °C. When the temperature increases by 1 time, the empirical aging effect evaluation will increase by about 1.5 times.

The actual test temperature is greater than 50 °C. In this part, we set it to be 50 °C.

Based on the above analysis, we obtained a relationship between the estimated UV aging time and the accelerated UV-aging time

14.05*50/11.6*1.5=90.8

In this work, the actual time of UV irradiation test was 10 month (about 300 days), hence, the estimated UV aging time in real environments was calculated to 10 month*90.8 =908 month (which equals to 75.6 years).
Because the annual outdoor sunlight irradiation time is about 8 hours per day (considering the removal of cloudy or rainy days), therefore, the total estimated UV aging time is about 908 month*24/8=2724 months (which equals to 227 years).