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Tailoring TiO₂ Nanotube-Interlaced Graphite Carbon Nitride Nanosheets for Improving Visible-Light-Driven Photocatalytic Performance

Yang Wang, Xueqin Liu,* Cunchuan Zheng, Yinchang Li, Songru Jia, Zhen Li,* and Yanli Zhao*

Rapid recombination of photoinduced electron–hole pairs is one of the major defects in graphitic carbon nitride (g-C₃N₄)-based photocatalysts. To address this issue, perforated ultralong TiO₂ nanotube-interlaced g-C₃N₄ nanosheets (PGCN/TNTs) are prepared via a template-based process by treating g-C₃N₄ and TiO₂ nanotubes polymerized hybrids in alkali solution. Shortened migration distance of charge transfer is achieved from perforated PGCN/TNTs on account of cutting redundant g-C₃N₄ nanosheets, leading to subdued electron–hole recombination. When PGCN/TNTs are employed as photocatalysts for H₂ generation, their in-plane holes and high hydrophilicity accelerate cross-plane diffusion to dramatically promote the photocatalytic reaction in kinetics and supply plentiful catalytic active centers. By having these unique features, PGCN/TNTs exhibit superb visible-light H₂-generation activity of 1364 μmol h⁻¹ g⁻¹ (λ > 400 nm) and a notable quantum yield of 6.32% at 420 nm, which are much higher than that of bulk g-C₃N₄ photocatalysts. This study demonstrates an ingenious design to weaken the electron recombination in g-C₃N₄ for significantly enhancing its photocatalytic capability.

Semiconductor photocatalysis as a type of green technology for solar energy conversion has exhibited fascinating application prospects in environmental and energy-relevant areas, such as photocatalytic degradation of pollutants and photocatalytic hydrogen generation.[1] In the wave of photocatalysis research, graphitic carbon nitride (g-C₃N₄)-based photocatalysts have been significantly flourishing[2] since its promising utilization for water splitting under visible-light irradiation reported in 2009.[3] As a new generation of metal-free photocatalysts, g-C₃N₄ exhibits various extraordinary features, such as desirable visible-light response (λ < 460 nm) with medium bandgap (2.7 eV), environmental friendliness, reasonable cost, and high stability.[4] However, its photocatalytic performance is severely hampered by high recombination rate of photoinduced electron–hole pairs, low specific surface area, and limited active sites.[5] Thus, extensive research has been carried out to enhance its photocatalytic activity.[6] To date, the most common approaches to improve the photocatalytic activity of g-C₃N₄ include doping for narrowing the bandgap,[7] morphology engineering for light harvesting,[8] and composite structure control for effective charge separation.[9,10]

Inspired by the graphene research,[10] the exfoliation of layered bulk materials into 2D counterparts induces unique physicochemical properties including increased surface area, ultrahigh charge carrier mobility, and pronounced changes in the energy band structure. Indeed, reducing the dimensionality of bulk g-C₃N₄ into 2D nanosheets could increase the surface area that is beneficial to the improvement of photocatalytic efficiency via efficient exposure of active sites, optimized light harvest, and charge separation.[11] Moreover, lacunaris materials are highly appreciated as catalysts due to their accessible porous nature with a large surface area for the mass transfer.[12] Hence, porous 2D g-C₃N₄ nanosheets have been prepared to show photocatalytic performance enhancement.[11]

In addition to the nanostructure design, the development of g-C₃N₄ hybrids for photocatalysis is another promising approach.[13] Among various routes, the introduction of an appropriate band structure at the heterojunction interface is the most important prerequisite to enhance the charge separation efficiency for increasing photocatalytic performance.[13a,12a,15] Since TiO₂ has an advantage in electron–hole separation for its special energy band,[16] the establishment of heterostructure...
between g-C$_3$N$_4$ and TiO$_2$ is an effective way to improve the efficiency of charge separation in g-C$_3$N$_4$/TiO$_2$ photocatalyst systems.$^{[17]}$ 1D TiO$_2$ nanotubes having the advantage of intimate contact with 2D nanosheets and cross-linked network$^{[18]}$ present a significant potential as photoinduced electron sink in g-C$_3$N$_4$/TiO$_2$ heterojunction hybrids for the electron–hole separation.

Herein, by utilizing the mixture of ultralong TiO$_2$ nanotubes (Figure S1, Supporting Information) and melamine as precursors for the polymerization, we developed a template-based strategy to generate the perforated ultralong TiO$_2$ nanotube-interlaced g-C$_3$N$_4$ nanosheets (PGCN/TNTs) having in situ formed in-plane holes. As compared to intact ultralong TiO$_2$ nanotube-interlaced g-C$_3$N$_4$ nanosheets (GCN/TNTs), the as-prepared PGCN/TNTs with in-plane holes can improve the electron–hole separation and solar energy utilization. Meanwhile, the surface hydrophilicity of PGCN/TNTs was well optimized via the alkaline solution treatment, resulting in greater H$_2$O affinity. Accordingly, PGCN/TNTs were expected to display high visible-light photocatalytic activity in H$_2$-generation and dye degradation.

At the initial stage of the preparation reaction, melamine was fully covered on the surface of TiO$_2$ nanotubes due to the formation of hydrogen bonds after ultrasonic treatment. Then, the hybrid thermally polymerized into bulk g-C$_3$N$_4$/TNTs. In the bulk g-C$_3$N$_4$/TNTs, TNTs as the skeleton were implanted into the 2D framework of g-C$_3$N$_4$. Under the tailoring treatment of alkali ions$^{[19]}$ with embedded TNTs as the template, PGCN/TNTs were obtained (Figure 1a). To verify this synthetic process, the hydrolysis was first studied by scanning electron microscope (SEM) and transmission electron microscope (TEM). Figure 1b shows that pristine bulk g-C$_3$N$_4$/TNTs were composed of stacked lamellar textures with sizes around micrometers, and TNTs were not found in the bulk material on account of possible embedment by 2D g-C$_3$N$_4$ nanosheets during the copolymerization process. Figure 1c presents the TEM image of the GCN/TNTs. It was found that the nanotubes interlaced with the 2D nanosheets without holes. After tailoring process, holes were distributed uniformly on the nanotubes interlaced with the 2D nanosheets without holes. The exfoliated nanosheets were enriched with holes of ≈ 100 nm in diameter. The ultralong nanotubes homogeneously interlaced with the retained g-C$_3$N$_4$ nanosheets, and no naked nanotubes were observed in holes or edges of nanosheets. Figure 1g shows high-resolution TEM (HRTEM) image of PGCN/TNTs. Interestingly, an apparent heterointerface was observed between TNTs and g-C$_3$N$_4$ nanosheets. Two sets of lattice fringes with vertical interplane spacing of 0.24 and 0.19 nm in TNTs are attributed to (001) and (100) planes of the anatase TiO$_2$ phase, respectively, and the heterointerface is roughly along the [100] direction of the anatase crystal. In addition, the tailoring function of alkali ion on g-C$_3$N$_4$ was also proven by microscopic images (Figure S2, Supporting Information). It was found that without TNTs as the template, the g-C$_3$N$_4$ nanosheets were cut into nanofibers. Clearly, the 2D g-C$_3$N$_4$ framework was restricted when they contacted with TiO$_2$ nanotubes, indicating the template effect of the interlaced TiO$_2$ nanotubes. Elemental mapping images from the selected areas of PGCN/TNTs show a homogenous distribution of TiO$_2$ nanotubes (Figure 1h).

The crystal structures of the as-prepared PGCN/TNTs were analyzed by powder X-ray diffraction (XRD), as shown in Figure 2a. It is well known that the two dominant diffraction peaks at 13.0° and 27.3° in the powder XRD pattern of g-C$_3$N$_4$ are associated with the in-plane trigonal N linkage of tri-s-triazine motif and the periodic stacking of layers for conjugated aromatic systems, respectively. The peak located at 25.3° is ascribed to (101) facet of the anatase TiO$_2$ (JCPDS no. 89–4921). As compared with bulk g-C$_3$N$_4$ and g-C$_3$N$_4$/TNTs, the decreased intensity of peak at 27.3° in GCN/TNTs and PGCN/TNTs verified that the layered g-C$_3$N$_4$ structure was successfully exfoliated into nanosheets. Almost disappeared peak of PGCN/TNTs at 13.0° (Figure S3, Supporting Information) is mainly due to the fact that the alkali ion interaction with the in-plane trigonal nitrogen during prolonged tailoring process for the formation of in-plane holes may decrease the ordering degree of in-plane structures.

The chemical structures of g-C$_3$N$_4$, g-C$_3$N$_4$/TNTs, GCN/TNTs, and PGCN/TNTs were studied by Fourier transform infrared (FTIR) spectra as shown in Figure 2b. All of the bands attributable to a typical g-C$_3$N$_4$ were observed, further demonstrating that the chemical structure of tri-s-triazine ring unit is robust against ultrasonic and alkali treatment. Specifically, broad peaks between 3000 and 3500 cm$^{-1}$ are assigned to amine group and water. Compared with bulk g-C$_3$N$_4$ and g-C$_3$N$_4$/TNTs, peaks of PGCN/TNTs between 3000 and 3500 cm$^{-1}$ became stronger, suggesting higher exposure of surface amino (N$\equiv$H) and hydroxyl (O$\equiv$H) groups in PGCN/TNTs. The presence of surface amino and hydroxyl groups indicates high exposure edges and surfaces in PGCN/TNTs, which is consistent with the direct observation by TEM. When compared to GCN/TNTs, it was noted that a new sharp peak at 1153 cm$^{-1}$ appeared in the FTIR spectrum of PGCN/TNTs. This peak is ascribed to the vibration of C=O$\equiv$C group$^{[20]}$ indicating that O atoms are preferentially bonded to C atoms by substituting coordinated N atoms. The O doping could optimize the band structure of g-C$_3$N$_4$, leading to the light absorption increase of PGCN/TNTs in the visible region (Figure S4, Supporting Information). When compared with g-C$_3$N$_4$ and g-C$_3$N$_4$/TNTs, the FTIR spectrum of PGCN/TNTs displays an additional peak at around 2150 cm$^{-1}$, which can be assigned to the vibration of cyano terminal group (C≡N)$^{[21]}$. The presence of C≡N proves that the tri-s-triazine ring was partly fractured and restructured for atomic substitution during ultrasonic tailoring treatment.

Brunauer–Emmett–Teller (BET) surface areas of the as-prepared samples were determined by N$_2$ adsorption/desorption measurements at 77.4 K. As shown in Figure 2c and Figure 5 (Supporting Information) (pore size distributions), PGCN/TNTs exhibit a large BET surface area of 90.505 m$^2$ g$^{-1}$, which is almost four times higher than that of g-C$_3$N$_4$/TNTs (18.123 m$^2$ g$^{-1}$), indicating efficient exfoliation of PGCN/TNTs. Significantly, high-specific surface area can present more active sites for the adsorption and surface reaction. The quantitative compositional analysis of TiO$_2$ content in different composites was conducted by thermal gravimetric analysis (TGA) as shown in Figure 2d. The main weight loss appears at the range of 500–700 °C due to the combustion of g-C$_3$N$_4$. The total weight
loss is 100% for pure g-C$_3$N$_4$, implying that it decomposes completely when the temperature reaches 700 °C. The retained weight is the weight percent of TiO$_2$ in the composites. The amount of TiO$_2$ in the composites was measured to be 3.19 and 8.41 wt% for GCN/TNTs and PGCN/TNTs, respectively. As compared to GCN/TNTs, higher TiO$_2$ content in PGCN/TNTs is due to the tailoring treatment of g-C$_3$N$_4$ by the alkaline solution for the hole formation.

The compositions of surface elements and their oxidation states in PGCN/TNTs, together with the interaction between TiO$_2$ nanotubes and g-C$_3$N$_4$ were analyzed by X-ray photoelectron spectroscopy (XPS). The XPS survey spectra (Figure S6, Supporting Information) confirm that all g-C$_3$N$_4$-based samples are mainly composed of carbon, nitrogen, and oxygen, and g-C$_3$N$_4$/TNTs and PGCN/TNTs contain some titanium. The strong peak (Figure 3a) pertaining to N 1s (398.3 eV) from g-C$_3$N$_4$ is assigned to sp$^2$ hybridized N (C=–N=C).[8a] The peaks in N 1s region at around 399.3 and 401.2 eV are assigned to ternary nitrogen (N–C3) and C–N–H groups, respectively.[5a] As compared with g-C$_3$N$_4$, PGCN/TNTs show unaltered functional
groups, indicating stable graphite carbon–nitrogen structure in PGCN/TNTs. Interestingly, N 1s core level spectrum of PGCN/TNTs shifts to higher binding energies by 0.4 eV, implying the interface formation between TiO\textsubscript{2} nanotubes and g-C\textsubscript{3}N\textsubscript{4} in PGCN/TNTs. Figure 3b shows the binding energy region of C 1s with the C\textsubscript{\textsuperscript{sp}}\textsubscript{2} peak located at 284.6 eV. Another peak observed at 288.1 eV in this region is related to sp\textsuperscript{2}-hybridized carbon (N=C=N) from g-C\textsubscript{3}N\textsubscript{4}. A new peak at 286.1 eV observed in PGCN/TNTs is assigned to C\textsubscript{\textsuperscript{sp}}\textsubscript{3}O species. These results suggest that there are some C=O groups on the surfaces of PGCN/TNTs after the ultrasonic/alkali ion treatment. The high-resolution O 1s peak (Figure 3c) of g-C\textsubscript{3}N\textsubscript{4}/TNTs at around 529.42 eV is associated with the Ti–O bond in TiO\textsubscript{2} nanotubes. After the hydrolyzing process of g-C\textsubscript{3}N\textsubscript{4}/TNTs, the peak at 531.58 eV representing hydroxyl (O=H) group increases dramatically. The deconvoluted spectra of C and O essentially indicate that layered bulk g-C\textsubscript{3}N\textsubscript{4}/TNTs were successfully disintegrated with functional groups such as O=H at surfaces under alkaline condition, which would be beneficial for the dispersion of PGCN/TNTs in aqueous solution (Figure S7, Supporting Information). The peaks in the Ti 2p spectrum of PGCN/TNTs display a shift of 0.8 eV to lower binding energy as compared with that of TNTs (Figure 3d), further confirming a synergistic effect between g-C\textsubscript{3}N\textsubscript{4} and TiO\textsubscript{2} in PGCN/TNTs.

The photocatalytic performance of PGCN/TNTs was evaluated by hydrogen production and dye degradation under visible-light irradiation. The H\textsubscript{2} production activities were compared among bulk g-C\textsubscript{3}N\textsubscript{4}, g-C\textsubscript{3}N\textsubscript{4}/TNTs, GCN/TNTs, and PGCN/TNTs in Figure 4a,b. g-C\textsubscript{3}N\textsubscript{4} exhibits a rather low H\textsubscript{2}-generation rate of 6.85 µmol h\textsuperscript{-1}, arising from its poor charge transport capability and low-specific surface area, while the g-C\textsubscript{3}N\textsubscript{4}/TiO\textsubscript{2} hybrids show an apparent enhancement in the H\textsubscript{2}-generation rate, primarily due to effectively constructed heterojunction for the electron–hole separation. Specially, PGCN/TNTs show the highest H\textsubscript{2} evolution rate of 68.20 µmol h\textsuperscript{-1}, which is 9.0, 3.9, and 0.6 times higher than that of g-C\textsubscript{3}N\textsubscript{4}, g-C\textsubscript{3}N\textsubscript{4}/TNTs, and GCN/TNTs, respectively. The excellent H\textsubscript{2}-generation performance of PGCN/TNTs makes it one of leading materials with high H\textsubscript{2} production rate in the reported melamine-based g-C\textsubscript{3}N\textsubscript{4} photocatalysts (Figure S8, Supporting Information). The photocatalytic H\textsubscript{2} evolution stability of PGCN/TNTs was confirmed by cycling experiments. As shown in Figure 4a, the hydrogen evolution activity of PGCN/TNTs was maintained without noticeable deactivation of photocatalytic performance in four cycles for 20 h, indicating good stability of the photocatalyst. The apparent quantum efficiency (AQE) versus the wavelength of the incident light was exhibited in Figure 4c, suggesting that AQE at 420 nm could reach up to 6.32%. Interestingly, the AQE of PGCN/TNTs remains 1.04% at 500 nm, which is higher than most of other g-C\textsubscript{3}N\textsubscript{4}-based materials (Table S1, Supporting Information). In sharp contrast, the quantum efficiency of pristine g-C\textsubscript{3}N\textsubscript{4} is much lower in both...
the UV and visible-light regions (0.86% at 420 nm). Therefore, these results demonstrate PGCN/TNTs as promising and efficient photocatalysts possess excellent quantum efficiency. In addition, the photocatalytic degradation of rhodamine B (RhB) was also measured under visible-light irradiation (λ > 400 nm). As presented in Figure S9 (Supporting Information), the degradation rate of PGCN/TNTs (0.051 min⁻¹) for RhB is about 3.6 and 12.8 times higher than that of GCN/TNTs and g-C₃N₄/TNTs respectively, which is consistent with the photocatalytic activity of H₂ evolution.

For gaining the insights into the photocatalysis mechanism, photoelectrochemical properties of the prepared samples were also investigated. The photoelectrochemical properties of g-C₃N₄/TiO₂ hybrids before and after the tailoring process were investigated by measuring their transient photocurrent response and electrochemical impedance responses. As shown in Figure 5a, all of the samples exhibit rapid photocurrent responses upon the light is turned on and off, and the responses remain at a relatively stable value during the irradiation process. Such phenomenon was well repeated in each on–off cycle, indicating good reproducibility of the samples. Further observation shows that the photocurrent of the g-C₃N₄/TNTs electrode (0.01 mA cm⁻²) is doubled as compared to that of g-C₃N₄ (0.005 mA cm⁻²), demonstrating that the separation efficiency of the photoinduced electron–hole pairs in g-C₃N₄/TNTs is improved due to unique interfacial charge transfer of heterojunction. As compared to bulk g-C₃N₄/TNTs, GCN/TNTs exhibit higher photocurrent intensity, as the exfoliated nanosheet structure exposes more active sites to restrain the recombination.²¹ PGCN/TNTs show the highest photocurrent density (0.049 mA cm⁻²), demonstrating that the charge carrier separation in tailored nanostructure of PGCN/TNTs is further improved. A similar tendency was obtained in the electrochemical impedance spectrum (EIS), as shown in Figure 5b. By fitting the obtained impedance spectra with the transmission line model (Figure S10, Supporting Information), the parameters including series resistance (R₁) and electron transport resistance (R₂) were obtained (Table S2, Supporting Information). The arc radius of PGCN/TNTs on EIS Nyquist plot is smaller than that of GCN/TNTs and g-C₃N₄, indicating that PGCN/TNTs have the lowest R₂ value (1.04 × 10⁵ Ω), enabling fast interfacial charge carrier transfer. These results further reveal that faster interfacial charge migration and more effective separation of charge carriers on the PGCN/TNTs surface are responsible for enhanced photocatalytic activity.

The effect of nanostructure tailoring on the carrier recombination in PGCN/TNTs was then studied by photoluminescence (PL) spectroscopy. g-C₃N₄ exhibits the strongest PL with a peak emission at about 460 nm, which is in good agreement with the analysis of UV–vis absorbance spectra (Figure S4, Supporting Information). As compared with GCN/TNTs, PGCN/TNTs show a reduced recombination loss of photogenerated electron–hole pairs by the efficient charge transfer. Some studies showed that redundant g-C₃N₄ nanosheets of GCN/TNTs would
be the electron–hole recombination center. Herein, the shortened electron transfer distance (from g-C$_3$N$_4$ to g-C$_3$N$_4$/TNTs interface) after the tailoring treatment was obtained for effective electron–hole separation in the case of PGCN/TNTs.

Hereafter, the photophysical characteristics of the photogenerated charge carriers for these samples were carried out using time-resolved transient PL spectra (Figure 5d). A triexponential functions fitting was employed to analyze the PL decay curves, and the average PL lifetime ($\tau_{avg}$) was deduced by the following equation

$$\tau_{avg} = \frac{A_1\tau_1 + A_2\tau_2 + A_3\tau_3}{A_1 + A_2 + A_3}$$

where $\tau_i$ is the estimated lifetime value and $A_i$ is the corresponding amplitude. The resulted average lifetime is 3.41 ns for PGCN/TNTs, which is 1.42 times longer than that of GCN/TNTs. The long average lifetime of PGCN/TNTs means that the recombination of the electron–hole pairs is significantly restrained and greatly depended on the trimming of excessive g-C$_3$N$_4$ nanosheets in PGCN/TNTs. Moreover, PGCN/TNTs show electron diffusion length ($L_n$) of 3.9 times higher than that of GCN/TNTs (Figure S11, Supporting Information). The longer $L_n$ of PGCN/TNTs indicates reduced electron traps during the electron transferring. By combining these results, it was rationally proposed that the electron transfer pathway of PGCN/TNTs was greatly optimized by the template-based alkali ion tailoring strategy, leading to efficient photoexcited electron–hole separation and long photocarrier lifetime.

On the basis of the above analysis, the photocatalytic mechanism of PGCN/TNTs was suggested as shown in Figure 6. First, in the synthesized PGCN/TNTs, the heterojunction was formed in matched energy band between g-C$_3$N$_4$ nanosheets and TiO$_2$ nanotubes to separate photoinduced electron–hole of g-C$_3$N$_4$ under visible light. According to the conduction band (CB) and valence band (VB) edge potentials of g-C$_3$N$_4$ ($-1.12$ and $1.57$ eV) and TiO$_2$ ($-0.29$ and $2.91$ eV), g-C$_3$N$_4$ in g-C$_3$N$_4$/TiO$_2$ could easily absorb the visible light. Once they are irradiated under visible light (>400 nm), electrons are able to jump from the VB to the CB of g-C$_3$N$_4$ and then transfer from the CB of g-C$_3$N$_4$ nanosheets to that of anchored TiO$_2$ nanotubes. Such electron transition between heterojunctions efficiently reduces the recombination of charge carriers and prolongs the charge lifetime. Second, a plenty of electron–hole recombination in g-C$_3$N$_4$ nanosheets was eliminated through tailoring redundant g-C$_3$N$_4$ nanosheets in PGCN/TNTs. Due to intrinsic features of easy photoinduced electron–hole recombination, g-C$_3$N$_4$ nanosheets show a short electron diffusion length. Electrons are induced and consumed by g-C$_3$N$_4$ nanosheets at a distance away from the heterojunction interface.
of GCN/TNTs. After template-tailoring strategy, the retained PGCN/TNTs could minimize the electron transport path and more solar energy could be utilized, leading to weaker electron–hole recombination and higher photogenerated electron yield (Figure 4a). Furthermore, the in-plane holes in PGCN/TNTs are also beneficial for rapid cross-plane diffusion of mass, photogenerated carriers, and hydrogen, which dramatically accelerate the photocatalytic reaction in kinetics. Third, features including large accessible surface area and high hydrophilicity not only afford plentiful catalytically active centers, but also improve the reactant adsorption and diffusion process during the reaction. At last, H₂O or dye molecules could experience the redox reactions around catalytic centers, generating H₂ or being degraded.

In summary, we have designed and developed perforated ultralong TiO₂ nanotube-interlaced g-C₃N₄ heterostructures

Figure 5. a) Transient photocurrent response and b) electrochemical impedance spectra of g-C₃N₄, g-C₃N₄/TNTs, GCN/TNTs, and PGCN/TNTs. c) PL spectra and d) time-resolved transient PL spectra of GCN/TNTs and PGCN/TNTs.

Figure 6. Schematic diagram showing the photocatalytic reaction over PGCN/TNTs as the photocatalyst under visible-light irradiation.
higher than that of bulk g-C₃N₄ photocatalysts. This unique solar energy conversion.

developing highly efficient visible-light photocatalysts toward a template-tailoring strategy presents a significant improvement in g-C₃N₄ nanosheets could be suppressed. The in-plane holes and high hydrophilicity from PGCN/TNTs not only accelerate cross-plane diffusion to dramatically promote photocatalytic activity of 1364 µmol h⁻¹ g⁻¹ (λ > 400 nm), which is ninefolds higher than that of bulk g-C₃N₄ photocatalysts. This unique template-tailoring strategy presents a significant improvement in reducing photoinduced electron–hole recombination for developing highly efficient visible-light photocatalysts toward solar energy conversion.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

Keywords
graphite carbon nitride, photocatalysis, template-tailoring methods, TiO₂ nanotubes, visible light

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