第 45 卷　第 1 期　上 海 理 工 大 学 学 报　Vol. 45　No. 1　2023
J. University of Shanghai for Science and Technology

文章编号:1007－6735(2023)01－0001－07　DOI: 10.13255/j.cnki.jusst.20230201001

光催化剂在牙科研究中的应用进展

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摘要：光催化剂有两个主要作用：有机化合物的降解和超亲水性，这些作用使其具有抗菌和自清洁的效果。目前，光催化剂已经被应用于医学临床实践。两种主要的口腔疾病(龋齿和牙周病)影响着全球几十亿人，而治疗口腔疾病则可以直接接触到口腔内的病灶。牙科领域会使用到各种先进的牙科材料。因此，光催化剂可以成为牙科临床实践的有用工具。本文综述了光催化剂在牙科研究中的应用研究，并展望了光催化剂在牙科研究中的未来前景。

关键词：光催化剂；牙科临床实践；牙科研究；牙科材料
中图分类号：O 644　　**文献标志码：**A

Recent advances of application of photocatalyst for dental research

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Abstract: Photocatalyst has two major effects: degradation of organic compounds and superhydrophilicity. These effects lead to the antimicrobial and self-cleaning effects. Photocatalyst has been applied for medical clinical practices. Two major oral diseases (dental caries and periodontal disease) globally influence on several billons of people. Treatment of oral diseases allows direct access to lesions in the oral cavity. And there are various advanced dental materials in dentistry. Therefore, photocatalyst can be a useful tool for dental clinical practice. The aim of this article is to summarize the research of application of photocatalyst for dental research and to find out the future prospective for photocatalyst to dentistry.

Keywords: *photocatalyst*; *dental clinical practice*; *dental research*; *dental materials*

Dr. Fujishima, president of Institute of Photochemistry and Photofuntional Materials of University of Shanghai for Science and Technology, found out the photocatalytic reaction mechanism [1]. This phenomenon has two major effects: degradation of organic compounds and superhydrophilicity.

Photocatalyst is known to decompose various organic compounds under illumination by generating various radicals. Organic compounds adsorbed on the surface can be self-cleaned by the photodecomposition.

收稿日期: 2023−02−01
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In addition, super hydrophilic effect makes the organic compounds hard to attach the surface. Photocatalysts have multiple applications in air purifiers, paints, and self-cleaning coatings for medical devices such as catheters, as well as in the elimination of microbes. Photocatalyst is also applied to dental research and clinical dental practice.

Dental caries and periodontal diseases are two major oral diseases. Globally, it is estimated that 2 billion people suffer from dental caries of permanent teeth and 514 million children suffer from caries of primary teeth. Prevalence of the main oral diseases continues to increase globally with growing urbanization and changes in living conditions. Periodontal disease affects the tissues that both surround and support the teeth. The disease is characterized by bleeding or swollen gums, pain and sometimes bad breath. Severe periodontal diseases are estimated to affect around 19% of the global adult population, representing more than 1 billion cases worldwide[2].

These two major oral diseases are affected by oral pathogenic bacteria. In human body, there are two major bacterial flora: intestinal bacterial flora and oral bacterial flora. An oral microbiome consists of more than 700 species of bacteria [3-9]. To control oral bacterial flora in the prevention of oral diseases, treatment of oral diseases allows direct access to lesions in the oral cavity, and there are various advanced dental materials in dentistry. Therefore, photocatalyst can be a useful tool to control oral bacterial flora. Finally, photocatalyst can be a useful tool for the prevention of oral diseases.

The aim of this article is to summarize the research of application of photocatalyst for dental research and to find out the future prospective for photocatalyst to dentistry.

1 Martials and Methods

1.1 Search strategy

Electronic database, PubMed was searched for eligible studies. Abstracts were reviewed, and full-text articles were inspected for inclusion criteria. The reference lists from reviewed studies were also examined to seek additional sources.

Key word search terms using medical subject headings and text words included. Search details were as follows: (“photocatalyst” [MeSH terms] or “photocatalyst” [all fields]) and ((“dental” [MeSH Terms] or “dental” [all fields]) or (“oral” [MeSH terms] or “oral” [all fields]))(“photocatalyst” [all fields]) and ((“dental” [all fields]) or “oral” [all fields])). Studies were included if they were written in English.

1.2 Study selection

An initial search for studies using saliva for the evaluation of periodontitis generated 46 articles. During the preliminary analysis, 15 articles were excluded because they were not concerned with dental research. After reviewed the articles, 4 review articles were removed. Finally, 27 articles were independently reviewed and summarized by two authors (Nomura Y and Hanada N).

1.3 Classification

According to the review of the abstracts, articles were classified based on the application of photocatalyst for dental materials, clinical dental practice or dental research into 3 major categories: antibacterial effect on novel dental materials, dental materials (implant, resin, others) and dental practice (tooth whitening/bleaching). After classification, the articles were summarized.

2 Results

A total of 46 articles were hit on the PubMed database, 27 articles were included for the review. According to the classifications described in materials and methods, 6 articles were classified into antibacterial effect of novel dental materials, 10 articles were implant, 4 articles were resin, 3 articles were other dental materials, and 5 articles were tooth whitening/bleaching.

2.1 Antibacterial effect of novel dental materials

Six articles were met the requirement for antibacterial effect of novel dental materials. Without one article[10], photocatalysis activity leaded in antimicrobial activity (Tab. 1). One article investigated the changes of bacterial inner cell molecules which leaded the mechanism of the antimicrobial activity of photocatalyst[11]. One article had shown that combination of hydroxyapatite, which was the major component of tooth enamel, and TiO_2 acted for remineralization for demineralized tooth enamel surface[12](Tab. 1).

表 1　钛的新型合成纳米颗粒和改性对口腔细菌的抗菌作用

Tab.1　Anti-oral bacterial effect by the novel synthesized nanoparticles and modifications of titanium

Article	Materials	Evaluation	Results
Choi J Y, et al[10]	TiO_2 (Rutile and anatase) oxidation thermal and anodic Near-ultraviolet (UV) light UVA	Lactobacillus acidophilus	The antibacterial activity of the coated specimens was similar to that of the uncoated group.
Al-Serwi R H, et al[11]	Newly developed bioinorganic titanium dioxide (Bi-TiO_2)	Methylene blue degradation *E. coli* and *S. aureus*	Effective against Gram-positive bacteria Docked with bacterial thymidylate kinase (TMPK: potential therapeutic goal for the preparation of novel antibacterial drugs), rosmarinic acid, vicenin-2, orientin, vitexin, and isoorientin.
Wang R X, et al[12]	Titanium dioxide/nano-hydroxyapatite composite Dental curing lamp (385~515 nm)	Biofilm formed by *S. mutans* Enamel surface remineralization	Five minutes exposure of light *S. mutans* significantly reduced. Promote enamel mineralization by release of calcium and phosphorus ions The Ca/P ratio of demineralized enamel surface increased after 15 days.
Westas E, et al[13]	Titanium discs were spin-coated with photocatalytic TiO_2 nanoparticles UVA	Bacterial viability of *S. oralis*	After 24 h of exposure, the proportion of viable cells was reduced by 40% on the discs compared to 0 and 6 h.
Guo J N, et al[14]	Acridinium-grafted g-C_3N_4 nanosheets Ultraviolet to near-infrared region	Gram-negative and gram-positive bacteria	Effective sterilization and biofilm elimination activities against both gram-negative and gram-positive bacteria Novel material may move toward, tile and insert the bacterial lipid bilayer membrane.
Kunnamareddy M, et al[15]	Cobalt Sulfur codoped Titanium Dioxide (Co-S codoped TiO_2) photocatalysts	Methylene blue degradation Catalytic activity for *E. coli* and *S. aureus*	Exhibited strong antibacterial activity

2.2 Application of photocatalyst for dental materials

2.2.1 Dental implant

Dental implants involve the insertion of synthetic roots into the jawbone where teeth have been lost and the fabrication of artificial teeth thereon. The major materials used for the dental implant is now titanium. At initial stage, antibacterial activity of photocatalyst coating or surface treatment of titanium were confirmed [16-19]. Other than antimicrobial activity, effect of photocatalysis on bone formation was also investigated [20-24](Tab. 2).

2.2.2 Resin

Poly methyl methacrylate is often used in clinical dental practice. It was used as denture base, provisional restoration, artificial teeth on denture base etc. Two studies investigated the antimicrobial effect of photocatalyst on resin [25-28]. One article investigated the effect of photocatalyst on the curing reaction [26] (Tab. 3).

2.2.3 Other dental materials

By the tooth extraction, wearing ill fit denture, or inflamed gum disease, the gum attached to the denture surface becomes sensitive. Tissue conditioner is temporally used on the mucosal surface of the denture until the inflammation of gum is healing. Two articles investigated the antimicrobial effect of photocatalyst applying tissue conditioners[29-30]. One article investigated the effect of the photocatalyst containing toothpaste[31](Tab. 4).

2.3 Dental clinical procedures

For dental clinical procedures, 5 articles were applied photocatalyst for whitening/bleaching (Tab.5). Teeth whitening involves bleaching teeth to make them lighter. It can't make teeth brilliant white, but it can lighten the existing color by several shades. At initial stage, three articles investigated the degradation of pigments by photocatalyst[32-34]. Then materials used for the teeth bleaching were modified[35-36].

表 2 有关种植体的光催化剂研究

Tab.2 Photocatalyst research for implant

Article	Materials	Evaluation	Results
Suketa N, et al[16]	Titanium discs coated with photocatalytic TiO_2 nanoparticles, Ultraviolet-A	Biofilm formation of *S. oralis*	After 24 h UVA of exposure, the proportion of viable cells was reduced by 40% on the TiO_2 coated discs compared to 0 and 6 h.
Ahn S J, et al[17]	Surfaces preparation: anodized at 270 V, heat-treated, and machined UV illumination	Adhesion and survival rates of *S. sanguinis*	Significantly reduced bacterial adhesions were observed by anodized and heat-treated surfaces. The photocatalytic bactericidal effect was maximal on the anodized surface.
Kawano T, et al[18]	A newly developed blue-violet semiconductor laser (λ = 405 nm)	Contact angle Methylene blue degradation Adherence of *P. gingivalis* Osteoblast and fibroblast proliferation Histological examination after implantation in rats	The contact angle was significantly decreased. Methylene blue was significantly degraded. The number of attached *P. gingivalis* organisms was significantly reduced. New bone was observed around exposed specimens in the histological evaluation.
Yamada Y, et al[19]	Titanium disks Ultraviolet-C	*S. aureus, S. pyogenes*	Ultraviolet-C irradiation reduced the attachment and biofilm formation.
Hirakawa Y, et al[20]	Anatase TiO_2 film with plasma source ion implantation followed by annealing UV-A for 24 h	Serum fibronectin attachment Hydrophilicity Initial osseointegration responses in the dog mandibles	Strong hydrophilicity under UV-A illumination and serum fibronectin attachment were observed. The experimental implants enhanced the bone formation with the bone-to-implant contact of 42.7% after 2 weeks of healing (control: 28.4%).
Yeniyol S, et al[21]	Anatase/rutile mixed-phase TiO_2 thin films via the electrochemical anodization and annealing	Rhodamine B dye degradation *A. actinomycetemcomitans* colonization	Twenty-six percent photocatalytic decolorization toward Rhodamine B dye solution. *A. actinomycetemcomitans* colonization was significantly decreased.
Nagay B E, et al[22]	TiO_2 coated by nitrogen and bismuth via plasma electrolytic oxidation Visible light	Hydrophilicity Albumin adsorption Degradation of methyl orange	Treated titanium surfaces showed greater hydrophilicity and albumin adsorption. Incorporated Bismuth into the gap of TiO_2 facilitated higher degradation of methyl orange in the dark, and visible-light irradiation.
Venkei A, et al[23]	TiO_2 Ag-TiO_2 Visible light	*S. mitis*	Ag-TiO_2 containing surfaces showed photocatalytic activity and eliminated the highest amount of bacteria.
Al-Serwi R H, et al[11]	Ultraviolet-C Ultraviolet-A	Bacterial accumulation: *S. aureus* and *S. pyogenes*	Ultraviolet-C irradiation was no effect on the bacterial attachment and accumulation Ultraviolet-C irradiation changed wettability to superhydrophilicity and reduced carbon contents on any titanium surface.
Janani B, et al[24]	Surface preparation: anodized at 270 V, heat-treated, machined UV illumination	Bacterial adhesion	Anodized and heat-treated surfaces showed significantly decreased bacterial adhesion.

表 3 有关树脂的光催化剂研究

Tab.3 Photocatalyst research for resin

Article	Materials	Evaluation	Results
Cheng Y. et al.[25]	Photocatalyst mixed in a denture base resin (PMMA-MMA) Photocatalyst: Ca(II) in the hydroxyapatite was changed to Ti(IV) UV irradiation	Methylene blue degradation	Photocatalyst containing denture resin significantly degrade methylene blue.
Ingrosso C. et al.[26]	Resin coated with colloidal TiO_2 nanorods	Curing reaction	TiO_2 behaves as a free-radical donor in the curing reaction, accelerate the cure rate and increase cross-linking density, promoting multiple covalent bonds between the resin prepolymers.
Kuroiwa A. et al.[27]	A coating of a UV-responsive photocatalyst, titanium dioxide (TiO_2), was coated to an orthodontic resin	*S. mutans, S. sobrinus, S. gordonii, S. oralis, S. mitis*	Bacterial CFU were markedly decreased in photocatalyst coating sample with UV irradiation.
Shibata T. et al.[28]	Denture base resin coated hydroxyapatite-TiO_2	*C. albicans*	Hydroxyapatite-TiO_2 had statistically significant antifungal properties.

PMMA: Polymethyl Methacrylate, MMA: Methyl Methacrylate, CFU: Colony Forming Unit

表 4　有关义齿软衬材料和牙膏的光催化剂研究

Tab.4　Photocatalyst research for tissue conditioner and toothpaste

Article	Materials	Evaluation	Results
Akiba N, et al[29]	Tissue conditioner containing TiO_2 UV irradiation	*C. albicans*	Ninety min irradiation, *C. albicans* was significantly reduced, however, it remained on the coated surfaces.
Uchimaru M, et al[30]	Tissue conditioner containing a photocatalyst UV-irradiation	*E. coli, S. mutans, S. aureus* *C. albicans*	Tissue conditioner containing photocatalyst showed significant antibacterial effect.
Minato K, et al[31]	Toothpaste containing hydroxyapatite-tryptophan complex with gray titania visible light	*P. gingivalis*	Toothbrushing resulted in coat peeling and reduced the antibacterial effect.

表 5　有关牙齿美白或漂白的光催化剂研究

Tab.5　Photocatalyst research with tooth whitening/bleaching

Article	Photocatalyst	Evaluation	Results
Suyama Y, et al[32]	Hydrogen peroxide with visible light-activated TiO_2 or ultraviolet light-activated TiO_2 Low- and high-intensity halogen lamps or blue LED LCUs	Methylene blue	High-intensity halogen with hydrogen peroxide with visible light-activated TiO_2 caused the most significant reduction in Methylene blue concentration.
Kishi A, et al[33]	Visible-light activating titanium dioxide photocatalyst A halogen light unit or an LED unit with violet mode or blue mode	Stained by black tea	LED unit with violet mode showed better bleaching effect.
Tano E, et al[34]	405 nm diode laser on bleaching reaction of visible light-activated TiO_2	Methylene blue	Using 405 nm diode laser, the bleaching effects of visible light-activated TiO_2 depended upon the irradiation time and the average output power, regardless of pulse duration or duty cycle.
Iqbal A, et al[35]	TiO_2-modified ZnO quantum dots (QDs) were immobilised on a linear low-density polyethylene (LLDPE) polymer	Tetracycline	The highest removal (89.5%) of TC (40 mg/L) was achieved within 90 min at pH 9.
Li Q, et al[36]	Injectable sodium alginate hydrogel membrane doped with bismuth oxychloride and cubic cuprous oxide nanoparticles green light	Biofilm removal	Eradicate the biofilm and whiten the tooth non-destructively.

3　Discussion

In this article, we systematically reviewed the article concerned with photocatalyst applying for the dental research and clinical practice.

In the treatment of oral diseases, lesions in oral cavity can be directly accessible. Complex drug delivery systems are not necessary. In addition, dentistry has various advanced dental materials. Restorative and prosthetic treatments are major dental practice. Fine dental materials are indispensable for these treatments. These two major characteristics of dentistry make the application of novel materials for the dental treatment easy. All of the articles reviewed in this study are concerned with the dental materials. Among the 27 articles reviewed in this report, 10 articles were concerned with dental implant.

Dental implant, which is an artificial root of the teeth, consisted of titanium. Therefore, photocatalyst application for the surface of the implant is technically easy. Biocompatibility and antimicrobe are important factors for the prognosis of dental implant. The prognosis of dental implant mainly depends on the infection in the gap between dental implant and supportive tissue[37]. It is called as peri-implantitis or peri-implant disease. Peri-implantitis has become a global burden[38-39]. Prevalence of peri-implantitis were 1%～47%[40], 29.48%[41] or 80%[42]. Eight articles out of ten concerned with dental implant investigated antimicrobial effect of the photocatalyst. Photocatalyst effectively reduced bacterial adhesion[17-19,21,24], biofilm formation[16]. After insertion of dental implant into the jaw bone, irradiation to obtain the photocatalystic effect is impossible. However, complete sterilization is indispensable during the dental implant surgery. Bacterial contamination largely effects on the prognosis of dental implant[43]. Even the sort of time periods, contentious antimicrobial effect of photocatalyst may effectively work for the prevention of contamination during the surgery.

Four articles reported the application of photocatalyst for resin. In dental practice, PMMA is frequently used. One of the characteristics of PMMA is hydrophilia. Absorbency of complex oral fluid makes the prothesis and provisional restorations filthy. In addition, microbes colonize on the surface of PMMA. Coating photocatalyst on the surface of resin effectively works for the sanitization of the dental materials [27-28]. In addition, it promotes the curing reactions of PMMA[26].

There were two articles concerned with tissue conditioner [29-30]. Tissue conditioner is a denture lining material for inflated gum that support the denture. The major component of tissue conditioner is a gel type PMMA. Even through application of tissue conditioner is temporal, it is easy to be staining, coloration, stinking and moulding. Including photocatalyst in tissue conditioner has antifungal effect. Until the inflammation of gum heeling, patients visit dental clinic. Irradiation can be applicable during the patient visit dental clinics. Effect of photocatalyst may be desirable to keep tissue conditioner clean.

Whitening/bleaching the discolored teeth improve the life quality of the patients[44]. Hydrogen peroxide is the major reagent for whitening/bleaching. Photocatalyst enhances the removal of the stain on tooth surface[32-34]. It is also effective for discolored teeth by tetracycline[35]. Taking tetracycline during fetus or infant age pigments sediment inner teeth[7]. Photocatalyst containing whitening/bleaching materials are already packaged and commercially available. As UV irradiation is incapable for human oral cavity, the research concerning whitening/bleaching were focused on the safety and effectiveness of the light source[32-34].

Several articles are concerned with application of photocatalyst for dental research and dental clinical practice. However, information is not enough. Photocatalyst is a useful tool for dental clinical practice. For the application of photocatalyst for dental clinical practice, accumulation of evidences and commercialization of dental materials are necessary.

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