Hybrid ZnO-AuDAPT Nanoparticle Coating for Enhanced Antimicrobial Performance of Clear Orthodontic Aligners

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Clear aligners have become increasingly popular as orthodontic alternatives to traditional fixed appliances due to their transparency, comfort, and removability. Primarily manufactured from polymers like polyethylene terephthalate glycol (PETG) and thermoplastic polyurethane (TPU), these materials offer desirable mechanical characteristics such as flexibility, strength, chemical resistance, superior transparency, impact resistance, elasticity, and abrasion resistance [1]. However, a significant limitation persists: clear aligners are vulnerable to bacterial colonization and biofilm formation, potentially causing dental complications like enamel demineralization and gingival inflammation. Addressing this issue is crucial for improving patient outcomes and maintaining oral health.

Recent antimicrobial strategies for clear aligners, including zinc oxide (ZnO), magnesium oxide, and quaternary ammonium compound coatings, have shown some success but also notable drawbacks. ZnO coatings maintain transparency yet rapidly lose effectiveness under oral conditions, magnesium oxide may discolor aligners, and quaternary ammonium compounds lack proven long-term efficacy. Additionally, gold-based coatings like AuDAPT demonstrate strong anti-biofilm properties but have limitations due to distinct purple coloration and potential health risks from thiol group leaching.

To overcome these challenges, our proposed solution employs a hybrid antimicrobial coating, strategically combining ZnO nanoparticles with low concentrations of AuDAPT embedded within a UV-crosslinked acrylic matrix. This approach aims to leverage ZnO's robust antibacterial properties alongside AuDAPT's powerful anti-biofilm capabilities. Utilizing a low concentration of AuDAPT with dip-coating methods significantly reduces coloration and potential thiol-related health concerns. The UV-crosslinked acrylic matrix ensures improved durability, transparency, and nanoparticle retention throughout the aligner's typical 7-day wear cycle. This innovative coating strategy provides a long-lasting, aesthetically acceptable, and biocompatible solution, significantly enhancing clear aligners' antimicrobial performance and patient oral health outcomes.

Studies have explored coatings such as zinc oxide, magnesium oxide, and quaternary ammonium compounds to reduce bacterial buildup on clear aligners. While these materials show antibacterial effects, each has notable limitations. Zinc oxide coatings maintain transparency but lose much of their effectiveness after 24 hours of simulated brushing and oral conditions [1]. Magnesium oxide can lead to visible discoloration, and quaternary ammonium coatings lack long-term data in real-world use [5]. Gold-based coatings, including AuDAPT, show strong anti-biofilm performance and good biocompatibility [4], but are limited by their purple coloration and possible health concerns from thiol groups.

These findings highlight a gap in existing solutions. There is still a need for a coating that is long-lasting, transparent, and safe for daily use. Our proposed approach combines zinc oxide with a low concentration of AuDAPT in a UV-crosslinked acrylic matrix to improve durability, limit discoloration, and reduce the risk of chemical leaching. This hybrid strategy aims to maintain antimicrobial effectiveness throughout a typical 7-day aligner wear cycle. While this approach addresses key weaknesses in current coatings, further testing is needed to confirm long-term safety and performance under real use conditions.

We propose a hybrid coating is the combination of the antibacterial power ZnO and the anti-biofilm efficacy of AuDAPT, aiming for a synergistic effect against a broader range of oral bacteria. To address the purple coloration and potential health concerns from AuDAPT's thiol groups, we will use low concentration of AuDAPT and a dip-coating method to maintain transparency and coating uniformity. For durability, an acrylic component or carbon black material is added to the nanoparticle solution. Upon UV irradiation, it undergoes crosslinking to form a robust matrix that anchors ZnO and AuDAPT onto the aligner surface, improving wear resistance and nanoparticle retention over a 7-day period.

All the materials have been selected for their unique properties and their contribution to the final coating's result. ZnO nanoparticles have been incorporated into the coating due to their renowned antibacterial, antifungal, and antiviral properties. These properties are achieved by Zn^{2+} ion release, the generation of reactive oxygen species, and the direct interference of bacterial cell membranes. These nanoparticles are also biocompatible and naturally transparent and are therefore very useful for use in clear aligners where antimicrobial effectiveness and aesthetic optimization is key.

AuDAPT is incorporated for its anti-biofilm and antibacterial effect, specifically against drug-resistant oral pathogens. Its ability to inhibit biofilm growth is crucial for preventing bacterial growth, especially from chronic bacterial colonies, on the aligner surface. There are some problems associated with the application of AuDAPT in dental coatings. The first issue is its potential to leave a purple coloration on the aligner. This discoloration is a result of the optical properties of the gold nanoparticles themselves; 4,6-diamino-2-pyrimidine thiol-modified gold nanoparticles possess a conventional surface plasmon resonance (SPR) which results in a maximum absorbance at a wavelength of around 510 nm. This absorbance is a direct cause of the purple/reddish hue in solutions or coatings. The intensity of this coloration is directly correlated with the concentration of the particles and the size of the aggregates: higher concentrations and larger aggregates can lead to a deeper hue. There are some safety concerns with the thiol functional groups present in AuDAPT. These groups can potentially leach out of the coating and lead to biocompatibility problems if not properly contained.

To tackle these issues, our approach uses AuDAPT at a low concentration, thus preventing the color change from being observed while optimizing the anti-biofilm action/antibacterial properties. In addition, to obtain a thin, uniform coating, a dip-coating process will also be used. Arguably most important, AuDAPT is integrated into a UV-crosslinked acrylic (or alternatively a carbon black material) matrix. This crosslinked structure anchors the nanoparticles securely to the aligner service to enhance their retention and wear resistance. This structure also confines the AuDAPT and significantly minimizes the risk of leaching of both the gold nanoparticles and thiol groups.

This approach addresses current limitations of nanoparticle coatings particularly in short lifespan and discoloration which provides a long-lasting, clear, and safe antimicrobial coating suitable for clinical use in clear aligner treatments.

To verify that our coating performs as intended, we propose the following characterization and testing methods:

1. Optic Characteristic (Transparency and Band Gap):
   UV-Vis spectroscopy and photoluminescence (PL) analysis will be used to evaluate the transparency and optical properties of the coating. These techniques can help determine the optical band gap, especially if the coating exhibits visible coloration (e.g., a purple hue). PL can further confirm the presence of multiple band gaps or defect states, providing insight into the material's optoelectronic behavior. Both methods offer fast and effective characterization of optical properties.
2. Coating Characteristic:

    - Alpha-step profilometer is suitable for measuring film thicknesses around 100 nm and provides a straightforward approach to quantifying uniformity.
    - Atomic Force Microscopy (AFM) will be used to examine the nanoscale surface morphology and can provide localized thickness measurements with atomic-level resolution.
    - For more detailed structural analysis, cross-sectional Scanning Electron Microscopy (SEM) or Transmission Electron Microscopy (TEM) can be employed following alpha-step or AFM analysis.
3. Material Uniformity:
   The uniformity of ZnO within the coating can be assessed using Energy-Dispersive X-ray Spectroscopy (EDX) mapping in both SEM and TEM, providing elemental distribution and identifying any agglomeration or non-uniform deposition.
4. Adhesion Testing:
   The coating's adhesion to the substrate will be evaluated using standard tape tests and scratch tests to assess mechanical robustness under stress.
5. Crosslinking Efficiency:
   Fourier-transform infrared spectroscopy (FTIR) will be used to monitor changes in functional groups before and after UV irradiation, indicating the degree of crosslinking.
   Raman spectroscopy and X-ray Photoelectron Spectroscopy (XPS) will provide further information on bonding types, including carbon bonding states and interactions between ZnO and the polymer matrix.
6. Antimicrobial Testing:
   Aligners coated with our hybrid film, along with uncoated controls and samples coated with ZnO or AuDAPT alone, will be incubated with relevant oral bacteria. The bacteria will be fluorescently labeled, and confocal microscopy will be used to visualize colonization. Image analysis software such as Ilastik will be applied to quantify bacterial counts and assess antibacterial performance.
7. Biocompatibility (Cytotoxicity Assays):
   In vitro cytotoxicity will be assessed by exposing human oral cells to coated samples. Cell viability, proliferation, and morphology will be evaluated using standard assays (e.g., MTT or live/dead staining) to ensure the coating does not compromise biocompatibility.
8. Durability and Longevity Testing:
    - Mechanical durability will be tested by subjecting coated aligners to a 7-day simulated brushing routine to mimic daily oral hygiene practices.
    - Chemical stability will be evaluated by exposing coated samples to artificial saliva at room temperature for 7 days to assess resistance to degradation and leaching under physiologically relevant conditions.

Our proposed hybrid ZnO-AuDAPT nanoparticle coating holds significant promise for consumers, the environment, and the clear aligner manufacturing industry. For consumers, or patients in our case, the enhanced antimicrobial efficacy offered by synergistic action of ZnO and AuDAPT promises reduced risk of developing oral issues like gingivitis and enamel demineralization which are caused by P. gingivalis biofilm formation on the current state-of-the-art. These benefits can be seen without compromising the aesthetic transparency and comfort that consumers desire. By proactively implementing our solution, biofilm formation will be severely inhibited lowering the patient's potential future dental treatments, thus reducing the frequency of dental visits for oral hygiene.

From an environmental point of view, the use of UV-crosslinked acrylic components provides a thin yet durable coating which works to minimize material waste. Moreover, both zinc and gold are relatively inert and, as our proposal indicates the use of a low concentration of AuDAPT, the thiol concentration and its effect on the environment would be minimized. Despite these benefits, life-cycle assessments should be conducted to confirm that added manufacturing steps do not introduce net energy costs or generate harmful chemical byproducts when scaled.

In terms of manufacturing, our proposal is feasible without requiring intense equipment modifications. As we are proposing a coating, a post-form dip coat step followed by the proposed UV crosslinking could be applied directly after the current manufacturing process without intense capital reinvestment. Also, the aforementioned material compatibility between PETG and TPU allows the product to tolerate the acrylic nanoparticle formation. Limitations of the proposal include potential regulatory hurdles for nanoparticle-coated medical devices, specifically highlighting the need for long-term biocompatibility studies. Also, it is unknown if the adhesion between the acrylics and TPU would pose an issue, warranting further study in this area. Future work should focus on the thiol-modified AuDAPT component, as well as optimizing the nanoparticle size and potential surface chemistry to further minimize discoloration and leaving any trace chemicals from the coating well below FDA regulated toxicity thresholds. Importantly, clinical trials are also essential to validate antimicrobial performance under real-world wear and cleaning routines.

Overall, the benefits to adopting our hybrid ZnO-AuDAPT coating, including enhanced patient outcomes, potential reductions in dental interventions, and relatively straightforward manufacturing modifications strongly support its development. If adoption of our proposal is halted, it is most likely that the regulatory framework for nanoparticle-coated dental appliances requires robust long-term data. Should this hurtle persist, it may be worth looking into alternative strategies focused on optimizing the properties of the polymer bulk, such as intrinsic antimicrobial monomers. While we believe following the proposed Hybrid ZnO-AuDAPT Nanoparticle Coating is the most promising solution, other biocompatible antimicrobial agents such as quaternary ammonia compounds can be explored if required, achieving comparable biofilm resistance without the use of nanoparticles.

References:

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