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Advances in Dental Remineralization Materials: From Conventional Fluoride to Novel Bioactive Materials

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Abstract. Dental caries develops when demineralization outpaces remineralization in dental hard tissue. Fluoride has the strongest evidence base for preventing caries and promoting remineralization, but it supplies no mineral ions of its own, acts mainly at the lesion surface, and cannot rebuild lost structure. Casein phosphopeptide–amorphous calcium phosphate (CPP-ACP), nano-hydroxyapatite, bioactive glass, and polymer composites have been proposed as non-invasive options for early lesions. This review first sets out how fluoride limits demineralization, drives mineral redeposition, and raises the acid resistance of enamel. It then looks at how CPP-ACP stabilizes and delivers calcium and phosphate, how nano-hydroxyapatite offers nucleation sites, and how bioactive glass releases ions and changes the local microenvironment. Polymers are discussed as ion carriers, particle stabilizers, and biomimetic templates, including their use in polymer–inorganic composites. Set against fluoride, the newer materials look more like adjuncts than replacements. Controlled ion release, biofilm modulation, responsive design, and individualized combinations are the main directions ahead. Most of the evidence, though, is in vitro or short term, formulations and outcome measures differ widely, and long-term safety and clinical data are still thin.

Keywords: Dental remineralization, Fluoride, Bioactive materials, Biomimetic materials, Dental caries

1. Introduction

Dental caries is among the most common chronic diseases of the mouth. Global Burden of Disease data show that untreated caries in permanent teeth is still the most prevalent oral condition worldwide, affecting both children and adults [1]. Enamel is the most heavily mineralized tissue in the body, built mainly from hydroxyapatite, and once a real structural defect forms it cannot grow back. The disease starts in plaque. Cariogenic bacteria ferment dietary carbohydrates into organic acids, the local pH drops, and calcium and phosphate keep leaving the enamel. This loss of mineral is demineralization. Left alone, a lesion keeps advancing. It affects chewing and appearance, and it can eventually reach the pulp and cause pulpitis. That is why restoring mineral to early lesions, before the enamel is damaged beyond repair, has become a central strategy in early caries management.

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Fluoride has long been the main tool for preventing caries and for treating early lesions without drilling, and it remains the best-supported basis for remineralization [2]. It is delivered in toothpastes, gels, and varnishes. The fluoride ions slow mineral dissolution during acid attack and help calcium and phosphate redeposit as fluorapatite, which resists acid better than the original mineral. In practice this strengthens enamel against acid erosion and builds up mineral in the surface layer of early lesions. The approach has clear limits, though. How well it works depends heavily on the free calcium and phosphate in saliva [3]. Deposition also tends to concentrate at the lesion surface, which may keep ions from reaching the deeper, more damaged microstructure and leaves substantial defects largely unrepaired. And frequent or excessive exposure to high-concentration fluoride carries a risk of dental fluorosis and excess fluoride intake.

Over the past few years, a number of bioactive materials have entered the field. Some are meant to raise the calcium and phosphate available at the lesion, others to serve as templates for mineral nucleation, and they may help recover mineral deeper in the lesion body. Nano-hydroxyapatite (nHA) has a crystal size close to that of natural enamel mineral. It can slip into the micropores between enamel rods after demineralization and pull in calcium and phosphate to grow new crystals in place, which gives deeper and more even deposition [4]. Bioactive glass exchanges ions at its surface when it meets saliva, releasing calcium, phosphate, and silicon; the local pH rises and a mineralized layer forms on the lesion surface [5]. Casein phosphopeptide–amorphous calcium phosphate (CPP-ACP) uses casein phosphopeptide to bind and stabilize amorphous calcium phosphate (ACP) and carry calcium and phosphate to the tooth surface. Free ACP clumps easily and is unstable, so working formulations usually add polymer-type stabilizers [6]. What these materials have in common is a closer imitation of natural mineralization and, potentially, more sustained mineral release and tissue repair. The evidence is still mostly in vitro or short term, however, and long-term stability and clinical benefit have yet to be shown. This review summarizes where remineralization materials stand today. It covers how conventional fluoride and the newer bioactive materials work in enamel repair, what each does well and badly, and which challenges remain, and it closes with an outlook for caries prevention and oral health.

2. Application of fluoride in dental remineralization

2.1. Mechanisms of fluoride-mediated enamel remineralization

Fluoride is one of the best-established remineralization strategies in caries prevention and early lesion management, and modern preventive dentistry rests heavily on it [2, 3]. Caries is what happens when the balance between acid-driven demineralization and the remineralization supplied by saliva and local mineral ions tips the wrong way. Once bacteria produce organic acids, the pH at the tooth surface falls, the mineral phase of enamel starts to dissolve, and calcium and phosphate leave the crystal. If demineralization goes on long enough that remineralization cannot make up the loss, early lesions such as white spot lesions appear. The aim of remineralization therapy, then, is to rebuild a local chemical environment in which mineral can be redeposited.

Fluoride shifts this balance in several linked ways. One is that fluoride ions join in reorganizing the mineral phase at the enamel surface, so that new mineral resists acid better. The classic model describes some of the hydroxyl groups in hydroxyapatite being replaced by fluoride, giving fluorapatite or a fluoride-rich, fluorapatite-like structure. Fluoride-rich apatite holds up better in acid than ordinary hydroxyapatite, so later acid attacks remove less mineral. Fluoride also influences dissolution and deposition at the crystal surface, which makes the local environment more favorable for redepositing calcium, phosphate, and other ions. At clinically relevant concentrations, however,

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protection does not mainly come from forming large amounts of stoichiometrically pure fluorapatite deep in the tooth. It comes more from the steady presence of low fluoride concentrations at the tooth surface and in saliva and plaque fluid, and from calcium fluoride–like reservoirs that high-concentration topical products leave behind.

In short, fluoride does three things. It lowers the solubility of apatite in acid. It promotes redeposition of calcium and phosphate onto crystal surfaces that remain. And under some conditions it suppresses acid production by plaque microorganisms. Together these tilt the demineralization–remineralization balance toward a net gain in mineral.

One point needs emphasis: fluoride does not supply minerals for remineralization. Enamel still needs enough calcium and phosphate, so fluoride is better described as an active factor that promotes and regulates redeposition. This matters when comparing old and new materials. Fluoride mainly makes better use of mineral ions already present in the tooth and oral fluids, whereas many newer bioactive materials try to answer further questions: where calcium and phosphate come from, how to keep them stable, and how to deliver them into deeper parts of a lesion. Whether calcium and phosphate supply really limits remineralization depends on salivary status, the plaque microenvironment, lesion depth, and the formulation, so it should not be treated as a universal bottleneck.

2.2. Clinical forms of fluoride and their effects

Clinically, fluoride is delivered topically, as toothpaste, gel, or varnish. Toothpaste has by far the widest daily use. Its main value is keeping fluoride levels at the tooth surface up through frequent, low-intensity exposure, which suits the chronic, ever-shifting nature of caries better than a single intense treatment.

Fluoride toothpaste reduces enamel demineralization and also promotes remineralization in early lesions, and for this reason it is central to caries prevention in the general population. Concentration and mode of use have to be adjusted to age, caries risk, and level of exposure. In children especially, the purpose of fluoride is to set up long-term prevention, not to repair, with one high-concentration treatment, structural damage that has already formed.

Gels and varnishes are used more for professional prevention and for patients at high caries risk. A varnish creates a locally high fluoride concentration on the tooth surface and, under some conditions, a calcium fluoride–like reservoir that releases fluoride during later acid attacks [2]. Existing systematic reviews broadly support professionally applied topical fluoride for preventing caries and controlling lesions in particular populations, though results are not fully consistent across products, dentitions, and risk groups. One systematic review of 33 studies and more than 16,000 children found that professional fluoride intervention helped prevent and control caries in some children, but the size of the effect depended on the product and the clinical setting. A higher fluoride concentration, in other words, is not automatically more effective.

The goals of fluoride use fall into three levels. The first is primary prevention: keeping enamel acid resistance up before any obvious lesion appears. The second is control of active non-cavitated lesions, above all white spot lesions and early enamel demineralization. The third is managing the risk of continuing demineralization, where regular exposure keeps fluoride ions bioavailable in the mouth and lowers the chance that a lesion progresses.

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2.3. Advantages and limitations of conventional fluoride strategies

Fluoride has kept its central clinical position for two main reasons, the first being its evidence base and availability. Compared with the many materials still at the laboratory or early clinical stage, it has a long record of use, and how to use it and what it prevents are well understood. It is also inexpensive and comes in many forms, so it fits into everyday household oral care, which is a real advantage for public health [2].

None of this means fluoride can handle every kind of enamel damage on its own. Its most obvious weakness is limited ability to repair tissue with marked structural defects. When a lesion extends past the surface layer into the body of the enamel, supplying fluoride ions from the outside will not necessarily restore the full three-dimensional microstructure. More importantly, fluoride-driven remineralization still relies on local calcium and phosphate, so its effect can be restricted where mineral ions are hard to come by [3].

Exposure is the other issue. The effect of fluoride does not follow a simple rule that more is better; there is an effective range and a safe range. For children, the route of intake and total exposure need particular attention. High fluoride exposure is clearly linked to dental fluorosis, which is why fluoride use has to be standardized, not escalated to ever higher concentrations.

Overall, conventional fluoride strategies have improved control of early caries by raising the acid stability of the mineral phase, inhibiting demineralization, and promoting mineral redeposition. Limited deep structural repair and dependence on calcium and phosphate supply remain the main shortcomings. The way forward is therefore not simply a higher fluoride dose but combining fluoride with calcium–phosphate supplementation, nanomaterials, and biomimetic delivery systems.

3. Application of novel bioactive materials in dental remineralization

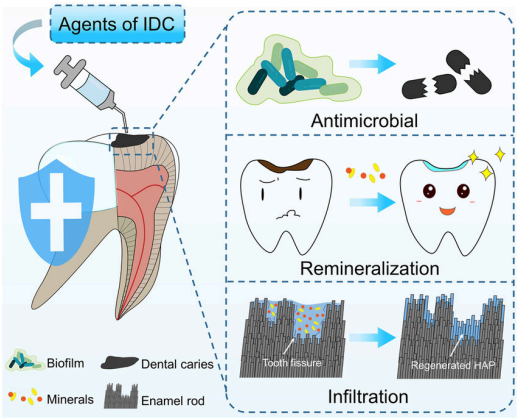


Figure 1. Caries management under the "interrupting dental caries" (IDC) concept, combining antimicrobial, remineralization, and infiltration approaches [8]

Conventional fluoride does promote remineralization to a degree, but it cannot fully deal with the supply of mineral ions, delivery into deep lesions, or reconstruction of natural structure. As biomaterials science and nanotechnology have advanced, attention has moved to newer materials that supply calcium, phosphate, and other mineral elements directly, or that copy natural tooth mineralization more closely [7]. Three systems have drawn the most study: CPP-ACP, nano-hydroxyapatite, and bioactive glass. None is a simple substitute for fluoride. They differ from it, and from each other, in indications, mechanism, and how mature the evidence is. Figure 1 shows where

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remineralization sits among the strategies used to interrupt caries, alongside antimicrobial and infiltration approaches [8].

3.1. Casein phosphopeptide–amorphous calcium phosphate (CPP-ACP)

CPP-ACP is a typical remineralization system that supplies calcium and phosphate. The idea is not to hand over finished crystalline hydroxyapatite. Casein phosphopeptide stabilizes amorphous calcium phosphate, so that highly reactive calcium and phosphate species stay in a form the tooth can use more readily [3].

ACP has high biomineralization potential, but it is thermodynamically unstable and readily turns into more stable crystalline calcium phosphates [9]. CPP is rich in phosphate groups and forms stable nanocomplexes with calcium ions and ACP. That delays the quick transformation of the amorphous phase somewhat and keeps calcium and phosphate highly available locally. In the mouth these complexes raise the effective concentration of calcium and phosphate near the tooth surface and provide raw material for mineral deposition in early demineralized areas.

What sets CPP-ACP apart from fluoride is that it goes straight at the shortage of calcium and phosphate. It does not just duplicate fluoride's action; it raises the local remineralization potential by supplying the two ions. Recent clinical studies and systematic reviews broadly suggest some value for early enamel remineralization, but clinical superiority has not been firmly established [10]. Studies differ widely in lesion type, in whether fluoride was used alongside, and in outcome measures, and some carry a high or unclear risk of bias. For now CPP-ACP is better seen as a possible adjunct than as a standard treatment that has displaced fluoride.

The effect of CPP-ACP also depends on how stable the material is and on the oral environment. Protein-derived peptide systems can be affected by oral enzymes, the plaque environment, and mechanical clearance, so the material does not stay on the tooth surface indefinitely. Early laboratory work and clinical studies also differ markedly: mineral gain under experimental conditions does not necessarily mean a patient's long-term caries risk drops significantly. In judging clinical value, lesion arrest or reversal, caries increment, and long-term adverse effects should come first, ahead of surrogate laboratory endpoints.

3.2. Nano-hydroxyapatite (nHA)

nHA is a different technical route. Natural enamel is essentially highly ordered apatite nanocrystals, so what makes nHA attractive is not just that it contains calcium and phosphate, but that its nanoscale form is so close to natural enamel mineral in structure and chemistry [4].

nHA can interact with damaged enamel through surface adsorption, particle filling, and mineral deposition. The large specific surface area of nanoparticles offers plenty of active surface and may let them act as nucleation sites for crystal growth. In early enamel lesions, nHA may fill some of the microscopic pores and encourage new mineral phases to form in the lesion. Particles entering pores or forming a deposited layer on the surface is not the same as rebuilding the ordered hierarchical structure of natural enamel rods, though, and structural and long-term functional evidence is still needed.

Systematic reviews on white spot lesions show good potential for nHA in improving surface hardness and mineral content in early lesions [10], and some studies found pure nHA outperforming fluoride alone on certain measures [11]. The great majority of studies are still in vitro or short term, however, clinical trials are few, and heterogeneity between studies is large. It is too early to say that nHA is better than fluoride across the board.

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nHA also suffers from particle agglomeration and weak dispersion stability. Nanoparticles have high surface energy and readily form aggregates in liquids through forces such as van der Waals interactions. Once agglomeration is marked, the effective specific surface area and the ability to enter tiny lesion pores may drop. Research on nHA therefore has to look beyond remineralization potential to dispersion stability, retention on the tooth surface, penetration into the lesion body, and long-term clinical effect.

3.3. Bioactive glass (BAG)

BAG works differently from CPP-ACP and nHA. It supplies calcium and phosphate to the local environment, and it can also alter the local chemistry through ion exchange between the material and oral fluids. When a typical BAG meets an aqueous phase, ion exchange, network hydrolysis, and surface reactions follow. Calcium and phosphate gradually accumulate near the material surface and go on to form a hydroxycarbonate apatite–like layer [5].

This gives BAG the potential to induce mineral deposition in situ at its surface. Conventional materials usually depend on the surroundings to provide enough mineral ions; BAG can act as an ion source itself, so in theory it can create more favorable conditions for mineralization where minerals are scarce. Some BAG systems also raise the local pH by releasing ions such as sodium and calcium, and may affect the growth or metabolism of cariogenic biofilm. These effects depend on glass composition, ion release concentration, and experimental conditions, however, and cannot be equated directly with clinical antibacterial or caries-preventive effects.

Early systematic reviews found that BAG increased enamel remineralization in vitro and tended to beat conventional materials on some outcome measures, but the central problem in this field has always been thin clinical evidence [5, 10]. One systematic review states outright that the remineralizing advantage of BAG comes mainly from laboratory evidence, and studies showing long-term clinical effectiveness are still lacking. BAG performs well in many experimental models, yet methods differ widely between studies, so more high-quality clinical trials are needed.

The actual effect of BAG also depends on glass composition, particle size, ion release rate, and product form. Different formulations can show quite different mineralization and pH-response profiles, so BAG should not be treated as a single material. That tunability helps tailor the material to different uses, but it also makes it harder to compare results across studies, standardize products, and move them into the clinic.

Taken together, CPP-ACP, nHA, and BAG add to the mechanisms of conventional fluoride from three directions: calcium–phosphate stabilization and delivery, nano-mineral biomimicry, and ion release with control of the microenvironment. Even so, there is not enough high-quality evidence that any one of them can fully replace fluoride. A more sensible path forward may be synergistic combinations of materials with different mechanisms, not one material displacing another.

4. Polymer composite remineralization materials

As research has gone on, it has become clear that inorganic active materials used alone share several problems. Nanoparticles agglomerate easily, release of active ions is hard to control precisely, retention on the tooth surface is limited, and a single inorganic material often cannot meet the demands of remineralization, mechanical performance, and long-term stability at the same time. Work has therefore moved gradually toward organic–inorganic composite systems.

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4.1. Polymers as ion carriers, stabilizers, and biomimetic templates

In composite remineralizing materials, some functional polymers carry carboxyl, phosphate, or other groups that bind ions. These groups interact with mineral ions such as calcium and phosphate and so regulate their local concentration, stability, and release. Polymers can also stabilize amorphous mineral precursors, control where nucleation happens, or limit the space available for crystal growth, copying some functions of the organic matrix in natural mineralization [6].

Natural enamel formation itself involves complex regulation by organic molecules, which is why biomimetic remineralization research tries to build a similar microenvironment with polymers. Some polymers can stabilize precursors such as amorphous calcium phosphate and may help mineral diffuse into or deposit within demineralized tissue. Mature enamel, however, contains very little organic matter, and artificial polymers imitate only part of the regulatory role the organic matrix plays during enamel formation, not the whole process by which natural enamel forms.

4.2. Construction of polymer–inorganic composites

Combining polymers with ACP, nHA, or BAG may let the two kinds of material complement each other. The inorganic component supplies mineral or promotes mineralization; the polymer stabilizes particles, improves adhesion, and regulates release. The resulting composite may combine bioactivity with good material properties, but the two do not necessarily improve together. The actual result depends heavily on polymer type, inorganic filler content, interfacial bonding, and the preparation process [12].

This approach suits dental restorative materials especially well. A conventional resin that aims for high strength but has no mineral activity can still develop secondary caries around it, while adding large amounts of active inorganic particles can damage the mechanical integrity of the polymer network. Research on modern composites has therefore begun to look for a balance point: functional components that release calcium, phosphate, or other active ions over time without giving up much in mechanical properties.

By combining mineral activity, ion delivery, material stability, and interface control, polymer composites may offset some limits of single inorganic materials. Highly active fillers can still affect mechanical properties, and long-term stability and behavior at the interface in vivo remain uncertain. The molecular design of the organic–inorganic interface needs further work, and overall performance needs to be checked in long-term clinical studies.

5. Development trends and future perspectives of remineralization materials

Remineralization materials have been moving from inhibiting demineralization and topping up mineral ions toward imitating natural mineral formation. Early work concentrated on using fluoride to lower enamel solubility. Calcium–phosphate supply systems came next, followed by biomimetic materials such as nano-hydroxyapatite, bioactive glass, self-assembling peptides, and polymer templates [4, 7].

Natural enamel has a highly ordered hierarchical structure, so future research will have to go beyond raising mineral content and also deal with crystal orientation, spatial arrangement, and interfacial bonding of the mineral.

The real oral environment is shaped at once by biofilm, salivary flow, dietary acid challenges, temperature changes, and mechanical wear, and a single mineralization-promoting function is hard to sustain for long. Materials are therefore moving toward multifunctional designs that combine

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mineral ion release, adhesion to the tooth surface, controlled sustained release, biofilm regulation, and environmental responsiveness. Stimulus-responsive materials, for example, can stay stable near neutral pH and release more ions or show more antibacterial activity during an acid challenge.

Studies still differ markedly from one another, though, and long-term stability, biosafety, and clinical benefit all need to be verified.

Clinically, the best target for remineralization materials is the active non-cavitated lesion with an intact surface: early enamel caries, white spot lesions, and some post-orthodontic demineralization lesions. Once a cavity has formed, remineralization materials usually cannot stand in for the restorative treatment that is needed. Where the tissue can still repair itself, non-invasive or minimally invasive mineral regulation has the most to offer.

Future practice may therefore settle into a stratified system built on caries risk, lesion activity, and the level of evidence behind each material. Fluoride would stay the baseline measure for most patients, while calcium–phosphate supplying materials and other bioactive systems would be kept for specific indications or used in combination. High-quality clinical studies that directly compare stratified protocols and material combinations are still missing, so this model remains a direction awaiting validation.

In short, dental remineralization materials are developing from fluoride-only caries prevention into a multilayered set of materials that combine calcium–phosphate supply, nano-minerals, bioactive glass, polymer templates, and smart responsive functions. The goal has shifted from raising surface mineral content to remineralizing the lesion body, bonding at the interface, and restoring long-term function. A clear gap remains between laboratory results and real clinical performance.

6. Conclusion

Remineralization materials have moved on from fluoride alone to calcium–phosphate delivery systems, nano-biomimetic materials, bioactive glass, and polymer composites. Fluoride, even so, is still the best-supported basis for preventing caries and for managing non-cavitated early lesions. CPP-ACP, nano-hydroxyapatite, and bioactive glass each show promise in delivering calcium and phosphate, providing nucleation sites, and adjusting the local microenvironment, and polymer composites may improve particle stability, retention on the tooth surface, and ion release. For now, the newer materials work better alongside fluoride than in place of it. They are aimed at active early lesions with an intact surface and at white spot lesions after orthodontic treatment. Once a cavity has formed, restoration is still needed.

The evidence has clear weaknesses. Most studies are in vitro or short term, formulations are not standardized, outcome measures differ from study to study, risk of bias is a concern, and long-term clinical data are scarce. A rise in surface hardness or mineral content should not be read as full remineralization of the lesion body, or as a lower caries risk for the patient. Future work should standardize material characterization, agree on core outcome measures, model the oral environment more realistically, assess long-term biosafety, and run multicenter randomized controlled trials. Longer term, material choice could be matched to a patient's caries risk and lesion activity, and multifunctional composites that control ion release, regulate biofilm, and respond to the oral environment deserve further development.

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