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REVIEW ARTICLE

NATURAL NON FLUORIDE REMINERALIZING AGENTS: A REVIEW

Dr.Poorvika. S, [1*], Dr. Suma.G [2]

[1]Post Graduate, Pediatric and Preventive Dentistry, Vokkaligara Sangha Dental College and Hospital, Bangalore, India

[2]Professor, Pediatric and Preventive Dentistry, Vokkaligara Sangha Dental College and Hospital, Bangalore, India

[*]Author of Correspondence: Dr.Poorvika. S, Post Graduate, Pediatric and Preventive Dentistry, Vokkaligara Sangha Dental College and Hospital, Bangalore, India

Email: poorvikasuresh41@gmail.com

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ABSTRACT

Dental caries remains a prevalent biofilm-mediated disease characterized by an imbalance between demineralization and remineralization of dental hard tissues. Contemporary preventive dentistry emphasizes minimally invasive strategies aimed at arresting early, non-cavitated lesions through remineralization. While fluoride has been the cornerstone of caries prevention, concerns regarding over exposure and toxicity, particularly in pediatric populations, have led to growing interest in alternative approaches.

Natural, non-fluoride remineralizing agents have emerged as promising substitutes due to their biocompatibility, safety, and multifaceted therapeutic properties. These agents, include various plant-derived polyphenols, which exert their effects through diverse mechanisms such as enhancement of salivary function, provision of calcium and phosphate ions, inhibition of cariogenic bacteria, promotion of collagen cross-linking, and facilitation of hydroxyapatite crystal regeneration.

In addition, herbal agents like neem, aloe vera, and tea polyphenols contribute antimicrobial and antioxidant benefits, further supporting the remineralization process. Emerging biomimetic materials such as agarose also demonstrate potential in mimicking natural mineralization pathways.

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Introduction

Dental caries is a dynamic process that affects the hard tissues of the tooth. The progression of early, non-cavitated lesions depends on the delicate balance between demineralization and remineralization. Modern dentistry has shifted its focus from Black's principle of "extension for prevention" to a more conservative approach aimed at preventing lesion progression. Traditional oral hygiene methods often prove inadequate in managing dental decay. Consequently, contemporary clinical practices prioritize non-invasive strategies, such as remineralization, to manage non-cavitated lesions.

Remineralizing agents play a significant role in treating initial and early carious lesions. This natural repair process restores minerals in ionic form to the hydroxyapatite (HAP) crystal lattice. Under near-neutral pH conditions, calcium and phosphate ions from saliva are re-deposited within the carious lesion, facilitating mineral recovery [1].

This review article will focus on natural, non-fluoride remineralizing agents that have shown potential in promoting enamel repair and enhancing oral health.

Xylitol

Xylitol is believed to be a “tooth-friendly”, non-fermentable sugar alcohol containing a five-carbon sugar polyol and is naturally found in fruits, vegetables and berries.The main properties of this sweetener is that it is not fermented to acids, forms less of plaque and reduces the number of Mutans streptococci in saliva . Some researchers indicated that xylitol might be involved in the enamel demineralization and remineralizing process by acting as a calcium carrier required for remineralization. A novel method of delivering remineralizing ions (calcium and phosphate) in combination with xylitol has been developed using a NaF varnish (Embrace Varnish, Pulpdent).The xylitol coating prevents early reaction and produces a sustained release of the remineralizing ions[2].

Saliva exposure dissolves the xylitol and frees the calcium and phosphate ions. They then react with the fluoride in the varnish to form protective fluorapatite on the teeth. Xylitol-containing chewing gums could potentially have as a preventive measure and increases salivary flow rate, improves protective properties of saliva and enhancing remineralization [3].

Agarose

Agarose, a natural polysaccharide composed of repeating d-galactose and 3,6-anhydro l-galactose units, functions as an organic matrix template for biomimetic mineralization. It forms agarose fibrenanoscale-amorphous calcium phosphate complexes that act as precursors for mineralization.The agarose hydrogel serves as a reservoir for these mineral precursors, with its confined network ensuring uniform and controlled complex sizes. This leads to the aggregation of amorphous particles, resulting in the calcification of demineralized collagen fibrils and the occlusion of dentinal tubules [4].

Hesperidin

Hesperidin, a flavonoid glycoside found in citrus fruits, is esteemed for its antimicrobial and remineralizing qualities. Initially isolated from citrus peel by French chemist Lebreton, it's also present in bergamot, lemon, banana, and other plants like leeks. Hesperidin has anti-inflammatory, anti-microbial, antioxidant, and collagen cross-linking effects which limit the development of caries and enhance the remineralization process. Hesperidin has been added to dental adhesive in three different ratios producing four experimental adhesive groups (control, 0.2%, 0.5%, and 1%). Results showed that 0.5 wt% HPN incorporated dental adhesives could achieve a promising antibacterial effect without adversely affecting the adhesive characteristics. Structurally, it consists of aglycones such as hesperetin or methyl eriodictyol bound to rutinose. Its versatility extends to treating diabetes, cancer, cardiovascular disease, and skin health. Studies indicate hesperidin's efficacy in remineralizing both superficial and subsurface lesions, possibly through collagen protein interactions. When incorporated into dental adhesive at specific ratios, it exhibits promising antibacterial effects without compromising adhesive properties. In clinical trials, hesperidin has been used to induce remineralization and halt active carious dentin due to its solubility in water [5].

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Grape Seed Extract (Polyphenols)

Polyphenols possess antioxidant, antitumoral, antiinflammatory, and antibacterial effects. Proanthocyanidins, with high antioxidant capacity, are present in grape seed extract. One of the essential properties of proanthocyanidin is its ability to strengthen tissues containing collagen by increasing cross-linking of collagen. Some studies have reported that proanthocyanidin indirectly promotes hydroxyapatite growth by increasing exogenous collagen cross-links and inhibiting the glucosyltransferase enzymes of S. mutans, preventing caries [6].

In a study by Benjamin et al. grape seed extract was reported to be significantly effective in remineralization. Mirkarimi et al. noted in an in-vitro study on primary teeth that grape seed extract significantly increased the microhardness of teeth with initial enamel caries. A study by Nagi et al. found grape seed extract to be effective in remineralization, consistent with the findings of other studies [7].

Sorbitol

Sorbitol is referred to as a non-cariogenic sugar because it is fermented more slowly by cariogenic bacteria compared to sugars like glucose, sucrose, and fructose. Therefore, it causes less reduction in plaque pH. The caries-preventive effect of sorbitol has not been found to be as successful as xylitol when compared [8].

Isomalt

Isomalt is commonly found in the content of sugar-free candies and gums. Like xylitol, it cannot be fermented by cariogenic bacteria, making it antimicrobial and noncariogenic. It has the ability to bind calcium, thereby enhancing remineralization. Although isomalt's effect on caries is reported to be less successful than xylitol, it has been suggested that it may be more effective when used in combination with fluoride.

Chitosan

Chitin is an amino polysaccharide found in the cell walls of insects, the shells of fungi, and the exoskeletons of crustaceans. Chitosan, obtained by the deacetylation of chitin, is biocompatible and has been used in medicine since the 1960s in wound dressings, surgical sutures, and bandages. It later found applications in wound treatment, cholesterol medications, and contact lenses. Chitosan is preferred in dentistry due to its antifungal and antibacterial effects. It has been reported to elevate plaque pH by buffering the effects of acids produced by cariogenic bacteria, thus supporting remineralization [9].

In a study by Hayashi et al. individuals chewing gum containing chitosan reported a significant decrease in S. mutans counts in their saliva. In an invitro study examining the penetration of chitosan into enamel and its inhibition against demineralization, Arnaud et al.reported that chitosan intervened in the demineralization process by inhibiting phosphorus release, resulting in higher microhardness values for teeth treated with chitosan [10].

Aloe Vera

Aloe, the largest genus in the Xanthorrhoeaceae family, thrives primarily in arid climates across Africa, India, and other regions of the Old World. With approximately 140 species, Aloe plants boast a rich repository of phytochemicals, particularly concentrated in their leaves and gel. These compounds span a broad spectrum, including anthraquinones, anthrones, chromones, coumarins, pyrans, pyrones, alkaloids, benzene, naphthalene, and furan derivatives, flavonoids, sterols, tannins, polysaccharides, vitamins, and minerals [11].

This diverse phytochemical composition underlies the multifaceted biological activities observed in Aloe species. Traditionally, Aloe has been utilized for various health and medicinal purposes, prompting extensive scientific exploration into its pharmacological potential. Arginine, the predominant amino acid in Aloe vera gel, is joined by salicylic, uronic, and galacturonic acids, as well as sugars like fructose, mannose, and glucose. Enzymes such as oxidase, amylase, and catalase, alongside essential minerals like sodium, potassium, calcium, and magnesium, enrich its composition. Due to enamel's porous nature, Aloe vera dentifrices, comprising 35% gel, may foster remineralization by aiding arginine-calcium deposition. Acid exposure from bacterial metabolism can strip minerals from hydroxyapatite crystals, leading to increased surface porosity and opaque lesions, potentially reversible with remineralization [12].

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Gum Arabic

Gum arabic, derived from Acacia trees, has a long-standing history of versatile applications, particularly in confectionery and diverse industries. Recent research highlights its potential as an antimicrobial agent against oral pathogens, aiding in plaque and decay prevention. Gum arabic acts as a prebiotic, supporting gastrointestinal health and potentially benefiting conditions such as chronic renal failure and diabetes. Moreover, gum arabic contains essential minerals like calcium, magnesium, and potassium, which are known to promote tooth remineralization. Specifically, the calcium ions present in gum arabic can replenish Ca2+ ions that have been depleted from hydroxyapatite crystals, thereby preventing further demineralization of enamel.
Research by Onishi T et al. has shown that the concentration of insoluble Ca2+ and PO4_ ions in gum arabic aids in facilitating tooth remineralization. Additionally, gum arabic exhibits the ability to inhibit acid-dependent demineralization and maintain remineralization even in fluoride-free environments. Incorporating gum arabic into oral care products, such as toothpaste, shows promising results in reducing plaque, gingival inflammation & demineralization, thereby supporting oral health maintenance.With its diverse applications and multifaceted benefits, gum arabic emerges as a valuable natural resource for various health and industrial purposes, inviting further exploration and development in the realm of remineralization and oral care [13].

Licorice Root

The medicinal use of Glycyrrhiza glabra, or licorice root, dates back to ancient times. It possesses anti-inflammatory, antiviral, antiallergic, and antioxidant effects. In dentistry, it is preferred in pediatric toothpaste formulations. The presence of glycyrrhizic acid in licorice root has been found to inhibit the glucosyltransferase activity of S. mutans, preventing the synthesis of glucans in the biofilm. Due to this effect, it has been reported to have both caries-preventive and remineralization-promoting properties [14].

Theobromine

Theobromine, an alkaloid from the methylxanthine family, is found in high amounts in cocoa beans. While belonging to the same xanthine family as caffeine, their effects on teeth are different. It has been reported that caffeine increases the solubility of dental hard tissues, whereas theobromine reduces solubility. In an in-vitro study, Amaechi et al. reported that theobromine increased the remineralization potential of teeth comparably to fluoride .

Two types of substances are present in theobromine which provides cariostatic property: One which exhibits anti-bacterial activity and the other anti-glucosyltransferase.
According to Amaechi et al., theobromine and fluoride toothpaste has given a significantly higher mineral gain with relative to artificial saliva showing the theobromine [15].

Propolis

Propolis is a mixture containing resin produced by honey bees (Apis mellifera) from plant exudates to fill gaps in their hives. The active components of propolis, flavonoids, have antioxidant, antibacterial, antiviral, antifungal, and anti-inflammatory properties. It is effective against both Gram-positive and Gram-negative bacteria, particularly exhibiting strong efficacy against Staphylococcus aureus and Salmonella. The effectiveness of propolis in inhibiting the glucosyltransferase enzyme activity of S. mutans and Streptococcus sobrinus, both in vivo and in vitro, has been supported by studies.
Additionally, propolis has been reported to have antibacterial activity against some anaerobic oral pathogens. Zaleh et al. reported that propolis significantly increased the microhardness of enamel lesions in initial enamel caries, and some studies on propolis's remineralization effect have yielded similar positive results [16].

Ginger

Ginger (Zingiber officinale Roscoe, Zingiberacae) is a plant that has been used since ancient times worldwide. It is known for its anti-inflammatory, antibacterial, and nontoxic properties and has been approved for safety by the US Food and Drug Administration (FDA). It finds applications in various medical fields, including gastrointestinal diseases, cardiovascular diseases, joint diseases, cancer, and symptomatic relief (sore throat, nausea) in some viral diseases. Gingerol, found in ginger, imparts

antibacterial properties by causing the dissolution of bacterial cell membranes. Polyphenols in ginger, such as beta-carotene, ascorbic acid, flavonoids, and flavonols, provide antioxidant properties. Especially flavonoids and their derivatives, being lipophilic, disrupt bacterial cell membranes, exhibiting antibacterial effects. Inhibition efficacy against both

Gram-positive and Gram-negative bacteria has been observed. Ginger has also been reported to inhibit the growth of respiratory pathogens such as Haemophilus influenza, Staphylococcus aureus, Streptococcus pyogenes, and Streptococcus pneumoniae. Ginger has demonstrated inhibitory effects on oral flora, including S. mutans, Porphyromonas gingivalis, Prevotella intermedia, and Porphyromonas endodontalis.

In an in vitro study conducted by Hassan et al. the remineralization capacities of ginger, rosemary, and fluoride varnishes were examined, and all three materials were reported to be usable for remineralization, with the ginger group showing the highest remineralization efficacy. In another in-vitro study by Hossam, the remineralization activities of ginger, rosemary, and fluoride varnishes were investigated for initial enamel caries, and the ginger group exhibited the highest remineralization efficacy [17].

Neem

Neem has been extensively used in Ayurveda, Unani and Homoeopathic medicine and has become a wonder tree of modern medicine. It has been used traditionally for the treatment of inflammation, infections, fever, skin diseases and dental problems. Azadirachta indica is an evergreen tree having potential medicinal values. It has been found to be active against many dreadful disorders like hepatitis, viral infections, malaria and cancer. It is also effective against periodontal pathogens, and oral acidogenic bacteria responsible for dental caries and dental plaque

Nimbidin, a major active principle isolated from seed kernels of A. indica exhibits several biological actions. Neem dental care products contains Neem leaf or bark extract. Neem leaf is rich in antioxidants and helps to boost the immune response in gum and tissues of the mouth

Neem offers a good remedy for curing mouth ulcers, tooth decay and acts as a pain reliever in toothache problems [18].

Dental Applications of Neem

Antibacterial activity

Neem is a natural antibacterial agent. Various scientific studies have revealed its antibacterial activity. The antimicrobial effects of Neem have been reported against S. mutans and S. faecalis. Ethanolic extract of Neem leaves and sticks and bark exhibited significant antibacterial activity. Dried chewing sticks of Neem showed maximum antibacterial activity against S. mutans compared to other dental caries-causing organisms, S. salivarius, S. mitis, and S. sanguis.

Anti-candidial activity

Ethanolic and aqueous extract of Neem leaf showed significant anti-candidial effect against C. albicans. A clinical study demonstrated the effects of the leaf aqueous extract from Azadirachta indica (Neem) on adhesion, cell surface hydrophobicity and biofilm formation, which may affect the colonization by Candida albicans. The results suggest that Neem leaves have a potential anti-adhesive effect on the sample studied in vitro.

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Anti-cariogenic activity:

Mango and Neem extract showed antimicrobial activity against S. mutans, S. salivarius, S. sanguis and S. mitis. Antimicrobial activity of commercially available Himalaya herbal dental cream containing neem and fluoride-containing cheerio gel toothpaste has been assessed in school children. The study reported both the toothpastes showed a good antimicrobial effect on caries producing salivary streptococcus mutans. The toothpaste

containing Neem as well as fluoridated toothpaste were equally efficacious against caries-producing bacteria. Acetone extract from the bark of Neem is bactericidal against S. sobrinus hence indicates its anti-cariogenic activity [19].

Anti-plaque activity:
Aqueous extract of Neem stick and the gallotannin-enriched extract from Melaphis chinensis inhibited insoluble glucan synthesis and results in bacterial aggregation. It reduces the ability of streptococci to colonize tooth surfaces]

Mucoadhesive dental gel containing *Azadirachta indica* is found to be beneficial in reducing the plaque index and salivary bacterial count comparatively better than chlorhexidine gluconate mouthwash.

Efficacy of Neem extract against acidogenic oral

bacteria in fixed orthodontic appliance patients:

Ethanolic leaf extract of Azadirachta indica shows significant antibacterial activity against selected acidogenic oral bacteria causing dental plaque in fixed orthodontic appliance patients. The study conducted by us evaluated the anti-plaque activity of the extract against S. mutans, S. sanguis, and S. mitis. The extract did not inhibit L. acidophilus when tested.

Aqueous and ethanolic extract of Neem leaf inhibits S. mutans and E. faecalis which cause root canal failure in endodontic procedure. Its antioxidant and antimicrobial properties makes it a potential agent for root canal irrigation as an alternative to sodium hypochlorite.

Neem bark and leaf extract is most effectively used in preventing cavities and gum disease. Mouthwash containing Neem is a remedy for tooth decay, oral infections, prevents bleeding and sore gums. Twigs of Neem tree are used as chewing sticks by people all over India [20].

Oleum Limonis (Lemon Oil)

Lemon essential oil (LEO), extracted from lemon peel, is primarily composed of limonene (LIM). Research has shown that both LEO and LIM can inhibit cariogenic bacteria by preventing their growth, adhesion, acid production, and the formation of virulence factors such as insoluble glucans. Additionally, Ma et al. reported that essential oils like lemon oil can help stabilize tooth structure by protecting against collagen degradation and promoting remineralization in the early stages of dental caries. While LIM is the major active component, it remains important to determine whether LIM alone or other constituents of LEO contribute to these protective effects against early caries development [21].

Coconut

Coconut (Cocos nucifera), a staple in many households, is widely used in the food industry in various forms. Raw coconut milk, in particular, contains key minerals such as potassium, calcium, and phosphorus, with calcium being the most abundant. Oil pulling—a traditional oral hygiene practice in Indian culture—involves swishing edible oils like sesame, sunflower, or coconut oil in the mouth. Studies have shown that oil pulling may help reduce halitosis, tooth discoloration, and allergic reactions, while also inhibiting oral microbiota and preventing gingivitis.

Notably, research involving virgin coconut oil (VCO) for oil pulling demonstrated a reduction in Streptococcus mutans, a major contributor to tooth decay. These findings suggest that VCO, coconut milk (CM), and coconut water may exhibit similar enamel-remineralizing effects [22].

Galla Chinensis

It is an extract derived from a traditional Chinese plant. Its use as a caries preventive agent is under investigation. Chu et al. reported that this plant prevented demineralization and enhanced remineralization. In a study examining the mechanism of action of G. Chinensis, Zhang et al. stated that it slowed down demineralization on the outermost layer of carious lesions, allowing ion penetration towards the lesion body [19]. In in vitro studies, Huang et al. reported that using G. chinensis in combination with

nanohydroxyapatite significantly increased the remineralization of initial enamel lesions. In comparative studies by Abdel-Azem et al. it was reported that NaF and G. chinensis yielded similar results in terms of remineralization efficacy [23].

Tea Polyphenols

Green tea, extracted from the Camellia sinensis plant, enjoys global popularity, with variants like green, black,white and Oolong widely consumed. However, green tea is particularly renowned for its health benefits.Recent studies have emphasized its potential in cancer prevention, inflammation reduction, and brain health protection, largely due to its rich polyphenol content, notably epigallocatechin-3-gallate (EGCG) [24].

Tea leaves, containing polyphenols and fluoride, aid in combating dental caries. Catechins such as epicatechin (EC), epigallocatechin (EGC), epicatechingallate (ECG), and EGCG found in white and green teas act as MMP inhibitors. Furthermore, white tea exhibits antioxidant properties, counteracting oxidants. Both green and white teas have been observed to protect demineralized enamel by enhancing surface microhardness and reducing enamel wear, owing to their high catechin content, thus enhancing enamel surface quality [25].

Rosemary

Rosemary (Rosmarinus officinalis Lamiaceae) is known for its antioxidant, anticarcinogenic, anti-inflammatory, antifungal, and antibacterial properties. The polyphenols it contains, such as carnosic acid and rosmarinic acid, provide these characteristics. It has been reported to have an inhibitory effect on Gram-positive bacteria such as S. mutans. In a study by Al-Duboni et al. the effectiveness of rosemary extract in the remineralization of initial enamel caries was examined. The results of fluorescence and microhardness evaluations indicated the effectiveness of rosemary in remineralization. Bilgin et al. in their studies examining the remineralization capacities of various plant products, reported that a mixture of ginger-honey-rosemary increased the microhardness of initial enamel caries and was effective in remineralization [26].

Conclusion

In conclusion, non-fluoride remineralizing agents represent a significant advancement in modern dentistry, offering a safe, effective, and biologically sound approach to enamel remineralization. While fluoride remains an important tool in caries prevention, these natural alternative agents provide a compelling solution for individuals seeking fluoride-free options. Their ability to restore enamel integrity without the risks associated with fluoride overexposure makes them particularly valuable in pediatric dentistry, where safety is a primary concern. As research continues to refine and validate these alternatives, their integration into mainstream dental practice will likely play a crucial role in the future of caries management and preventive dentistry.

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