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In vitro fluoride release from fluorinated elastic denture material

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Elastic denture material has been routinely applied to replace missing teeth. Its physical and chemical properties, as well as its flexibility enable its application to cover concave abutment areas, reduce food impaction, and improve the retention of dentures. However, after long-term elastic prosthesis wear, the self-cleaning ability during the chewing process weakens, resulting in a drastically increased chance of local plaque deposition on the abutment and microecological imbalance, which can lead to caries and periodontal diseases. In this study, to effectively prevent and reduce the occurrence of caries around dental restorations such as dentures, different ratios of sodium monofluorophosphate (Na_2PO_3F or MPF) were added to the elastic denture material, and the time-dependent fluoride release process was investigated after soaking in artificial saliva at a temperature maintained at 37ºC. The elastic denture materials containing 0%, 3%, 5%, 10%, 15% and 20% (weight ratio) of MPF were soaked in100 ml and 200 ml of artificial saliva solutions at a constant temperature of 37ºC. The solutions were periodically replaced, and a 2 ml sample of each solution was tested just before replacement. The fluoride release rates and patterns were determined using the fluoride selective electrode method. All fluorinated elastic dentures released fluoride at different concentrations, and the higher the added fluoride content, the higher was the fluoride release. The elastic dentures with added MPF demonstrated an effective fluoride slow-release effect. The controlled release of fluoride can effectively prevent and reduce the occurrence of caries around dental restorations such as dentures.

Key words: Caries, Slow-releasing fluoride, Elastic denture material, Release rate

INTRODUCTION

Elastic denture material has been routinely applied to replace missing teeth. Its physical and chemical properties, as well as its flexibility, enable its application to cover concave abutment areas, reduce food impaction, and improve the retention of dentures. In addition, it has excellent biological compatibility and imposes no adverse stimuli on patients' oral mucosa, making it an ideal base material. However, after long-term elastic prosthesis wear, the self-cleaning ability during the chewing process weakens, resulting in a drastically increased chance of local plaque deposition on the abutment and microecological imbalance, which can lead to caries and periodontal diseases. Currently, there is very little literature on the effects of adding fluoride to elastic denture materials; therefore, in this study, different ratios of MPF were added to elastic denture material, and the time-dependent fluoride release process was investigated after soaking in artificial saliva at a temperature maintained at 37ºC. The findings of this paper can provide an experimental basis for further clinical application.

EXPERIMENTAL

Materials

Analytical methods

The elastic denture material used in this study was obtained from Changsha Aolun High Tech Co., Ltd; Na_2PO_3F (analytical grade) was obtained from Reagent No.1 Factory of Shanghai Chemical Reagent Co., Ltd; PF-2-01 fluoride selective electrode and saturated calomel electrode (Type 232) were obtained from Shanghai Yue Ci electronic technology Co., Ltd. Six types of elastic denture bases were made with 0%, 3%, 5%, 10%, 15%, and 20% (weight ratio) added Na_2PO_3F.

Analytical methods

The base samples were prepared with base wax in dimensions of 9mm × 7mm × 3mm weighing 1 g each. Five samples of each fluoride concentration were prepared, for a total of 30 samples that were divided into six ratio groups. The preparation process was the same as that of removable dentures. The casting wax was shaped in the same way as the samples, and the mold cavity was prepared accordingly with boxing wax. The MPF was dissolved in the self-cure monomer and then either mixed with the elastic denture material particles or sprayed on the surface of the particles. After the solvent volatilized, the MPF was evenly attached to the particles. Next, the conventional compression

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molding technique was performed. The samples were soaked in 100 ml and 200 ml artificial saliva solutions, respectively, maintained at a constant temperature of 37°C. The solutions were each replaced on day 1, 2, 3, 6, 9, and 16, and then every 7 days for a length of 65 days. Just before solution replacement on the specified days, 2 ml of solution from each sample was removed and measured using a fluoride ion selective electrode.

Fluoride standard curve

A volumetric pipette was used to transfer 0.05, 0.1, 0.2, 0.4, and 0.6 ml of fluoride standard solution (10 ppm), respectively, to a 50 ml volumetric flask. Then, 20 ml of TISABI solution was added, diluted with distilled water to 50 ml, shaken well and transferred to a 50 ml polyethylene beaker. The latter was placed on a magnetic stirrer with inserted fluoride electrode and saturated calomel electrode. After stirring for about 30 min the reading was stable and the result was recorded. Finally, the fluoride E(mv)-log C F-standard curve was drawn on semi-logarithmic coordinate paper.

Statistical analysis

The rates of fluoride release and the release patterns of the samples of elastic dental materials containing different added fluoride ratios were analyzed by direct chart comparison.

RESULTS AND DISCUSSION

For both the 100 ml and 200 ml artificial saliva solutions, the fluoride release of each fluoride-containing group on days 2 and 3 was not significantly different from that of the fluoride-free group; however, after day 3 the fluoride release rates of the fluoride-containing groups were significantly higher than that of the fluoride-free group.

For the tests in 100 ml artificial saliva solution, the fluoride release of the elastic denture material in the fluoride-containing groups varied significantly in the first 44 days, but stabilized after that. The sample with 20% fluoride had the highest fluoride release, with an average release of 22.87 μg/(g×d) and the highest value of 187.6 μg/(g×d). The next highest was the sample with 15% fluoride, with an average release of 17.16μg/(g×d); the 10% sample had an average fluoride release of 14.47 μg/(g×d). The samples with 5% and 3% fluoride had small amounts of release, with a value of 12.51 μg/(g×d) and 11.09 μg/(g×d), respectively. The release of the 5% and 10% fluoride groups was not significantly different from day 3 to day 23. The fluoride release of the 10% fluoride containing group was significantly lower than that of the 15% group in the first 30 days; however, the difference was not significant after day 30. The release of the 3% fluoride containing group was similar to that of the 5% and 10% fluoride groups from day 3 to day 9, but was significantly lower than all groups for the other time points.

For the tests in 200 ml artificial saliva solution, the fluoride release of the fluoride-containing groups varied significantly in the first 37 days and then stabilized between days 38 through 65. Similar to the results of the 100 ml artificial saliva solution test, the sample with 20% fluoride had the highest fluoride release, with an average value of 21.12μg/(g×d) and a highest value of 167.2 μg/(g×d), followed by the sample with 15% fluoride, which had an average fluoride release of 17.73 μg/(g×d).The 10% group had an average fluoride release of 15.73 μg/(g×d), and the 5% group had an average fluoride release of 13.95 μg/(g×d). The 3% group had the lowest average fluoride release of 12.08 μg/(g×d). The fluoride release of the 10% and 15% fluoride groups was not significantly different from day 2 to day 16, but after that the release of the latter group was significantly higher than of the former group. The fluoride release of the 3% fluoride group was similar to that of the other fluoride containing groups for the first three days, and was significantly lower after that. After day 37, the fluoride release of all groups stabilized.

The addition of fluoride to the denture material is thought to protect abutments from caries by increasing local fluoride concentrations. The fluoride released to the saliva forms fluoride ions, which bond to the calcium ions dissolved in the saliva from enamel to form calcium fluoride and then attach to the enamel plaque interface. Moreover, calcium fluoride can elevate the enamel pH by neutralizing acidic substances, and fluoride in its free state may also bind to calcium and phosphorus in the saliva, forming fluoro-apatite on the enamel surface and promoting enamel remineralization [1-4]. Studies have shown that fluoride released at low concentrations inhibits acid production by affecting the metabolism of bacteria; at high concentrations, the infiltration depth of fluoride into newly-demineralized enamel is significantly increased, which hardens enamel softened by caries and inhibits the continuation of demineralization [5-7]. Removable dentures have been widely applied clinically to replace missing teeth. The physical and chemical characteristics of the base material have a direct impact on the health of the close contacting abutment. By adding

fluoride to the denture material, fluoride ions can infiltrate the material surface or pass through internal pores to spread to the saliva and tooth surface [8], slowly releasing fluoride near the enamel and maintaining a low concentration of fluoride at the surrounding tissues to prevent caries. Due to the special structural features of the attachment of elastic dentures and the large extension range of the base, the self-cleaning ability of the abutment during the chewing process weakens after long-term wear, and the chance of dental plaque deposition increases, resulting in local microecological imbalance, which is a cause of dental caries and periodontal disease. Because the elastic denture material maintains long-term close contact with the abutment surface, it is necessary to add fluoride in the denture material to prevent caries.

MPF is a white powder that is water-soluble at room temperature (0-25ºC) and is internationally recognized as an excellent third-generation anti-caries agent. Moreover, it has tooth desensitization, water fluoridation, and antibacterial effects, and is widely used as a fluoride toothpaste additive, preservative, and fungicide [9,10]. Currently, it is mainly used as an anti-caries fluoride agent in toothpaste. The fluoride in MPF exists in a complex ion state (FPO_3) with a neutral pH; therefore, it is more stable than free fluoride. In addition, it is highly compatible with other materials. These advantages make it an ideal fluoride additive [11,12]. The results of this study demonstrated that the addition of MPF to the elastic denture material led to a slow-release of fluoride.

The fluoride release curves shown in Figures 1 and 2 illustrate how each group of fluorinated elastic denture samples reached a peak on the first day, declined sharply on days 2 and 3, and increased again on day 4.

In the 100 ml artificial saliva, the fluoride release increased irregularly in the first 44 days, and stabilized after that. In the 200 ml artificial saliva, the fluoride release increased irregularly in the first 37 days and stabilized after that. This may have been due to the fact that at the beginning of the soaking, the MPF on the surface of the fluoride-containing elastomeric denture material began to dissolve, which formed a release peak. Later, the release of the MPF deep inside the denture material slowed down because of limited diffusion channels. However, after further soaking, the material absorbed water and expanded in various degrees, forming many tiny gaps on the surface of the material, which resulted in an uneven release of deep MPF. At the end of the experiment, after soaking for a period of time (44 days in 100 ml solution and 37 days in 200 ml solution), the swelling rates of the material were basically uniform, which meant that the fluoride release was stable.

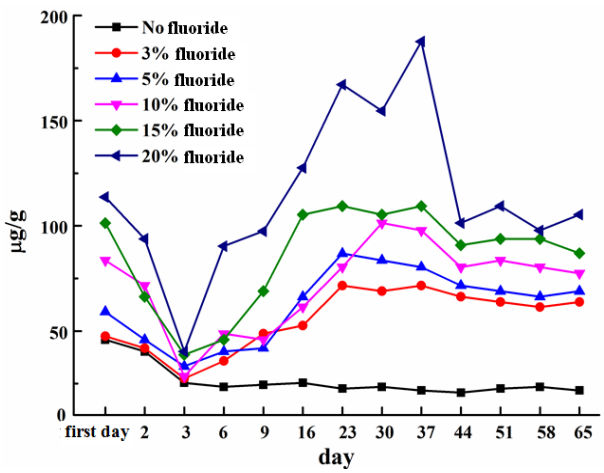

Fig. 1. Relationship between fluoride release (100 ml solution) and fluoride ratio in elastic denture material

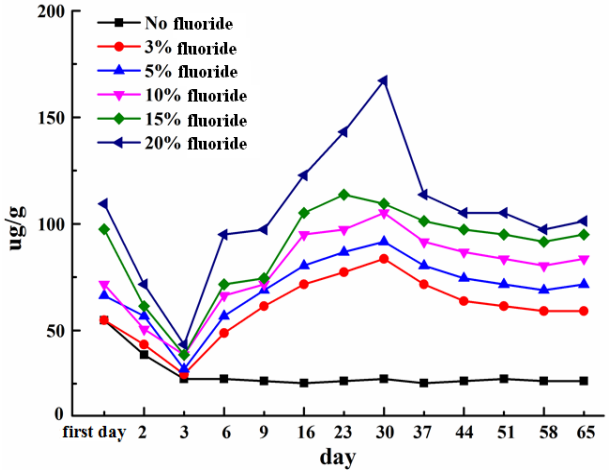

Fig. 2. Relationship between fluoride release (200 ml solution) and fluoride ratio in elastic denture material

Elastic denture material is an active polymer plastic composed of polycarbonate nylon composites. It has excellent flexibility, toughness, bending performance, corrosion resistance, and pressure forming ability. Moreover, it is resistant to high temperatures; it holds a stable volume without deformation at temperatures of 260-290ºC. In addition, it is non-toxic, tasteless, biologically safe, and inexpensive. It displays progressive osmosis and good water absorption. Although the fluoride itself has no slow-release capability, added *in gitto* to elastic denture material at low concentrations to form complexes in the form of “fluoride libraries”, it demonstrated sustained release action to the surface of the material. Foreign and domestic

studies have added fluoride to thermo-coagulation materials, self-curing resins, and a variety of filling materials in anti-caries experiments; however, there have been no experiments or tests performed with fluoride added to elastic dentures. For the first time, in this study fluoride was added to elastic denture material for anti-caries protection and improved the denture preparation process. For a more realistic simulation of the oral environment, to reduce interference factors, and increase the accuracy of the experiments, in this study samples were innovatively soaked in 100 ml and 200 ml of artificial saliva at a constant temperature of 37ºC and then fluoride release tests were performed. The scientific nature of the experiment was also enhanced by analyzing the release rates at different time points. As shown in the fluoride release curves, the higher the fluoride content of the elastic denture material, the higher are the fluoride release levels. The amount of fluoride in the material also affected the physical properties, color, and appearance of the material [13]. Therefore, the determination of the optimal ratio of fluoride requires more comprehensive consideration and should be further clinically validated. There is a wide range of oral fluoride doses that can cause acute poisoning; an oral dose of 6-9 mg/kg body weight can cause acute poisoning symptoms, and a one-time dose of 50mg/kg body weight is lethal [14-18]. The MPF doses added to the material in this study's experiments were much lower than the toxic dosage levels. Furthermore, exogenous fluoride is quickly diluted in saliva. Some of the fluoride in the saliva forms compounds and deposits in the enamel, some is deposited in the plaque, and some is excreted by oral self-cleaning. Most of the fluoride absorbed in the blood is excreted from the body with other body fluids. Clinically, it is highly safe.

CONCLUSION

The results of this study revealed that the addition of sodium monofluorophosphate into elastic denture material effectively released a certain concentration of fluoride when maintained at a constant temperature of 37ºC in artificial saliva over a long period of time. The controlled release of fluoride can effectively prevent and reduce the occurrence of caries around dental restorations such as dentures. These findings provide experimental and theoretical basis for further clinical research. However, due to limited experimental conditions and time, the fluoride release in this study was only observed for 65 days, and longer-term fluoride release experiments should be conducted for a more in-depth understanding.

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