

In vivo PIXE–PIGE study of enhanced retention of fluorine in tooth enamel after laser irradiation

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Available online 14 March 2008

Abstract

The presence of fluoride in tooth enamel reduces the solubility of hydroxylapatite by acid attack. Fluoride presence (even at low concentration) in the oral cavity is efficient against caries process. We propose a new approach of the explanation of the increase of fluoride retention in the tooth enamel when low power laser irradiation is applied after the treatment with fluoride gel (fluoridation). External beam PIGE measurements of fluorine on extracted teeth have been made in order to determine the best sequence of the operations. The laser irradiation after fluoride application is more efficient than the reverse procedure. This observation is in agreement with previous observations that the fluorine penetration in the enamel takes place first in the soft organic material present between the polycrystalline (prismatic) structure before being integrated in the crystalline composition of hydroxylapatite in order to produce fluoro-apatite. As those in vitro measurements do not reflect the whole process in the saliva, in vivo PIGE measurements have been also performed. We have demonstrated, by repeating the PIGE measurements (at least five times at various time intervals) that a significant increase of the fluoride retention took place even 18 months after the unique laser treatment. The complete experimental procedure is described: fluoride application, laser irradiation, PIGE measurements with 2.7 MeV protons (repeated measurements at the same place on the same tooth in order to follow the evolution) and safety tests before in vivo analyses.

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PACS: 25.40.Ep; 29.20.Ba; 29.40.Wk; 42.62.Be; 42.55.Lt; 61.80.Ed; 82.39.Wg

Keywords: Tooth enamel; External PIGE; In vivo analysis; Argon laser irradiation; Long term fluoride retention

1. Introduction

The presence of fluoride in tooth enamel reduces the solubility of hydroxylapatite by acid attack [1]. Fluorine may be introduced in the crystal lattice of hydroxylapatite during the enamel genesis or enamel maturation process. Oral fluoride administration during childhood seems to be less efficient against caries than the topical application of fluoride gel, toothpaste or mouth rinse solution [2]. Direct application by the patient may give rise, if applied in excess, to several diseases (fluorosis, poisoning, pruritus, nausea, vomiting, diarrhoeas and abdominal pain [3].

Topical application of the gel is ideally done in dental clinics. Practitioners avoid swallowing of fluoride during fluoridation by isolation and suction.

The presence of fluoride (even at low concentration) in the oral cavity is efficient against caries process. We have already demonstrated that the process of migration of fluorine in the tooth enamel takes place in two steps [4]. The first phase of the migration mechanism is a fast penetration of fluorine from the applied gel into the natural clefts existing between the micro-crystals (prisms) of hydroxylapatite which are mainly filled with soft organic material (Fig. 1). The second step is the migration of fluorine into the crystalline structure of enamel with the formation of fluoro-apatite. For an efficient protection against caries, a daily use of a fluoride paste is recommended because

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Fig. 1. Electron micrograph of tooth enamel. Horizontal arrows: soft regions between micro-crystals of hydroxylapatite where the first fluorine diffusion (fast procedure) takes place. Vertical arrows: crystals of enamel (hard region) where the second step (slow process of fluorine diffusion and transformation of hydroxylapatite into fluoro-apatite).

topical application by a dentist has a limited efficiency: about 5 weeks [4]. The fluoride deposited on the enamel surface resulting from fluoride topical application may be seen as a reservoir releasing fluoride [5–7]. This reservoir of CaF_2 releases fluoride during about 5 days [8].

We propose a new approach of the explanation of the increase of fluoride retention in the tooth enamel when low power laser irradiation is applied after the treatment with a fluoride reach gel.

2. Lasers in dentistry

Laser or more conventional light sources are of common use today in dental clinics. The argon laser is one promising source, as the wavelength of light emitted by this laser is optimal for the initiation of polymerization of composite resins [9]. Four main types of lasers are in use for dental research and practice [10]. Carbon dioxide (CO_2) lasers use CO_2 gas and are probably the most used in dentistry, namely in gingivectomies, biopsies and for the removal of benign and malignant lesions [11]. For lesions extending into tissue deeper than the 0.1 mm that the CO_2 laser penetrates, an Nd:YAG laser is appropriate.

Argon lasers (wavelengths of an argon laser at 514 nm “green” and 488 nm “blue” [12] with a power not exceeding 350 mW do not require tap water cooling, which adds great. Upon exposure to laser radiation, enamel powder mixed with NaF underwent an increase in crystallite size and/or perfection with a significant uptake of fluoride. Incisor teeth lased in the presence of NaF released significantly less calcium and phosphorus into sodium acetate (pH 4.0) compared with unlased controls, suggesting a possible role for the laser in caries prevention [13]. Argon laser has been used in the present investigation to demonstrate the enhanced retention of fluorine in tooth enamel. The

argon laser effects are inconsistent depending on whether the enamel surface is cleaned, but after cleaning, the superficial and deep increases of temperature are low: a few centigrades ($3^\circ\text{--}7^\circ$) [14]. With the CO_2 laser, the enamel and dentin surfaces reach very high temperatures, but only low temperatures are measured in the pulp chamber [15].

3. Laser enhanced retention of fluoride in enamel

Before experimenting the role of the laser irradiation in vivo, a preparatory protocol was established by in vitro study. Two-hundred and forty samples of enamel ($3\text{ mm} \times 3\text{ mm}$) were sampled from 120 recently extracted teeth (one sample from the face and one from the rear of each tooth). The “patients” belong to a large variety of ages. The original concentration of fluorine determined by PIGE was observed to be higher for young people than for older. No difference related to the face of sampling was observed. The fluorine concentration was measured before any treatment to obtain a reference value. These initial concentrations were very widely spread: from 100 to 5000 ppm of F. We will speak about the E group (enamel group) when referring to the results on this fresh material. The mean concentration of F calculated on the whole collection of samples is 3009 ppm.

The experimental procedure of PIXE–PIGE analysis was extensively described long time ago [16]. The full description: incident proton energy on the target: 2 MeV, beam extracted through a $8\text{ }\mu\text{m}$ thick tantalum foil, beam intensity 30 nA, detection of gamma-rays with a $\text{Ge}(\text{Li})$ of 60 cm^3 , 110 and 198 keV for F, 135 keV for Ta, 440 keV for Na and 1267 keV for P and detection of X-rays with a $\text{Si}(\text{Li})$ for Ca and heavier elements. The analyzed depth is about $20\text{ }\mu\text{m}$. As the fluorine concentration may vary from point to point on the same sample, the experimental set up was designed in order to reproduce the same beam impact at each step of the experimentation. The monitoring of the measurements is based on the detection of the 135 keV γ -ray line of tantalum.

After the measurement of the initial fluorine content, all of the samples were treated with acidulated phosphate fluoride (1.23% w/w fluoride ion; pH 3.5) by topical application for 5 min followed by a 1 min rinsing in distilled water. The whole collection of enamel samples was then divided in two parts of 120 samples having each the same distribution of initial fluorine concentration. From the 240 samples, 120 were irradiated with an argon laser delivering an energy of 10 J/cm^2 for 30 s (the EFL – enamel fluoride lased group) but no additional treatment was applied to the 120 other ones (the EF – enamel fluoride group). EF and EFL samples were immediately analyzed by PIXE–PIXE after the single (fluoridation only) or the double (fluoridation immediately followed by laser irradiation) treatment. This irradiation was performed with a maximum delay of 10 min. PIGE analysis gives the information on the increase of the fluorine content relative to the initial concentration. The control of any other modifi-

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