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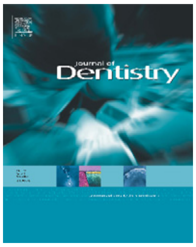

Niobium pentoxide as a novel filler for dental adhesive resin

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ABSTRACT

Objectives: The purpose of this study was to develop an adhesive resin with incorporation of niobium pentoxide and evaluate its properties.

Methods: Niobium pentoxide was characterised by X-ray diffraction, surface area, particle size, micro-Raman, scanning electron microscopy and the effectiveness of silanisation process by Fourier Transform Infrared (FTIR). An experimental adhesive resin was formulated with 0, 5, 10 and 20 wt% Nb_2O_5. The formulated adhesive resins were evaluated based on microhardness, degree of conversion, radiopacity and interface (resin/dentine) characterisation by micro-Raman.

Results: The particles used in this study presented a monoclinic crystalline phase with typical chemical groups and micrometre mean size. Microhardness and radiopacity increased with higher amounts of Nb_2O_5, and the particles were able to penetrate into the hybrid layers.

Conclusions: Therefore, Nb_2O_5 may be an alternative for polymer-based biomaterials.

Clinical significance: Niobium pentoxide could be used to produce adhesive resins with enhanced properties.

1. Introduction

In dentistry, the effectiveness of adhesion of resin to a tooth is directly related to several factors like demineralisation of substrate, resin infiltration and polymer setting.[1] This adhesion is vital for the success of long-term resin composites used to restore tooth cavities.[1] Poor adhesion to tooth substrates may produce postoperative hypersensitivity, marginal discoloration and less retention of restoration.[2–4] The following efforts have been made to increase bond strength to dental tissues: reducing the hydrophilicity of polymer,[5,6] adding fillers,[7] controlling the enzymatic activity[8–11] and changing other factors.[1]

Attempts to incorporate filler particles into adhesive resin to improve the resin/tooth bond strength were made in several previous studies and commercial products. Colloidal silica, hydroxyapatite, ytterbium trifluoride, tantalum oxide, glass and zirconia are among the filler particles that have been tested.[5,7,12–16] A hybrid layer that is less prone to degradation could be created by incorporating filler in adhesive resin,[17] thereby decreasing the water sorption and increasing the material properties. The reliable bonding of dental materials to tooth substrates depends on the mechanical and chemical

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features of the polymer. Therefore, improving the material's properties will lead to a more durable restorative treatment.[18]

Niobium is a transition metal and has the atomic number 41. This metal is widely used to enhance mechanical properties in the development of metal alloys.[19] Its application in the biomedical area was started recently and is due to the titanium and steel alloys that are used in the biomedical field.[20,21] Niobium pentoxide (Nb_2O_5) has shown bioactive properties, like hydroxyapatite crystal growth when in contact with human saliva[22] and has been used as an anti-allergic coating in endoprostheses with favourable results.[23] Therefore, it appears to be an alternative for composite development.

However, to the best of our knowledge, there are no reports on the use of niobium pentoxide in resin matrix production for biomedical use. The purpose of this study was to develop an adhesive resin, with niobium pentoxide, and evaluate the properties for using to restore tooth cavities.

2. Materials and methods

The monomers used in this study were bisphenol A glycol dimethacrylate (BisGMA), triethylene glycol dimethacrylate (TEGDMA), 2-hydroxyethyl methacrylate (HEMA), camphorquinone (CQ) and ethyl 4-dimethylaminobenzoate (EDAB) and were provided by Esstech Inc., USA. These materials were used without further processing. Niobium pentoxide (Nb_2O_5) was provided by Companhia Brasileira de Metalurgia e Mineração (CBMM, Araxá, MG, Brazil). The organic phase of the adhesive was produced by mixing 50 wt% Bis-GMA, 25 wt% TEGDMA and 25 wt% HEMA. CQ and EDAB were added at 1 mol% to all groups, according to the monomer moles. Nb_2O_5 was added at the following four different concentrations: 0, 5, 10 and 20 wt%. No radical scavenger was added. To improve the adhesion interface between filler particles and the matrix, Nb_2O_5 was subjected to a silanisation process with 5% of silane (γ-methacryloxypropyltrimethoxysilane, Aldrich Chemical Co., Milwaukee, WI, USA) and 95% of solvent (acetone), in weight.[24] After the silanisation process, the particles were stored for 24 h at 37 °C to allow the solvent to evaporate. All components were weighed using an analytical balance (AUW220D, Shimadzu, Japan), mixed and ultrasonicated for 1 h. To perform monomer photo-activation, a light-emitting diode unit (Radii Cal, SDI Ltd., Australia) was used. An irradiation value of 1200 mW/cm^2 was confirmed with a digital power meter (Ophir Optronics, USA).

2.1. *Characterisation of Nb_2O_5*

2.1.1. *X-ray diffraction*

To identify the crystalline phases in the powder, a Philips diffractometer (X'Pert MPD model) operating at 40 kV and 40 mA, using CuKα radiation was used. The scanning rate was 0.05°/min, and the time-step was 1 s. Analyses were performed within a 5–60° range.

2.1.2. *Micro-Raman spectroscopy*

The typical chemical groups of Nb_2O_5 compounds were identified by micro-Raman spectroscopy using Bruker Optics (SENTERRA model) equipment. The range of the analysis was 80–2700 cm^{-1}.

2.1.3. *Scanning electron microscopy*

Scanning electron microscopy (SEM) was used to evaluate the morphology of the Nb_2O_5 powder using Hitachi (TM3000 model) equipment.

2.1.4. *Surface area and particle size*

Using a Quantachrome NOVA1000 Autosorb Automated Gas Sorption System (Boynton Beach, FL, USA), the specific surface area of the Nb_2O_5 powder was determined through the Brunauer–Emmett–Teller (BET) method. Before the analysis, the sample was outgassed for 3 h at 300 °C in vacuum. The particle size distribution was assessed using a laser diffraction particle size analyser (CILAS 1180, Orleans, France).

2.1.5. *Evaluation of the silanisation process*

Fourier transform infrared (FTIR) spectroscopy was used to evaluate the attachment of silane to the Nb_2O_5 surface. The FTIR measurements were performed in the Vertex 70 FTIR spectrophotometer (Bruker Optics, Ettlingen, Germany). Nb_2O_5 powder was dispensed over a diamond crystal of Attenuate Total Reflectance (ATR) accessory. A total of 16 scans were collected from 400 cm^{-1} to 4000 cm^{-1} at a 4 cm^{-1} resolution. The C=C signal at 1636 cm^{-1} in the FTIR spectrum of Nb_2O_5 was used to verify the existence of the double bond following the surface treatment.

2.2. *Refractive index*

The refractive index values of co-monomer blend, before and after polymerisation, were evaluated by spectral ellipsometry. The sample was analysed using an ellipsometer SOPRA GES-5E (SEMILAB-SOPRALAB, Courbevoie, France) adjusted for wavelength range of 0.30–0.75 μm, at $\theta = 68°$.[25,26]

2.3. *Radiopacity*

The radiopacity of model adhesive resins was evaluated according to ISO 4049[27] standards. Five specimens per group ($n = 5$), 10.0 mm (±0.5 mm) in diameter and 1.0 mm (±0.1 mm) in thickness, were produced. X-ray images were obtained with the phosphorous plates Digital System (VistaScan, Dürr Dental GmbH & Co. KG, Bietigheim-Bissingen, Germany) using an exposure time of 0.4 s and a focus-film distance of 400 mm. The X-ray source (DabiAtlante model Spectro 70X) operated with a tungsten anode at 70 kV and 8 mA. Each of the five films contained one specimen of each of the four experimental groups. An aluminium step-wedge was exposed with the specimens in all images. The aluminium step-wedge thickness ranged from 0.5 mm to 5.0 mm in increments of 0.5 mm. The images were saved in TIFF format for less compressed files. Digital images were handled with Photoshop software (Adobe Systems Incorporated, CA, USA). The mean and standard deviation values of the grey levels (density of pixels) of the aluminium step-wedge, and the specimens were obtained in a standardised area.