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Metal-composite adhesion based on diazonium chemistry

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ABSTRACT

Objective. Composite resins do not adhere well to dental alloys. This weak bond can result in failure at the composite-metal interface in fixed dental prostheses and orthodontic brackets. The aim of this study was to develop a new adhesive, based on diazonium chemistry, to facilitate chemical bonding between dental alloys and composite resin.

Methods. Samples of two types of dental alloys, stainless steel and cobalt chromium were primed with a diazonium layer in order to create a surface coating favorable for composite adhesion. Untreated metal samples served as controls. The surface chemical composition of the treated and untreated samples was analyzed by X-ray photoelectron spectroscopy (XPS) and the tensile strength of the bond with composite resin was measured. The diazonium adhesive was also tested for shear bond strength between stainless steel orthodontic brackets and teeth.

Results. XPS confirmed the presence of a diazonium coating on the treated metals. The coating significantly increased the tensile and shear bond strengths by three and four folds respectively between the treated alloys and composite resin. Conclusion: diazonium chemistry can be used to develop composite adhesives for dental alloys.

Significance. Diazonium adhesion can effectively achieve a strong chemical bond between dental alloys and composite resin. This technology can be used for composite repair of fractured crowns, for crown cementation with resin based cements, and for bracket bonding.

1. Introduction

Composite resins are widely used in modern dental practice due to their ability to bind to acid etched tooth structure, and their good handling, mechanical, and esthetic properties. Composites are used in a variety of dental applications such as temporary and permanent restorations, cementation and repair of indirect restorations as well as bonding of orthodontic brackets [1,2]. However, the weak adherence of dental composites to dental alloys frequently leads to clinical problems such as failures at the resin-metal interface in fixed dental prostheses and de-bonding of orthodontic bracket [1,3,4].

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Porcelain fused to alloys have been used for many years for the fabrication of esthetic fixed prostheses. These prostheses present good clinical performance with 88.7% survival rates over ten years for metal-ceramic crowns and 80.2% for fixed partial dentures [5]. The most common cause for replacement of these restorations is dental caries followed by delamination of veneering porcelain [6]. Over time delamination of veneering porcelain can occur in 3–13% of the restorations [7–10]. This is usually caused by technical mistakes during their fabrication as well as difference between metals and ceramics in terms of modulus of elasticity, and coefficient of thermal expansion, moisture in the oral cavity, and mechanical fatigue of the ceramic itself [2,11]. The life of defective fixed prostheses could be prolonged by repairing the porcelain veneer with resin based composites [12], using surface conditioning methods and coupling agents to bond the resin composites to the metal substrates [13]. Surface conditioning can be done with hydrofluoric acid etching and air particle abrasion whereas the coupling agents are usually based on silane adhesives [2,10,12,14] alone or in combination with acidic or sulfur-containing adhesive monomers [15,16]. However, these procedures involve a number of drawbacks such as the hazardous intraoral use of hydrofluoric acid and air born abrasion, as well the special equipment needed for silica coating and the limited long-term performance [2,6,10,17–19].

In orthodontic treatments, metal brackets are usually made of stainless steel, and are bonded to teeth using commercially available resin based dental adhesives [20]. These bonding systems adhere well to etched tooth enamel but very poorly to metallic brackets [4,21–24]. Typically, bracket bases are designed with surface patterns that favor micromechanical interlocking to create mechanical retention at the resin-base interface [22,25]. However, mechanical failure often occurs at the bracket-composite interface resulting in bracket de-bonding [23]. Bracket de-bonding prolongs treatment duration and chair time, increasing treatment cost and causing inconvenience to the patient and the dentist [20,24]. To compensate for the lack of adhesion between metallic brackets and composite [4,21,22], the majority of research has been focused on modifications to the bracket base surface pattern in order to increase micromechanical interlocking, which has only achieved minor improvements [4,22,26].

A series of metal adhesives have been explored to improve the shear bond strength between brackets and the tooth or ceramic crown and between composite repairs and porcelain fused to metal (PFM) crowns, this include among others silane coupling agents, and bifunctional acidic monomers such as 10-methacryloxy ethyl diphosphate monomer (MDP) [16,27].

Silane coupling agents have been used to chemically bond resin composites to conditioned dental alloys, and ceramics. The silanol groups in silane coupling agents react with the OH group present on the oxidized metal surface, consequently bonding is to metal oxides and not to the metal itself [28]. Silane coupling agents have been improved over the years in an attempt to optimize their properties while rendering them durable and less clinically demanding. Nowadays silanes are supplied in a pre-hydrolyzed form in ethanol/water solutions to facilitate their clinical use [19]. However, Silane undergoes hydrolysis and this limits its shelf life and makes its long term efficiency doubtful [10], as its bond strength is sensitive to humidity, and decreases over time [29].

HEMA (2-hydroxy ethyl methacrylate) and adhesive acidic monomers that have a carboxylic group such as hydrolyzed 4-META(4-methacryloxyethyl trimellitic anhydride) or the phosphonic acid group [R–P(=O)$(OH)_2$] with or without sulfur groups, are also capable of bonding to non-precious metal alloys [30,31] resulting in a strong and durable bonding to various substrate materials. These monomers can also interact chemically with the metal oxide in base metal alloys [2,15,30], and they have superior hydrolytic stability, but they exhibit poor adhesion to precious metals and alloys and they require surface preparation of the substrate prior to their application [15].

Given the above-mentioned limitations of current bonding agents there is a need for the development of chemical adhesives that could increase the metal-composite bond strength and eliminate the difficulties encountered with the existing bonding strategies [2,32].

Aryldiazonium salts have been the subject of much recent research due to their ability to react with a wide range of surfaces including metals [33]. Aryldiazonium salts are easy to prepare, and are rapidly reduced to stable radicals that allow strong covalent bonding [33,34]. Diazonium ions can be produced from aromatic amines and grafted onto almost any surface using chemical reducing agents in acidic solutions [34]. The reducing agents activate the aryldiazonium salts and form aryl radicals that covalently bind to the surface of interest [35]. If an extra amino group is present on the aryldiazonium precursor, this group can be further activated and used to bind a second layer onto the original surface [35–37]. The chemical bond formed between aryl diazonium salts and the different substrates is stable at temperatures as high as 200 °C, which is much higher than the maximum temperature reached in the oral cavity [38]. It is very resistant to solvents and ultrasonication [28,34,39], and is electrochemically stable. Mechanically, the adhesive is also stable, it can withstand scratching, and just as other coupling agents, it is also capable of improving the bond strength between substrates [34,37].

The objective of this study was to develop a new surface treatment, based on diazonium chemistry, to facilitate chemical bonding between dental alloys and composite resins. This new simple and inexpensive bonding method would allow easy intraoral repair of fractured porcelain- fused-to-metal prosthesis and prevent de-bonding of orthodontic brackets.

2. Materials and methods

2.1. Materials

2.1.1. Treatment materials

P-Phenylenediamine (PPD), sodium nitrite ($NaNO_2$), sodium dodecyl sulfate (SDS), benzoyl peroxide (BP), bisphenol A-glycidyl methacrylate (Bis-GMA), hypophosphorous acid (H_3PO_2), and hydrochloric acid (HCl) were purchased from Sigma Aldrich (St. Louis, MO) and were used without any further purification. Concentrated hydrochloric acid (HCl) was diluted in distilled water to a concentration of 0.5 M.