

# Preparation of dental polymeric nano-adhesives with silica nano-porous (MCM-41) as novel fillers for improving the adhesive mechanical properties: Synthesis and application

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## Abstract

This study evaluates that the incorporation of silica nano-porous (MCM-41), as filler synthesized by sol-gel method, into an experimental dental polymeric adhesive improves the mechanical properties of the polymeric adhesive layer. MCM-41 filler was prepared by sol-gel method. Adhesive precursor with different wt. % of MCM-41 fillers (0-5.00 %) were mixed. The MCM-41 powder was characterized using FTIR, SEM, BET. The flexural modulus of seven adhesive systems containing different amount of MCM-41 were evaluated by three-point bending test. The results show that the mechanical properties of the new adhesive system improve when the amount of MCM-41 increases.

**Keywords:** dental polymeric adhesive, silica nano-porous filler (MCM-41), mechanical properties, flexural modulus.

## 1. Introduction

The primary aim of dental adhesives is to provide retention to composite fillings or composite cements. In addition to withstanding mechanical forces, and in particular shrinkage stress from the lining composite, a good adhesive also should be able to prevent leakage along the restoration's margins [1, 2]. With the recent improvements in dental adhesive, the behavioral characteristics of adhesive dentistry have received an increasing consideration [3, 4]. Nanotechnology created a new vision in the formation and evaluation of modified materials. The mechanical efficiency of nano-filler may in some situation be extraordinary to other filler size [5, 6]. The three-point bending test has been performed in the past by some researchers, for investigating the mechanical properties of dental adhesives. In these researches, the influence of inorganic filler particles of adhesive and composites on flexural strength and flexural modulus increased by using inorganic fillers [7, 8].

The higher elastic modulus observed in the filled adhesives, the powerful interaction between inorganic silica fillers and matrix enhanced by the silane coupling agent, resulting in reduction of the surface energy reported in the some literature [9, 10].

## 2. Materials and Methods

### 2.1. Synthesis of MCM-41

MCM-41 filler was prepared by mixing tetraethyl orthosilicate (TEOS, Merck company, Germany), distilled water and diethylamine (Merck company, Germany), at room temperature. Then, the pH was set at 8 which afforded the sol formation. Cetyltrimethylammonium bromide (CTAB, Merck company, Germany) was added to the sol and the clear solution was formed (gel formation). The gel was placed in an oven under vacuum to form MCM-41 filler.

## 2.2. Dental polymeric adhesive preparation

Adhesive precursor (2,2-Bis[p-(2-hydroxy-3-methacryloxypropoxy)-phenyl] propane (Bis-GMA, Degussa Group, Germany), 1,6-bis-[2-methacryloyloxyethoxycarbonylamino]-2,4,4-trimethylhexane (UDMA, Degussa Group, Germany), 2-hydroxyethyl methacrylate (HEMA, Merck company, Germany), 2-ethyl-2-(hydroxymethyl)-1,3-propandiol trimethacrylate (TMPTMA, Merck company, Germany) and ethanol (Merck company, Germany)) with different wt. % of MCM-41 filler (0-5.00 %) were mixed.

Then camphorquinone (CQ, Merck company, Germany) and N,N-Dimethyl aminoethyl methacrylate (DMAEMA, Fluka company, Germany) as photo-initiators were added to the above mixture. The mixture was placed under vacuum at suitable temperatures. The obtained dental adhesives were provided for mechanical testing.

## 3. Results and Discussion

FT-IR spectrum (Shimadzu FT-IR 8400S, Japanese) of the silica nano-porous filler is shown in Fig.1, which shows the peaks corresponding to the silica nano-porous filler structure.

The peaks characteristic of Si-O-Si appear at about 462, 794 and 1076  $\text{cm}^{-1}$ . Also, the peaks of Si-OH are located at 3100-3700  $\text{cm}^{-1}$  (Fig.1).

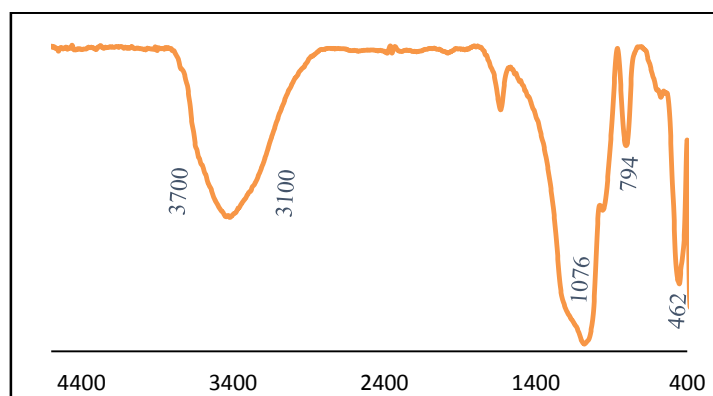


Fig.1: FT-IR spectrum of silica nano-porous filler

BET-plot (Adsorption/Desorption Isotherm) of silica nano-porous, which was reported by BET apparatus (Micrometrics AS AP 2020, Korea), shows that the filler is placed in the nano-porous categories and BJH-plot illustrates that this filler has a pore-size diameter of 3.37 nm (Fig.3).

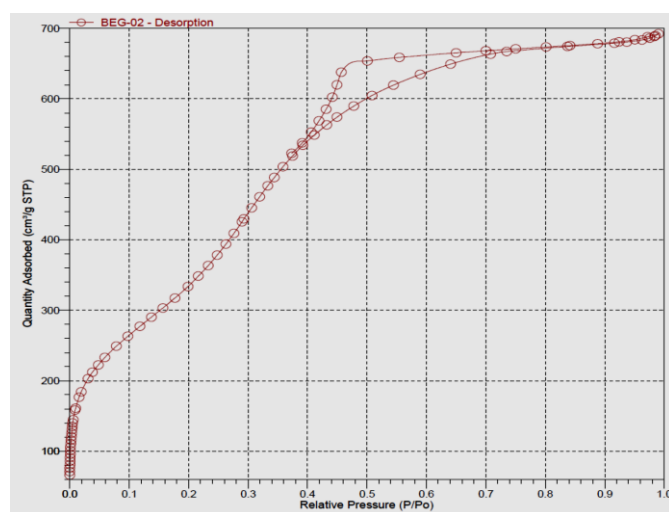


Fig.2: BET-plot of silica nano-porous filler

SEM image (MIRA FE-SEM IROST, USA) shows that the size of the silica filler is about 35.02 nm (Fig.3).

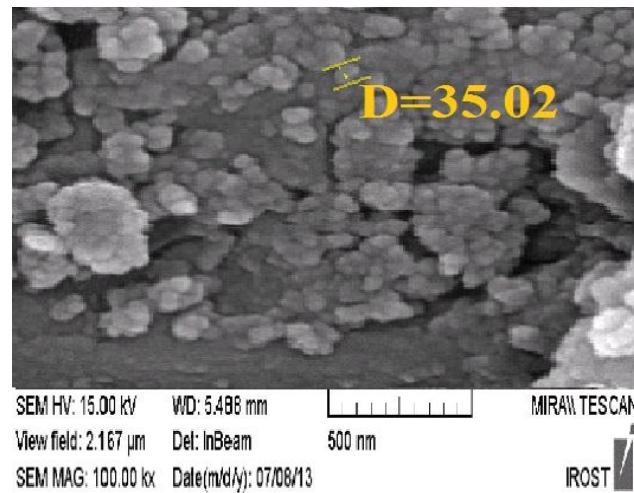


Fig.3: SEM image of silica nano-porous filler

The flexural strength of the unfilled and silica nano-porous filled adhesives was measured according to the ISO 4049(2000) standard method. The specimens were prepared in rectangular mold with dimensions of 2mm×2mm×25mm utilizing the same light-curing unit. After 1 day of storage in deionized water, a three-point bending test was performed using the universal testing machine at a cross-head speed of 0.5 mm/min.

The flexural strength and modulus were calculated based on the following equations:

$$\text{Flexural strength} = 3PL/2bd^2 \quad (1)$$

$$\text{Elastic modulus} = L^3.m/4bd^3 \quad (2)$$

where P is the maximum force applied, L is the span length, b is the width, d is the thickness and m is the slope of initial straight line deflection curve [9].

The results showed that the flexural modulus was increased for the sample containing 0–1.00 wt.% silica nano-porous filler to the maximum value of 644 MPa and then decreased with increasing the filler content (Fig.4).

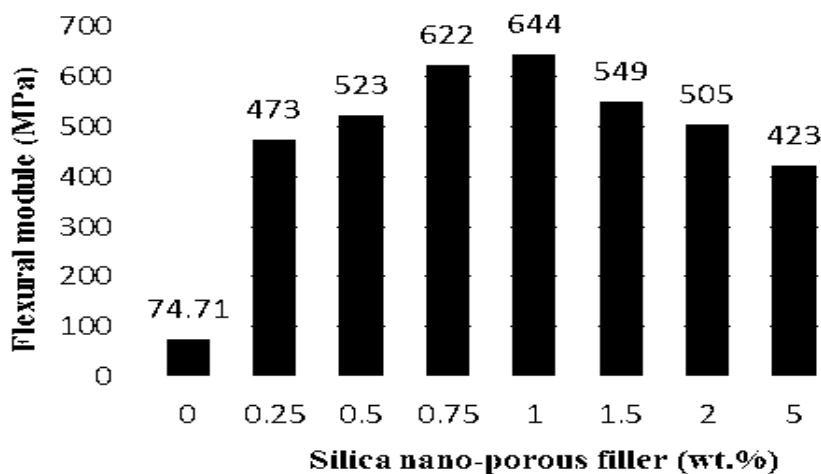


Fig.4: Flexural modulus of the adhesives containing different percentages of silica nano-porous filler

The fall of flexural modulus after 1.00 wt. % of filler content might be related to the presence of filler agglomerating as defect points. The second reason for the decline of flexural modulus with increasing the filler content is the incomplete curing of the adhesive.

#### 4. Conclusions

A direct effect of silica nano-sized filler was studied on the mechanical behavior, i.e. flexural modulus of dental adhesives, by using three-point bending test. The results showed that the flexural modulus increased for the sample containing 0–1.00 wt. % silica nano-porous filler up to the maximum value of 644 MPa, and then decreased with increasing the filler content. Adhesives synthesized in this paper show better mechanical properties due to the use of nanotechnology.

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