Available online at www.sciencedirect.com

ScienceDirect

Ceramics International 40 (2014) 4163–4171

CERAMICS INTERNATIONAL

www.elsevier.com/locate/ceramint

Low surface area nanosilica from an agricultural biomass for fabrication of dental nanocomposites

Mohammed Noushad*, Ismail Ab Rahman, Noor Sheeraz Che Zulkifli, Adam Husein, Dasmawati Mohamad

School of Dental Sciences, Health Campus, Universiti Sains Malaysia, 16150 Kubang Kerian, Kelantan, Malaysia

Received 23 June 2013; received in revised form 27 July 2013; accepted 19 August 2013
Available online 7 September 2013

Abstract

This study aimed to obtain nanosilica with desirable characteristics from an agricultural biomass waste using an organic acid. The effect of the feed rate of the precipitant and the mixing speed on the morphology and characteristics of nanosilica from rice husk for use as fillers in dental nanocomposites has been explored. The feed rate showed considerable effects on the agglomeration and the size of the silica nanoparticles. At a feed rate of 0.2 ml/min, the particles were bigger and highly agglomerated with a mean particle size of 261 nm. The mean particle sizes for the feed rates of 1 ml/min and 5 ml/min were 213 nm and 174 nm, respectively, exhibiting a decrease in the mean particle size with increasing feed rate. The shape of the silica nanoparticles depended on the mixing speed and it was possible to obtain spherical, dense, low surface area silica particles suitable for use in the fabrication of dental nanocomposites using this simple technique.

Keywords: B. Nanocomposite; Nanosilica; Rice husk; Dental; Filler

1. Introduction

Rice husk, a milling byproduct of rice, is an agricultural waste which is usually burnt in the open air or stacked on farmland, releasing large amounts of hazardous substances, occupying land resources, and polluting the environment [1]. It has a characteristic of hard surface, high silica content, small bulk density, not easily decomposed by bacteria and has traditionally been disposed in landfill. This can result in a source of pollution, eutrophication and perturbations in the aquatic and terrestrial life [2]. When used to generate electric power, the emission of rice husk ash into the ecosystem is associated with its persistent, carcinogenic and bio-accumulative effects, resulting in silicosis syndrome, fatigue, shortness of breath, loss of appetite and respiratory failure [3].

Rice husk was thought to be of no commercial value, but the silica in rice husk which is highly porous with light weight and high surface area has made it economically important [4]. Moreover, the commonly used raw materials used in the classic sol–gel method are relatively expensive [5]. The content of amorphous silica in rice husk is the highest in all gramineae plants [6] and several authors have concluded that rice husk is an excellent source of high grade amorphous silica [7–9].

Amorphous silica powder is a basic raw material that is widely used in industries associated with ceramics, rubber, electronics, catalysis, pharmaceutics, dental materials and other materials [10]. One application where the silica obtained from rice husk could be put into use is as fillers in dental composites which have not been studied widely. The dental composites available currently in the market use commercially prepared silica which greatly increases its cost due to the high cost of the precursors involved in its production. Hence, it would be useful if a relatively inexpensive source of fillers could be explored resulting in reduced cost.

The properties of dental composites are highly dependent on the characteristics of the fillers, like the shape, size, surface

*Corresponding author. Tel.: +60 179797396; fax: +60 97675505.
E-mail addresses: malcolmex13@yahoo.com (M. Noushad), arismail@usm.my (I. Ab Rahman), noorsheeraz87@yahoo.com (N.S. Che Zulkifli), adam@kck.usm.my (A. Husein), dasmawati@kck.usm.my (D. Mohamad).

0272-8842/$ - see front matter
http://dx.doi.org/10.1016/j.ceramint.2013.08.073

area and porosity. For example, a positive effect of the presence of nanofiller particles was observed by an improvement in flexural strength, surface hardness and fracture toughness [11]. Also, a combination of mesoporous and non-porous materials can be used to prepare stronger dental materials that may resist hydrolysis and wear [12]. Hence, the current study aimed to look into the manipulation of the morphology of the silica particles so that it could be used as fillers in dental nanocomposites.

Previous researches have mainly concentrated on obtaining silica with a high surface area [13,14]. Studies have shown that fillers with low surface area are favorable when used to fabricate dental composites. The larger surface area to volume ratio of the fillers has shown to increase water uptake resulting in the degradation of filler/matrix interface [15]. Moreover, a higher surface area also causes a reduction in the filler loading which is disadvantageous [12]. Several researchers have shown that filler loading reduces as a consequence of a high surface area to volume ratio, thereby limiting mechanical properties [16,17]. However, the inclusion of nanoclusters in the nanofilled material provided distinct mechanical and physical properties compared with those of the microhybrid resin based composites [15]. Therefore, fillers with lower surface area are preferable. The present study therefore intended to obtain silica with a low surface area ideal to be used in the fabrication of dental nanocomposites.

Studies have shown that spherical shaped fillers have a positive effect on the properties of the dental composites because spherical equiaxial fillers are free from entanglement and can be better dispersed in the matrix [18]. Composites with spherical filler particles are shown to exhibit lower shrinkage-stress values as compared to those with irregular filler particles. This is due to an increase in the ability of the dispersed phase to move within the matrix and relax stress, with increasing sphericity [19].

Deriving silica from rice husk has been extensively reported in the last two decades, but none has focused on obtaining silica ideal for use as fillers in dental composites. It is reported that active silica with a high specific area could be produced from rice husk ash after heat-treating at 973 K in air [20]. Moreover, it has been reported that the fungus *Fusarium oxysporum* rapidly biotransforms amorphous plant biosilica into crystalline silica and leach out silica extracellularly at room temperature in the form of 2–6 nm quasi-spherical, highly crystalline silica nanoparticles [8]. Mixed-phase bimodal mesoporous silicas (BMS) was synthesized by a simple sol–gel technique using rice husk ash-derived sodium silicate as a silica source [14]. A consecutive preparation of D-xylose and superfine silica from rice husk was also carried out [6]. Rice husk ash was treated with acid leaching and then boiled with base to leach silica [21]. Amorphous silica was successfully extracted at a 90.8% yield through base dissolution–acid precipitation of rice straw ash generated by a three-stage heating process [22]. A recyclable technology for preparation of silica powder using rice husk ash and NH_4F has also been proposed [23]. The current study employs a simple method to obtain spherical nanosilica particles.

Several researchers have looked into several aspects of the processing conditions on the morphology and characteristics of silica particles from rice husk. Previous studies have shown that it is possible to manipulate the morphology and characteristics of the silica particles by simple alterations in the extraction process. A study on the effects of calcination parameters, including temperature and time, on the silica phase of original and leached rice husk ash using X-ray diffraction (XRD) analysis showed the presence of totally crystalline silica in the original rice husk ash prepared at 800 °C but not in the leached rice husk ash [24]. They suggested that the leaching procedure, low calcination temperature and short calcination time to prepare more active amorphous silica from the rice husk. High purity of the silica gel has been found to be dependent on the reflux time and water loading by the addition of boiling deionized water to the silica gel prior to titration with 1 M H_2SO_4 [10]. A study utilized rice husk as an alternative silica source for the synthesis of MCM-22 showed that the duration required for zeolite crystallization was significantly decreased under varying-temperature conditions [25]. It was shown that the morphology of the silica particles from rice husk was dependent on several parameters like sodium silicate concentration, addition of solvent, pH, calcination temperature, etc. [26,27]. Although innumerable studies have been carried out taking into account different methods of utilizing rice husk in general and on obtaining amorphous silica in particular, there is no report looking into the effect of feed rate on the obtained silica particles to the best of our knowledge. In this study, the effect of the above mentioned parameters on the morphology and characteristics of nanosilica from rice husk with a proposal to use it as fillers in dental composites is reported.

2. Materials and methods

2.1. Materials

The rice husk was obtained from a local rice mill in Pak Badol, Kelantan, Malaysia. Sodium hydroxide (NaOH), hydrochloric acid (HCl) and 2-propanol were obtained from Merck (Germany). HCl was used in diluted form to remove the metallic impurities from the rice husk. All other chemicals were used without further purification. Deionized water was used wherever applicable.

2.2. Preparations

2.2.1. Extraction of silica from rice husk

Rice husks were washed thoroughly with water to remove the soluble particles and dust or other contaminants such as heavy impurities. They were then dried overnight in a hot air oven at 110 °C. The dried rice husks were heated in an acidic solution (1 M solution of HCl in a hot water bath) at 75 °C for 90 min to remove metallic impurities. The husks were thoroughly washed several times with deionized water. The wet solid was subsequently dried overnight in a hot air oven at 110 °C. The rice husks were heated in a 10% solution