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Komposit berpenguat struktur karbon aktif kayu (dari kayu eucalyptus-papuana)-zirkonia sebagai bahan alternatif pasak dental

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Abstrak

Latar belakang: Logam, keramik, dan polimer yang diperkuat telah digunakan sebagai bahan pasak dental. Komposit saat ini merupakan material yang sangat populer untuk pasak. Banyak jenis material telah dieksplorasi sebagai penguat serat untuk material pasak komposit tersebut, termasuk karbon. Dalam penelitian ini karbon dalam bentuk karbon aktif akan digabungkan dengan zirkonia (melalui *sol-gel templating*) dan PMMA, sebagai alternatif bahan pasak dental. Karbon aktif diekstrak dari kayu Eucalyptus-papuana, sumber daya alam yang melimpah dan ekonomis. **Tujuan**: 1) Membuat komposit berpenguat struktur karbon aktif-zirkonia sebagai bahan pasak dental. 2) Mempelajari pengaruh konsentrasi prekursor zirkonia terhadap sifat mekanis pasak komposit. Metodologi: Kayu Eucalyptus-papuana dipirolisis pada 750°C sehingga menghasilkan karbon aktif berpori tubular. Karbon aktif tersebut kemudian digabungkan dengan zirkonia melalui sol-gel templating dengan menggunakan 4 macam konsentrasi prekursor zirconia. Setelah kalsinasi pada 900°C, struktur karbon aktif-zirkonia diimpregnasi dengan PMMA. **Hasil**: Morfologi penampang transversal menunjukkan bahwa zirkonia terbentuk pada dinding tubular karbon aktif dan pori-pori tubular karbon ditutupi oleh PMMA dengan

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merata. Hasil *three-point bending* test menunjukkan bahwa semua pada material dengan semua konsentrasi prekursor tidak dapat menghasilkan kekuatan fleksural bahan yang setara dengan pasak komposit komersial (450-1100 Mpa), tetapi semua modulus elastisitas yang dihasilkan setara dengan pasak komposit komersial (8-50 Gpa). **Kesimpulan**: Nilai modulus elastisitas komposit yang dihasilkan oleh semua variasi konsentrasi zirconiaprecursor 0.05, 0.1, 0.4, and 0.8M, setara dengan pasak komposit komersial, dengan nilai yang tertinggi 24,247 GPa dihasilkan oleh konsentrasi 0,8 M. Semakin tinggi konsentrasi prekursor, semakin tinggi modulus elastisitas yang dihasilkan. Kekuatan fleksural bahan belum setara dengan pasak komposit komersial.

Kata kunci: pasak komposit dental, karbon aktif Eucalyptus papuana, PMMA, zirkonia, sol-gel

Reinforced composite with activated-carbon (from Eucalyptus-papuana wood)-zirconia as an alternative material for dental post.

Abstract

Background: Metals, ceramics, and reinforced polymers have all been used as dental post material. "FRC post = Fiber Reinforced Composite Post is very popular at this moment. Many kinds of materials have been explored as fiber reinforcement for this type of post, including carbon. In this study, activated carbon is combined with zirconia (through sol-gel templating) and PMMA, as an alternative material for composite dental post. Activated carbon is extracted from Eucalyptus papuana wood, a natural resource that is very abundant and economical. **Purpose**: 1) To make an activated carbon-reinforced zirconia composite structure. 2) To study the suitable zirconia-precursor concentration in enhancing the mechanical properties of the post material to besimilar to the commercial composite post. Methods: Eucalyptus-papuana wood was pyrolyzed at 750°C,producing activated-carbon with tubular pores. The synthesized carbon was then combined with zirconia, using sol -gel templating at 4 different zirconia-precursor molarities. After calcination at 900°C, the activated carbon-zirconia structure was impregnated with PMMA. **Result**: morphological image of the transverse surface zirconia showedattachment on the tubular wall of activated

carbon and the tubular pores and were evenly covered with PMMA. The 3-point bending test showed that all the zirconia-precursor concentrations failed to produce flexural strength in the range of commercial posts (450-1100 MPa), but all their elastic moduli are equal with commercial posts (8-50 GPa). The values of modulus of elasticity of this materialwere comparable to the commercial composite posts with the highest value of 24.247 GPa produced from 0.8 M precursor. The higher the precursor concentration, the higher the mechanical properties of the material **Conclusion**:the composite PMMA reinforced with activated carbon-zirconia structure was established, 2) the concentrations of the precursor 0.05, 0.1, 0.4, and 0.8 are correspondsto the modulus of elasticity ofcommercial composite post, but none of these concentrations could produce flexural strength equivalent to the commercial posts

Key words: dental composite post, Eucalyptus-papuana activated carbon, PMMA, zirconia, sol-gel.

Introduction

Dental postshave been widely used for dental restorations. Metal posts are still widely used today. These posts are less biocompatible because it tends to produce corrosion and these posts also have high mechanical properties that tend to cause root fractures.[1,2]

In the early‘90s, composite posts were developed. The most popular reinforcing materials used were glass fiber, silica fiber, and carbon fiber, with carbon fiber being regarded as having the best enhancedmechanical properties, but it is not aestheticallypleasing.These posts have been proven to be biocompatible, notcorrosive, anddo not have the tendency to cause a root fracture.[3-5]In Indonesia, the use of posts in dental restoration is on the rise. Based on a survey conducted by the Department of Health of the Republic of Indonesia, that 90% of Indonesian’s population suffers from dental diseases (caries).[6]However, all kinds of ready-made posts used in Indonesia are still an imported products. Therefore, it is important for research to be done in order to trigger the production of local products.

Indonesia’s natural resources, Eucalyptus-papuana wood can be used as carbon precursors. This isa promising alternative carbon source in terms of production costs and availability.[7] ·Now zirconia, a ceramic materials have been developed because of its aesthetic properties and mechanical properties that resembles metal. Zirconia can be attached to carbon to improve aesthetic and mechanical properties.

In this research, we will attempt to use carbon in its activated form, with sol gel technique (ceramic manufacturing technique that do not require high temperatures). This technique was chosen because it can be used in a simple dental laboratory without expensive equipments.The purpose of this study is to produce a reinforced composite material structure withactivated carbon-zirconia in aplication as an alternative material for composite posts and to study the effect of dental zirconia precursor concentrationon the mechanical propertiesofthe resultingmaterial.

Method

Below, is an explanation of the flow of the research:

Preparation of activated-carbon:

Eucalyptus-papuana wood were cut into cylinders measuring of 4mm x 3.5cm. The wood that had been cut was then preheated at 100°C for 4 hours to evaporate the water content. Then the wood was pyrolysed at 750°C under N2 atmosphere for 2 hours to produce active carbon [8].

Synthesis of zirconia by sol-gel method

Activated-carbon immersed inZrOCl2. 5H2Oprecursor with several concentrations: 0.05, 0.1, 0.4, and 0.8 molar. Ca $(NO_3)_2$ and chitosan were used as stabilizers and dispersants. The wholesystemwas vacuumed for 1 hour to remove the trapped air and then heated at 80°C for 4 hours. Carbon that has been soaked was then calcined at 900° C for 2 hours.[9]

Synthesis of post material: PMMA impregnation

PMMA was made of MMA monomer and BenzoylPeroxyde (BPO) initiator with the ratio of initiator/monomer is 2wt%. MMA-BPO solution was impregnatedto the carbon-zirconia structure with vacuum assistance. Carbon-zirconia structures that have been impregnated by monomer is then heated at 80°C for 8 hours.[10]

Material Characterization

1. Three-pointbendingtest; Fourgroupswere created according to the concentration of zirconia-precursor samples (0.05, 0.1, 0.4, 0.8). Each group consists of 3 pieces of the test samples. Tests were conducted according to ISO 4049:2009. The dimensions of the test samplesis a beam with 2x2x25mm size, the distance between the supporting points were 20 mm, with loading speed of 0.5 mm/min, and the load used is 5 kg.
2. Electron microscopy; Transverse cross-section morphology of activated-carbon, activated carbon-zirconia structure, activated-carbon-zirconia-PMMA were examined with an electron microscope JEOL JSM-6510LA.
3. Energy Dispersive Spectroscopy (EDS); Zirconia formation was confirmed by EDS.

Results

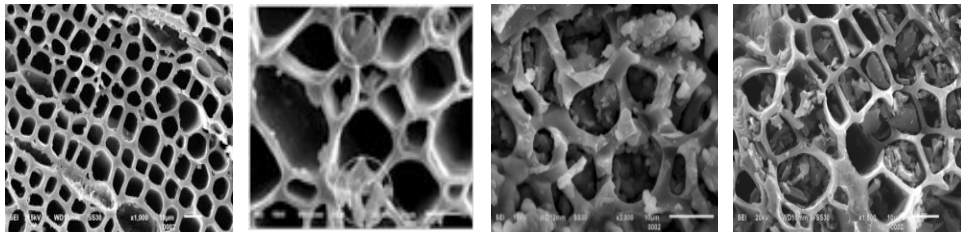
The results of mechanical tests such as flexural strength and modulus of elasticity can be seen in table 1.

Table 1. Flexural strength and modulus of elasticity of the material in several zirconiaprecursor molarities

Precursor molarity (M)	0,050	0,100	0,400	0,800
Flexural strength (Mpa)	37,081	65,096	64,459	103,989
Elastic modulus (Gpa)	13,316	16,796	17,258	24,247

The results of the activated-carbon structure formation

Figure 1.a shows the structure of activated-carbon from Eucalyptus-papuana wood. The structure consists of tubular pores that are not connected to each other and spread over the surface of the transverse surface resembling the original structure of Eucalyptus-papuana wood.[11]



a b c d

Figure 1. Transverse cross-sectional SEM micrography: a.) Activated-carbon, 1000x magnification, b.) Activated-carbon-zirconia (1 M precursor), 2500x magnification, c.) Activated-carbon-zirconia (0.8 M precursor), 1500x magnification, d.)activated-carbon-zirconia (0.1 M precursor), 2000x magnification.

The results of using activated-carbon from Eucalyptus-papuana wood templates for zirconia with sol-gel method.

The use of activated-carbon as a template in the synthesis of zirconia by sol-gel technique was intended to attach zirconia on the surface of activated-carbon. The formation of zirconia (ZrO_2) on the structure of activated-carbon is confirmed by tests using EDS.

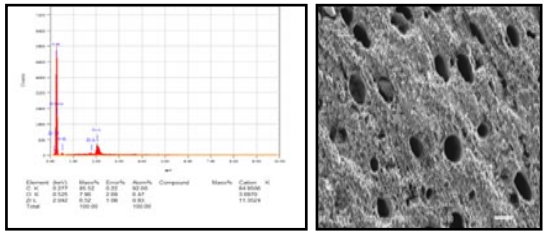


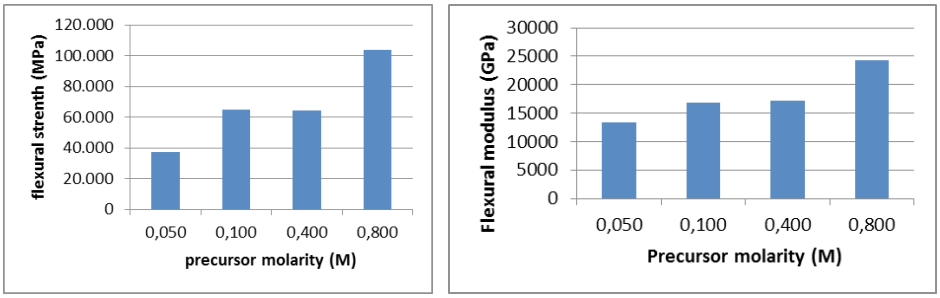
Figure 2. EDS results of activated-carbon-zirconia showed peaks of O and Zr, ZrO2 constituent elements StrengtheningEffect of Zirconia.

In figure 1.b, 1.c, and 1.d,shows that zirconia attached to the surface of activated-carbon tubular pores. From these photos, it can be observed visually that the amount of zirconia attached was higher,when higher concentrations were used . Comparison between the amount of zirconia formed by several concentrations of zirconia precursor used is illustrated in Table 2. An increased number of zirconia formed is due to the increasing amount of the zirconia particulatesat higher precursor concentrations.[12, 13]

Table 2. Comparison of amount of zirconia formed using different precursor concentrations.

Precursor Concentration(M)	Amount of Zirkonia
0,8	• • • •
0,4	• • •
0,1	• •
0,05	•

The intended use of zirconia in the composite is to improve the mechanical properties of resultant composites . The results of three-point-bendingtest supports the strengthening effect of the zirconia composites. The higher the concentration of zirconia precursor, the



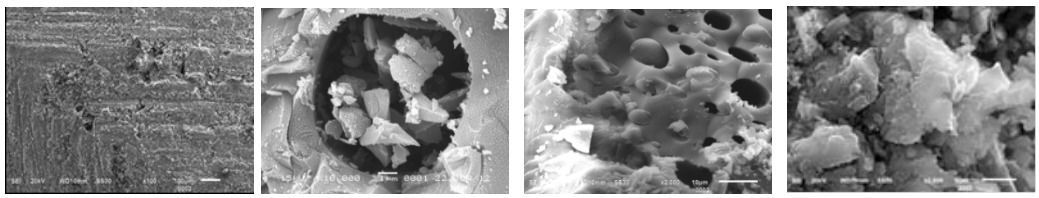
Graph 1.a.Flexural Strengthb. Elastic Modulus of activated carbon-zirconia-PMMA

higher the strength and elastic modulusof the composite (Graph 1.a and 1.b). In other words, thehigher the amount of zirconia in the composite, the higher the strength and modulus of elasticity of the composite.

Distribution and polymerization of PMMA after impregnation

SEM photos showed PMMA has formed evenly on the transverse cross section (Fig 3.a) although there are seen few voids, voids in the cross position were evenly distributed (not localized. Fig.3.b, and 3.c showed the different PMMA structures that form on the inside and the outside diameter of the samples.

PMMA on the inside diameter of the sample is forming an amorphous structure (Fig.3.b) and did not fill the entire the pore space. On the other hand, PMMA on the outside diameter of the sample formed a continuous structure, filling the entire pores homogeneously.



a b c d

Figure 3:Transverse cross-section of zirconia-PMMA activated carbon of the same specimen taken using SEM micrography a) 100x magnification, b) on the inner diameter sample, c) on the outer diameter sample (near the end of the sample), d) PMMA amorph

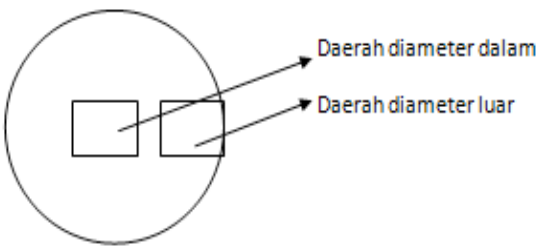


Figure 4. Illustration of specimen captured on SEM.

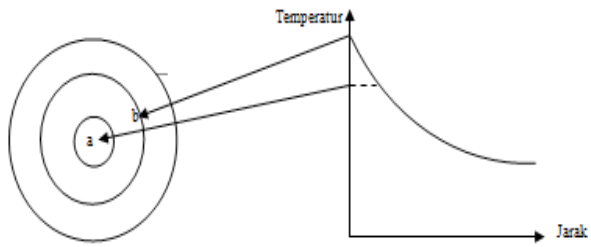


Figure 5. Illustration of the temperature gradient on the inside and outside of the especimen.

The inner structure of amorphous PMMA is similar to the snowflake structure described byliterature. Snowflake structure can be caused by inhomogeneous heating or the presence of additive materials. There is a difference in temperature between the outside and inside of the specimen as it is more difficult for heat to enter the center of the specimen.

Monomer and initiator used in the reaction arenot totally pure, they containedadditives.Additives produces gas when polymerization takes place. It is difficult for the gases formed in the middle of the specimen to diffuse out. However, it is easier for the gases formed on the outside of the sample to diffuse out.Lower polymerization temperature can also result in a lot of the remaining monomer (unpolymerized). The higher the polymerization temperature and the longer the polymerization time, the fewer the number of resisualmonomer.[15-17]

This reduces the flexural strength of the residual PMMA monomer present.Besides the influence of residual monomer, the number of voids due to incomplete polymerization

will also be the points of stress concentration at the three-point bending test, causing the specimen to fail easier than expected.

C**omparison with Fiber Composites (FRC) posts**

Flexural strength values obtained are not as high as expected, is not on par with commercial composite posts (450-1100 MPa). Thehighest flexuralstrength of the material is 103.989 was produced by using 0.8 M precursor.[2]In contrast, the modulus of elasticity is generated by all concentrations of the zirconia precursor is equivalent to the modulus of elasticity of commercial posts (8-50 Gpa).[2]The largest modulus of elasticity (24.247 GPa) was produced by a concentration 0.8 M zirconia precursor.

Conclusion

From this study it can be concluded that 1) the composite PMMA reinforced by activated-carbon-zirconia structure can be established, 2)the precursor concentrations of 0.05, 0.1, 0.4, and 0.8 corresponds to the modulus of elasticity of yielded composite which is equivalent to commercial composite posts, 3) but none of these concentrations could produce force equivalent to commercial flexural composite posts.

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