**RESEARCH ARTICLE**

# The Effect of Addition of Nano hydroxyapatite Powder of Anadara granosa Shells on Surface Roughness of Heat-cured Acrylic Resin

**Aris Aji Kurniawan[*], Dian Noviyanti Agus Imam[**], Helmi Hirawan[***]**

[*]Prosthodontic Department, Dentistry Programs, Medical Faculty, Universitas Jenderal Soedirman, Purwokerto, Indonesia

[**]Orthodontics Department, Dentistry Programs, Medical Faculty, Universitas Jenderal Soedirman, Purwokerto, Indonesia

[***]Oral Surgery Department, Dentistry Programs, Medical Faculty, Universitas Jenderal Soedirman, Purwokerto, Indonesia

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***ABSTRACT***

***Background:*** *Heat-cured acrylic resin is one of the most widely used denture base materials. However, the residual monomer can reduce its mechanical properties, which affect physical properties such as surface roughness, resulting in high porosity—the results in the growth of fungal colonies. One way to improve these properties is to add a reinforcement material such as nano-hydroxyapatite (HAp) from the Anadara granosa shell. Nano-hydroxyapatite has a large surface area to act as a filler because of its strong bonding capacity and a smoother surface, and a higher density to improve mechanical properties.* ***Purpose:*** *This study aims to determine HAp powder's effect from the Anadara granosa shells on the surface roughness of heat-cured acrylic resin.* ***Methods:*** *This type of research used an experimental laboratory, research design posttest-only control group design. Twenty-four samples were used and divided into three groups: the acrylic resin group with 1%, 3% blood cockles shell HAp, and acrylic resin without HAp with simple random sampling.* ***Results:*** *The results showed that the lowest mean value of surface roughness was in a group with the addition of 3% HAp, at 1.696 ± 0.25058 µm. One-Way ANOVA test on roughness showed a significant difference between groups with p = 0.000 (p <0.05).* ***Conclusion:*** *This study concludes that there is an effect of HAp on the roughness of heat-cured acrylic resin.*

***Keywords:*** *the Anadara granosa shell, heat-cured acrylic resin, nano-hydroxyapatite, surface roughness*

***Correspondence:*** *Aris Aji Kurniawan, Prosthodontic Department, Dentistry Programs, Medical Faculty, Universitas Jenderal Soedirman, Purwokerto, Indonesia, +6281285474035, arisajikurniawan@gmail.com*



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## INTRODUCTION

Heat-cured acrylic resin is one of the most widely used denture base materials. This type of resin has certain advantages such as lower toxicity, relatively prone to be processed and polished, more aesthetic values as well as affordable cost. However, the residual monomer can reduce mechanical properties which will affect physical properties such as surface roughness which results in higher porosity. This is due to higher growth on fungal colonies[1]. Surface roughness is one of the most significant physical properties of the denture base that needs further attention. Higher roughness on denture base will increase adhesions on bacterias, plaques and food leftovers. The tolerable surface roughness of the denture base shall be no more than 0.2 µm or less.[2]

Higher use of acrylic resin in dentistry has led to various measures in order to improve these properties, one of which is the addition of a reinforcing agent in the form of hydroxyapatite. Hydroxyapatite has osteoconductive properties due to approximately 65-70% of its components are bone-like components so that it shall not cause any allergic reactions. In addition, hydroxyapatite has also good biocompatibility and relatively stable thermodynamically.[3] Hydroxyapatite with calcium phosphate-based has its chemical compound $Ca_{10}(PO_4)_6(OH)_2$ (4). One of the most potential sources of hydroxyapatite is Anadara granosa shell/cockle. Cockle or Anadara granosa (A. granosa) is the most commonly consumed shellfish type by the public. The mineral compositions of anadara granosa shell are calcium carbonate and carbon, it content more than 98.7% of total mineral. The content of $Mg^{2+}$, $Na^{+}$, $P^{3-}$, $K_{+}$, $Fe^{2+}$, $Cu^{+}$, $Ni^{2+}$, it comprise approximately 1.3%[5]. Anadara granosa shell contains high calcium carbonate as well as with higher density on its particle size. Hydroxyapatite can be synthesized from Anadara granosa shell using low temperature hydrothermal method[6]. This material is widely used in Medicine and Dentistry sectors as a material to produce artificial bones and dentures due to it contains calcium phosphate with abundance of hydroxide.[4]

Hydroxyapatite can be made with certain forms, one of which is nanoparticle.[7] Nanoparticle is widely used as it has larger surface area so that it can be used as fillers.[8] The ratio between surface area and larger volume than similar particles on the nanometer scale makes its nano-particle become more reactive. Nano-hydroxyapatite has higher surface activity and a very fine structure similar to minerals found in hard tissues on the body and higher density which can improve its mechanical properties.[9,10]

Information on the addition of nano-hydroxyapatite on Anadara granosa shells to surface roughness of heat-cured acrylic resin is relatively small and has never been reported yet, nevertheless, certain researchers are becoming interested to carry out this study.

## METHODS

This study is purely laboratory experimental with using post test only control group design. A total of 24 heat-cured acrylic resin samples were used as research samples and divided into 3 groups, namely acrylic resin group with no addition of nano hydroxyapatite, by the addition of 1% and 3% nano-hydroxyapatite on Anadara granosa shell. The study sample was conducted at the Basic Dentistry Laboratory, Department of Dentistry, University of Jenderal Soedirman. Surface roughness of heat-cured acrylic resin was tested at Material Laboratory, Mechanical Engineering Department, Vocational School of Gadjah Mada University.

Procedures on this research commences with manufacture of metal disc molds measuring 10 mm in diameter and 2 mm thick. The next stage is to manufacture nano-hydroxyapatite on Anadara granosa shells. The dried Anadara granosa shells were mashed



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using a ball mill and weighed for 15 g. and then calcination was carried out in a furnace at $1000^0$ C for 3 hours. Calcined $Ca(OH)_2$ weighed to 18.03 g. $(NH_4)_2HPO_4$ was weighed to 16.63 g and then dissolved in 275 ml distilled water in a beaker, then to be homogenized in a magnetic stirrer with speed of 300 rpm for 150 minutes and to mix $Ca(OH)_2$ little by little. Formed precipitate is put into a petri dish and then dried in an oven for 12 hours with temperature of $120^0$C. After drying it then mashed it using a mortar and to be weighed. After that, TEM test was carried out in order to confirm shape and size of Hap.[5]

Nano-hydroxyapatite was added according to the concentration determined in polymer, then to be mixed with heat-cured acrylic resin monomer. The polymer in group I (with no addition of nano HAp powder) was 5 g with 2.5 ml of monomer. Group II with 1% concentration with polymer for 4.95 g and HAp nano powder for 0.05 g. Group III with 3% concentration with polymer for 4.85 g and nano HAp powder for 0.15 g.

The acrylic resin mix was put into a mold that had been smeared with *could mold* seal. Polymerized acrylic resin was cured using water bath at $70^0$C for 90 minutes and then continued to $100^0$C for 30 minutes and allowed to stand with room temperature. Nonetheless, finishing is done with using a tungsten carbide bur with speed of 15,000 rpm for 60 seconds on each sample.[11]

Surface roughness testing on acrylic resin samples was conducted by using Surface Roughness Tester SE 1700® (Profilometer). The profilometer has a radius-shaped stylus tip with a radius of 2 μm and movement speed of 0.500 mm/s. This instrument works by means of stylus tip touching and moving horizontally on surface of acrylic resin sample and acrylic resin sample must be placed with stable position. Tests were also conducted on 3 sample groups on cylindrical-type sample with diameter of 10mm and thickness of 2mm.[12]

One of surface roughness parameters is to calculate mean roughness of quadratic ($R_q$). Calculation on roughness mean of quadratic ($R_q$) with equation as follows (13):

$$Rq = \sqrt{\frac{(Yi - Ym)^2}{N}}$$

where:

Yi = Reference profile distance to measures profile
Ym = Reference profile distance to average lines
N = Quantity of Y

Then, obtained data was tabulated and analyzed with using One-Way ANOVA test as well as advanced test of Post Hoc LSD.

## RESULTS

The mean and standard deviation (SD) of surface roughness test results can be seen in Figure 1.

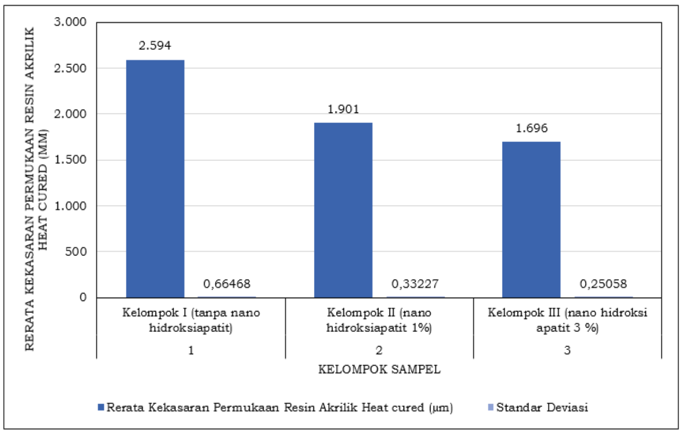


**Figure 1.** Mean and surface roughness on heat-cured acrylic resin.

Figure 1 shows that the highest surface roughness value is in group I with a mean of 2.594 ± 0.66468 μm and lowest surface roughness value is group III with a mean of 1.696 ± 0.25058 μm. Saphiro-Wilk test and Levene test showed that data were normally distributed and homogeneous ($p \geq 0.05$). Then, data on normally distributed and homogeneous were analyzed using One Way Anova test in order to determine is there any significant difference in surface roughness value between treatment group and control group. The results


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of One Way Anova test presented in Table 1 as follows.

**Table 1.** Result on *One-Way Anova* test on surface roughness value

| Sample group | F | Sig |
|---|---|---|
| Group I (with no nano hydroxiapatite) | | |
| Group II (1% of nano hydroxiapatite) | 8.644 | 0.002* |
| Group III (3% of nano hydroxiapatite) | | |

Note :
*= there is a significant difference inter-groups (p<0,05)

Based on result in Table 1, p value is 0.002 (p <0.05), which indicates that there is a significant difference between surface roughness values on each sample group. Nevertheless, Post Hoc Least Significant Differences (LSD) test was carried out in order to identify groups with significant differences. The results of Post Hoc Least Significant Differences (LSD) tests are presented in Table 2 as follows.

**Table 2.** Result on *post hoc* LSD test for surface roughness

| Group | Group I | Group II | Group III |
|---|---|---|---|
| Group I | | 0,006* | 0,001* |
| Group II | | | 0,376 |
| Group III | | | |

Table 2 shows that there is a significant difference in surface roughness of heat-cured acrylic resin (p≤0.01) in all treatment groups with control group. Meanwhile, there is no significant difference between groups II and III (p> 0.01).

## DISCUSSION

Objective of this study is to determine the effectiveness on adding nano-hydroxyapatite to Anadara granosa shells by reducing its surface roughness of heat-cured acrylic resin. In this study, nano-hydroxyapatite of Anadara granosa shells was used with concentration of 1%, 3% respectively and with no addition of nano-hydroxyapatite on Anadara granosa shells as a control group. Concentration selection is based on study of Kusumawardani et al.[3] by adding 2% of hydroxyapatite to acrylic resin can minimize residual monomers and increase its compressive strength, therefore, 1% and 3% concentrations were selected in this study.[3]

Results of this study indicate that lowest surface roughness value is in group with the addition of nano-hydroxyapatite for 3% and the highest is in group with no addition of nano hydroxyapatite. Significant differences were shown between groups with addition of 1% of nano-hydroxyapatite on Anadara granosa shells and 3% if nano-hydroxyapatite on anadara granosa shells and groups without addition of nano-hydroxyapatite on Anadara granosa shells. However, there was no significant difference between addition of 1% and 3% of nano-hydroxyapatite in Anadara granosa shells. Factors that can cause decrease of surface roughness include the presence of high interfacial bonding between nano-hydroxyapatite and polymer matrix, good dispersion and even distribution of nano-hydroxyapatite in polymer. Nanometer size and low concentration of nano-hydroxyapatite shall promote good dispersion in heat-cured acrylic resin.[14] In addition, nano-hydroxyapatite can play a role in binding heat-cured acrylic resin monomers.[15]

The capability of nano-hydroxyapatite on Anadara granosa shells can be made as a filler or nanofiller when manipulated polymer and monomer cause a decrease in surface roughness. Nano-hydroxyapatite on Anadara granosa shells has a large surface area so that its binding power is relatively solid. Based on TEM test conducted, it was found that particle size of nano-hydroxyapatite powder on Anadara granosa shells ranges from 20 to 200 nm. Nanometer particle size of nano-hydroxyapatite on anadara granosa shells will fill the porosity



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gap and increase inter-matrices binds. This conforms with the study of Chondro et al.[15] regarding addition of 2% and 5% concentration of hydroxyapatite powder which indicates that there is a decrease in porosity level of heat-cured acrylic resin.[15]

The addition of hydroxyapatite to heat-cured acrylic resin is able to minimize residual monomers as well as produce a homogeneous texture but without any chemical reactions (16). Nano-hydroxyapatite powder that fills porosity gaps on inter-matrices of heat-cured acrylic resin will bind mechanically. Reduced residual monomer in acrylic resin will affect physical properties such as decreased surface roughness.[17]

To mix with nano-hydroxyapatite during manipulation process into chain binds will not change its basic structure, however, it can react with polymethyl methacrylate. Nano-hydroxyapatite will fill gaps and bind to matrix walls in newly-formed gaps and fill gaps until it closed and form new surfaces (3). FTIR test result regarding the addition of hydroxyapatite to PMMA has no correlation chemically whatsoever. In PMMA spectrum, not only transformation on group C = C and C-C groups into polymers either before and after addition of hydroxyapatite. This means that addition of hydroxyapatite does not cause any chemical reactions, instead of a mechanical bond.[16]

On addition of nano-hydroxyapatite with a concentration of 3% resulted in number of hydroxyapatite nanoparticles becoming more and more evenly dispersed between chains of polymethyl methacrylate, causing some of microporosity to be closed and reducing surface roughness on acrylic resin.[13]

## CONCLUSION

The addition of nano-hydroxyapatite on Anadara granosa shells can lower surface roughness of heat-cured acrylic resin, the best reduction was the addition of 3% nano-hydroxyapatite on Anadara granosa shells.

## REFERENCES

1. Anusavice K J. Phillips science of dental material ed. 11th. St. Louis: Elsevier science; 2013. hal. 176-179.
2. Syafriani, Setiawan Y. Perbedaan kekasaran permukaan basis resin akrilik polimerisasi panas menggunakan bahan pumis cangkang telur dan pasta gigi sebagai bahan poles. J Ilm PPANMED. 2017; 12(2): 200–203.
3. Kusumawardani CDN, Chondro RT, Andrian I, Sari RP. Pengaruh penambahan hidroksiapatit terhadap porositas dan compressive strength basis resin akrilik heat-cured. Jurnal Kedokteran Gigi Universitas Padjadjaran. 2020; 32(2): 91-98.
4. Afrizal G. Analisa struktur mikro material substitusi hidroksiapatit cangkang kerang darah dan resin akrilik bahan pembuat gigi untuk aplikasi gigi tiruan. Surya Teknika. 2016; 4: 1-9.
5. AJ A-H, Bakar MZA, Noordin MM, Abu J, Norimah Y. Mineral composition of the cokle (anadara granosa) shells of west coast of peninsular malaysia and it's potential as biomaterial for use in bone repair. J Anim Vet Adv. 2005; 6(5):591–594.
6. Muhara, Ikhbal, Fadli A, Akbar F. Sintesis hidroksiapatit dari kulit kerang darah dengan metode hidrotermal suhu rendah. Jom FTEKNIK. 2015; 2(1): 7.
7. Mollazadeh S, Javadpour J, Khavandi A. In situ synthesis and characterization of nano-size hydroxiapatite in poly (vinyl alcohol) matrix. Ceram int. 2007; 33(8):1579–1583.
8. Maryam, Kasim A, Novelina, Emriadi. Review: teknologi preparasi pati nanopartikel dan aplikasinya dalam pengembangan komposit bioplastik. Majalah ilmiah teknologi industri. 2018; 15(1): 36-56.
9. Abdullah M, Virgus Y, Nirmin, Khairurrijal. Review: sintesis nanomaterial. J Nanosains dan Nanoteknologi. 2008; 1: 33–57.
10. Qulub F, Widiyanti P, Ady J. Variasi komposisi nano hidroksiapatit pada poly (1,8-Octanediol-co-citrate)(poc) sebagai biodegradable bone screw. Jurnal Sains Materi Indonesia. 2015; 16(4):188-192.
11. Riyadi W, Purasasmita BS, Imam DNA. Penambahan nanoselulosa sekam padi terhadap kekuatan fleksural basis gigi tiruan resin akrilik polimerisasi panas. E-Prodenta Journal of Dentistry. 2020; 4(2): 336-342.

12. Sundari S. Studi kekuatan fleksural antara resin akrilik heat-cured dan termoplastik nilon setelah direndam dalam minuman ulee kareng (coffea robusta). Journal Dental Sains. 2016; 1(1): 31-38.
13. Saputro H. Model matematika untuk memprediksi kekasaran permnukaan hasil proses cnc bubut tanpa pendingin. Traksi. 2010; 10(1): 18-31.
14. Karadi HR, Hussein BMA. Effect of modified nanohydroxyapatite fillers addition on some properties of heat-cured acrylic denture base materials. J Bagh College Dentistry. 2017; 29(2):49-54.
15. Chondro RT, Nanik CD, Sari RP. Efektivitas penambahan hidroksiapatit terhadap penurunan porositas basis resin akrilik heat cured. Denta.2019; 13(2): 37-42.
16. Mozartha M. Hidroksiapatit dan aplikasinya di bidang kedokteran gigi. Cakradonya Dent J. 2015; 7(2): 807-868.
17. Hassan ZJ, Hatim NA, Taqa AA. Study the FTIR of hydroxyapatite additive to heat-cured acrylic resin. Al-Rafidain Dent J. 2014; 14(1):32-36.