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- Betel leaf toothpastes inhibit dental plaque formation on fixed orthodontic patients
- Volatile sulphur compounds elimination: A new insight in periodontal treatment
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Research Report

The difference of residual monomer levels of acrylic resin with various polymerization method

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ABSTRACT

Background: After polymerization process, heat cured acrylic resin denture base actually still contains residual monomers that can become potential irritants later in oral cavity. Polymerization process is essential to obtain acrylic resin which can meet the requirements of the biocompatible and good physical properties. To meet the requirements, there are several methods of polymerization process used.

Purpose: The purpose of this study was to determine the differences of the residual monomer levels of acrylic resin processed by various polymerization methods. **Methods:** Acrylic resin powder and liquid were mixed based on the rules of factory, and sample was made with size of 30 mm × 50 mm × 3 mm and then polymerized by using microwave at 70° C for 24 hours based on the methods of Japan Industrial Standard (JIS). Each group of samples was cut with weight of ± 0.2 g, dissolved in 5 ml of methyl ethyl ketone in test tubes, and then stored at ± 5° C for four days. Residual monomer level was conducted by using gas chromatograph mass spectrometer. Data obtained were then analyzed by using One-Way ANOVA test with $p < 0.05$. **Results:** After the level of polymerizing residual monomer with JIS method was compared to that at 70° C for 24 hours using microwave, it is known that there were significant differences ($p < 0.05$). **Conclusion:** The highest level of residual monomer of acrylic resin was that polymerized at 70° C for 24 hours.

Key words: Acrylic resin, residual monomer, gas chromatograph

ABSTRACT

Latar belakang: Basis gigi tiruan yang berbahan dasar resin akrilik jenis heat cured setelah proses polimerisasi selesai masih mengandung monomer sisa yang berpotensi sebagai bahan iritan dalam rongga mulut. Proses polimerisasi sangat penting untuk mendapatkan resin akrilik yang memenuhi persyaratan biokompatibilitas dan fisik yang baik. Untuk persyaratan tersebut digunakan berbagai macam proses polimerisasi. **Tujuan:** Penelitian ini bertujuan untuk menentukan kadar monomer sisa resin akrilik yang diproses dengan metode polimerisasi berbeda menggunakan gas chromatograph mass spectrometer. **Metode:** Bubuk dan cairan resin akrilik dicampur sesuai aturan pabrik dan sampel dibuat berukuran 30 mm × 50 mm × 3 mm dipolimerisasi berdasarkan metode Japan Industrial Standard (JIS), suhu 70° C selama 24 jam, dan microwave. Setiap kelompok sampel dipotong seberat ± 0,2 gram dilarutkan 5 ml dalam metil etil keton pada tabung uji dan disimpan suhu ± 5° C selama empat hari, dilakukan analisis kadar monomer sisa dengan gas chromatograph mass spectrometer. Data yang diperoleh dianalisis ANOVA satu arah dengan nilai $p < 0,05$. **Hasil:** Kadar monomer sisa resin akrilik yang berpolimerisasi metode JIS dibandingkan polimerisasi suhu 70° C selama 24 jam dan microwave terdapat perbedaan bermakna ($p < 0,05$). **Kesimpulan:** Kadar monomer sisa paling tinggi pada resin akrilik yang dipolimerisasi suhu 70° C selama 24 jam.

Kata kunci: Resin akrilik, monomer sisa, gas chromatograph

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INTRODUCTION

Polymethyl methacrylate is the basic material of heat cured acrylic resin a major choice in the prosthodontics for making the denture base. It is still the main choice of dentists although metallic materials or polyamide-mixed materials have widely been used as removable denture base. Acrylic resin is easily manipulated and repaired if fracture occurs, it can meet aesthetic needs because its translucent and color stability are good enough, it is not toxic and not soluble in oral fluids, and its absorption is relatively low and relatively cheap price. In general, acrylic resin actually has low strength, either impact power or resistance strength to fracture, but it is still quite flexible. Another character is that acrylic resin can cause dimensional changes occurred during polymerization process and crack that might be found on the surface of acrylic resin.^{1,2}

Acrylic resin material used in dentistry is generally available in powder and liquid. Some polymerization methods and curing processes, such as chemical activation, visible light, water heating, microwave energy, are then needed to manipulate the material into a solid material. This type of activation is used to initiate the formation of free radicals as the initial polymerization process of acrylic resin.³ The polymerization process is important to obtain acrylic resin which can meet the requirement of the physical character which is biocompatible to the tissues of oral cavity. Thus, to meet the requirement, there are several methods of polymerization process that can be used. Thermal polymerization method or conventional one is most often used in addition to using microwave. Polymerization method using microwave has the advantages of saving time, clean, being not porous, with physical mechanical properties and dimensional changes similar to that processed conventionally.⁴ Japan Industrial Standard (JIS) is also introduced to the polymerization of acrylic resin, which is heated in water for 90 min at 70° C and was continued at 100° C for 30 minutes.⁵ The polymerization reaction of acrylic resin actually tends not to produce perfect unresidual monomer, but it still leaves residual monomer that has still not reacted yet, called the residual monomer of methyl methacrylate.³

The selection of the proper method of polymerization, as a result, is very important because it deals with the levels of residual monomer which will affect the quality of acrylic resin denture base. The residual monomer of acrylic resin may also be released into the water or saliva potentially causing irritation, inflammation, hypersensitivity, and allergic responses in mucosal tissue.^{1,6} therefore, it is necessary to know the residual monomer levels of acrylic resin which is processed by several polymerization techniques to avoid unacceptable impacts.

The residual monomer levels of acrylic resin can actually be determined by several kinds of methods, such as by using infrared spectroscopy, gas chromatography, high performance liquid chromatography (HPLC) and fluorescent flow injection.³ Gas chromatography is a

very precise and quick method to separate very complex mixtures. The duration needed varies from a few seconds for a simple mix up to hours for a complex mixture so that the method is widely used by researchers to analyze the residual monomer levels. Therefore, the purpose of this study was to determine the differences of the residual monomer levels of acrylic resin processed by various polymerization methods.

MATERIALS AND METHODS

Gypsum made of water and gypsum with ratio of 24ml: 10grams was stirred on vibrator, and then poured into the cuvette. Master model measuring 30 mm × 50 mm × 3 mm was placed in the middle of cuvette already containing the hard gypsum (New plastone GC, Japan), and then was abandoned until the gypsum got harder. After the gypsum got harder, could mould seal was applied on the surface, and the top of cuvette was filled with the dough on vibrator. The master model that had been planted in the cuvette was abandoned for 24 hours. Then, the master model in the cuvette was taken, so the mould was obtained. The cuvette was then filled with acrylic resin (Bioresin, Shofu Japan; and Acron MC, GC Dental Industrial Japan), with a ratio of 10 grams of powder: 4.5 ml of fluid (based on the manufacturer's instructions).

After 20 minutes, the dough reached the dough stage. The mould on which separator had already been applied was filled with acrylic resin dough. Before the cuvette was closed, acrylic was coated with celluloid plastics and pressed gently with hydraulic bench press (Yoshida, Japan). Next, the cuvette was reopened, and the excess of acrylic resin was cut and then closed again. Afterwards, pressing was conducted with pressure of 2200 psi or 50 kg/cm². This procedure was repeated three times, and then transferred to the clamp abandoned for 24 hours. Next, curing process was then conducted by heating water at 70° C for 90 min based on JIS, and then followed at 100° C for 30 minutes. This procedure was repeated for the other treatment groups, but the polymerization was conducted by heating water at 70° C for 24 hours. Making samples curing by microwave used a ratio of 10 grams of powder: 4.3 ml of fluid (based on the factory's instructions). Polymerization process was then conducted with microwave oven (500 watts) for 3 minutes.

Group I was acrylic resin polymerizing by using JIS method. Group II was acrylic resin polymerizing at 70° C for 24 hours. Group III was acrylic resin polymerizing with microwave. After all of the polymerization processes were completed, the samples were abandoned until got cool. The samples in each treatment group consisted of six pieces, so the total samples were eighteen pieces. Prior to being tested, the samples were stored in distilled water 37° C ± 1° C for 48 hours.⁵

The determination of the residual monomer levels of acrylic resin was then conducted by using gas chromatograph

mass spectrometer (JEOL JMS DX 303) and mass analysis/data processing system (JEOL JMA DA 5000) (JEOL Ltd., Japan). The samples that had already polymerized were cut into small pieces weighing ± 0.2 grams. Each treatment group sample was then dissolved in 5 ml of methyl ethyl ketone (Merck) in a test tube, and kept at $\pm 5^\circ\text{C}$ for four days (96 hours). The determination of the residual monomer was conducted based on the operating conditions of gas chromatograph mass spectrometer instrument as follows: helium carrier gas ($P = 0.5 \text{ kg/cm}^2$, flow $\pm 1 \text{ ml/min}$) using supelcowax column 10, fused silica capillary column (= carbowax 20 m = SP 1000), film thickness injector: splitless (60 seconds), ionization voltage 70 eV; current 100 A, acceleration 3 kV, chamber temperature 150°C , injector temperature 190°C , and program T1 (initial 40°C and T2 70°C with the increasing of 4°C/minute). Data obtained were then analyzed by using One-Way ANOVA test with $p < 0.05$.

RESULTS

It is known that the polymerization of acrylic resin at 70°C for 24 hours has the highest residual monomer level compared to the other polymerization methods (Table 1).

Table 1. The mean and standard deviation of the residual monomer levels of acrylic resin with different polymerization methods (mg/l)

Treatment groups	Mean \pm SD
Polymerization with JIS method	0.3700 \pm 0.1128
Polymerization at 70°C for 24 hours	2.0717 \pm 0.3100
Polymerization with microwave	0.6917 \pm 0.0572

After being tested with one sample Kolmogorov-Smirnov test ($p > 0.05$), it is known that the data obtained were homogeneous and normally distributed. To find out the differences of the residual monomer levels of acrylic resin processed by different polymerization methods, statistical calculations was conducted by using One-Way ANOVA. It is then known that there were significant differences with p value < 0.05 . Furthermore, to determine the differences of the polymerization methods towards the residual monomer levels of acrylic resin, LSD test was conducted (Table 2).

Table 2. The results of LSD test on the residual monomer levels of acrylic resin processed by different polymerization methods (mg/l)

Polymerization methods	Polymerization with JIS method	Polymerization at 70°C for 24 hours	Polymerization with microwave
Polymerization with JIS method	-	*	*
Polymerization at 70°C for 24 hours		-	*
Polymerization with microwave			-

Note: *: Significant

There were significant differences on the residual monomer level of acrylic resin polymerized with JIS method compared to that polymerized at 70°C for 24 hours and that polymerized with microwave.

DISCUSSION

Acrylic resin is a polymer that is popularly used in dentistry. It is known that ninety-five percents of patients use heat cured acrylic resin denture.⁷ Heat cured acrylic resin heat is a mixture of methyl methacrylate monomer and poly methyl methacrylate polymer through polymerization process conducted after heating. In this case, acrylic resin polymerizes additionally. If at the time of the polymerization, temperature increases to 60°C , then bensoil peroxide which acts as initiator will decompose into free radicals. Free radicals will then further react with monomer to form new free radicals resulting in from the occurrence of propagation reaction to the occurrence of termination. Polymerization process at too low temperature or with too short duration even will cause weak acrylic resin due to short chain polymer.¹

Method of acrylic resin polymerization used in this study showed the significant differences of the residual monomer levels. These results are also supported by the results of the previous researches.⁸ Some researchers also reported that the composition and the process of polymerization of denture base made of acrylic resin could affect on the release of residual monomer.⁹ The residual monomer levels of acrylic resin polymerize at 70°C for 24 hours was higher than that with JIS method and microwave. This is due to temperature used below the temperature of Tg (glass transition temperature), as a result, the polymerization of methyl methacrylate monomer will only slightly occur since methyl methacrylate during the polymerization is not mobilized, therefore, there are many residual monomer.¹⁰

Another opinion states that the residual monomer level can be reduced if the duration of the polymerization process is extended and the temperature is enhanced.¹¹ As a results, the polymerization process of acrylic resin using too low temperature or too short duration of curing process will generate residual monomer and high porosity that can reduce the strength of denture base made of acrylic resin which later can facilitate the occurrence of fractures. The

residual monomer that remains in the acrylic resin denture base will adversely affect the mechanical properties since the residual monomer acts as plasticizer that makes acrylic resin denture base soft and flexible.¹² Moreover, the residual methyl methacrylate monomer can also come out into the surrounding tissues¹³ which later may cause side effects, such as the hypersensitivity of oral tissue and the changing of color stability.¹⁴ Clinical signs and symptoms are often reported including erythema, erosion, oral mucosa, and burning sensation in mucosa and tongue.⁶ According to ISO 1567, the maximum residual monomer of denture base material shall not be more than 2.2% of weight.¹⁴

The residual monomer levels of acrylic resin used as a denture base, therefore, showed be reduced after the polymerization process. Some researchers even recommend to soak it in water for at least 24 hours before inserting it into patients in order to make the residual monomer become not potentially toxic. Other researchers, moreover, said that after polymerization, acrylic resin can be soaked in water at 50° C for one day to reduce the release of the residual monomer because the water heated can cause the monomer molecules more rapidly spread, so it can enhance the reaction of polymerization.⁶

Polymerization of acrylic resin with JIS method can produce the lower residual monomer than that with other polymerization methods. Acrylic resin polymerizing can usually reach the boiling point of monomer, and can also produce the lower residual monomer than that polymerizing without reaching the boiling point.¹⁵ It means that the use of high heat will cause the mobility of the molecule chain that facilitate the conversion of monomers into polymers.⁶ Thus, it is known that there was significant difference between the residual monomer levels of acrylic resin polymerizing with microwave and that polymerizing at 70° C for 24 hours. It is because the polymerization process with microwave can produce internal heat due to the high frequency of electromagnetic waves causing the faster monomer molecule movement and the more complete polymerization reaction, consequently, the residual monomer was reduced. Previous researchers even concluded that the polymerization of acrylic resins with microwave compared to that with the pressure and injection molding methods can cause the significant reduction of the

residual methylmethacrylate monomer.³ Finally, it can be concluded that the highest residual monomer level of acrylic resin is that polymerized at 70° C for 24 hours.

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