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THE PHYSICAL PROPERTIES OF AUTO-POLYMERIZING HARD DENTURE RELINING MATERIALS AFTER ULTRASONICAL CLEANED WITH ETHANOL SOLUTIONS

Miss Sutasinee Soontornwipath

A Thesis Submitted in Partial Fulfillment of the Requirements
for the Degree of Master of Science Program in Prosthodontics
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By	Miss Sutasinee Soontornwipath
Field of Study	Prosthodontics
Thesis Advisor	Associate Professor Chairat Wiwatwarrapan

Accepted by the Faculty of Dentistry, Chulalongkorn University in Partial Fulfillment of the Requirements for the Master's Degree

.....Dean of the Faculty of Dentistry
(Assistant Professor Suchit Poolthong, Ph.D.)

THESIS COMMITTEE

.....Chairman
(Assistant Professor Prarom Salimee, Ph.D.)

.....Thesis Advisor
(Associate Professor Chairat Wiwatwarrapan)

.....Examiner
(Assistant Professor Wacharasak Tumrasvin, Ph.D.)

.....External Examiner
(Assistant Professor Vanthana Sattabanasuk, Ph.D.)

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โดยทั่วไปทันตแพทย์นำวัสดุเสริมฐานฟันเทียมอะคริลิกชนิดแข็งมาใช้เสริมฐานฟันเทียมให้แนบสนิทกับสันเหงือกที่ยุบตัวลงไปจากการละลายตัวของสันกระดูกเมื่อมีการใช้งานฟันเทียมไประยะหนึ่ง แต่ปัญหาหลักของวัสดุเสริมฐานฟันเทียมชนิดเรซิน คือมีมอนอเมอร์ตกค้างภายหลังการเกิดปฏิกิริยาพอลิเมอร์ ซึ่งมีผลเสียคือ มีความเป็นพิษและทำให้สมบัติทางกายภาพของวัสดุเสริมฐานฟันเทียมต่ำลง การวิจัยพบว่าการใช้สารละลายเอทานอลในเครื่องล้างอัลตราโซนิกส์สามารถลดมอนอเมอร์ตกค้างได้อย่างมีประสิทธิภาพในวัสดุเสริมฐานฟันเทียมชนิดแข็งที่บ่มด้วยตนเอง วัตถุประสงค์ของการศึกษาครั้งนี้คือประเมินกำลังดัดขวาง โมดูลัสของแรงดัดขวาง ความแข็งผิว การดูดซับน้ำ และการละลายตัวในน้ำของวัสดุเสริมฐานฟันเทียมอะคริลิกบ่มด้วยตนเองชนิดแข็งทั้ง 4 ผลิตภัณฑ์ ได้แก่ Unifast Trad (UT), Kooliner (KL), Ufi Gel Hard (UG) และ Tukuso Rebase II (TR) หลังจากการแช่ในเครื่องล้างอัลตราโซนิกส์ที่มีสารละลายเอทานอลที่ความเข้มข้นต่างๆ สำหรับวัสดุแต่ละผลิตภัณฑ์ ผู้วิจัยเตรียมชิ้นงานจำนวน 90 ชิ้น และแบ่งออกเป็น 9 กลุ่ม ประกอบไปด้วย กลุ่มควบคุมผลลบ (NC) คือกลุ่มที่ไม่ได้ทำการลดมอนอเมอร์ตกค้าง กลุ่มควบคุมผลบวก ได้แก่ กลุ่มที่ทำการลดมอนอเมอร์ตกค้างโดยแช่ในน้ำอุณหภูมิ 50 องศาเซลเซียส เป็นเวลา 1 ชั่วโมง (PC1) กลุ่มที่แช่ในน้ำอุณหภูมิ 55 องศาเซลเซียส เป็นเวลา 10 นาที (PC2) และกลุ่มที่แช่ในเครื่องล้างอัลตราโซนิกส์ด้วยสารละลายเอทานอลความเข้มข้น 0%, 10%, 20%, 30%, 40% และ 50% ที่อุณหภูมิ 55 องศาเซลเซียส เป็นเวลา 5 นาที ทำการทดสอบกำลังดัดขวางและค่าโมดูลัสของแรงดัดขวาง โดยวิธี Three point transverse test จากนั้นนำส่วนที่หักของชิ้นงานไปทดสอบความแข็งผิวแบบวิคเกอร์ ส่วนคุณสมบัติการดูดซับน้ำและการละลายตัวในน้ำทำการทดสอบตามข้อกำหนดขององค์การมาตรฐานสากลหมายเลข 20795-1:2013 นำข้อมูลมาวิเคราะห์ทางสถิติด้วยการวิเคราะห์ความแปรปรวนแบบทางเดียว และเปรียบเทียบความแตกต่างระหว่างกลุ่มด้วยวิธีการของตูกี และวิธีการของดันเนท ที่ระดับความเชื่อมั่นร้อยละ 95 ผลการศึกษาพบว่าค่ากำลังดัดขวาง ค่าโมดูลัสของแรงดัดขวาง ค่าการดูดซับน้ำและการละลายตัวในน้ำของวัสดุเสริมฐานฟันเทียมอะคริลิกบ่มด้วยตนเองชนิดแข็งในแต่ละผลิตภัณฑ์ มีค่าแตกต่างกันอย่างมีนัยสำคัญ ($p<0.05$) ในขณะที่ค่าความแข็งผิวมีค่าไม่แตกต่างกันอย่างมีนัยสำคัญ ($p>0.05$) ในแต่ล่ะกลุ่มของวัสดุเสริมฐานฟันเทียมชนิดเดียวกัน นอกจากนี้เมื่อทำการเปรียบเทียบคุณสมบัติของวัสดุเสริมฐานฟันเทียมภายหลังการทดสอบด้วยวิธีการเดียวกัน พบว่าค่ากำลังดัดขวาง ค่าโมดูลัสของแรงดัดขวาง ค่าความแข็งผิว ค่าการดูดซับน้ำและการละลายตัวในน้ำมีค่าแตกต่างกันอย่างมีนัยสำคัญ ($p<0.05$) โดยวัสดุเสริมฐานฟันเทียมชนิด UT มีค่ากำลังดัดขวาง ค่าโมดูลัสของแรงดัดขวาง ค่าความแข็งผิว และค่าการดูดซับน้ำมากกว่าวัสดุชนิดอื่น ส่วนค่าการละลายตัวในน้ำวัสดุ TR มีค่ามากที่สุด สรุปผลการศึกษา วัสดุเสริมฐานฟันเทียมชนิด KL ภายหลังจากการลดมอนอเมอร์ตกค้างด้วยสารละลายเอทานอลความเข้มข้น 10% ในเครื่องล้างอัลตราโซนิกส์เป็นทางเลือกที่มีประสิทธิภาพที่ช่วยเพิ่มคุณสมบัติทางกายภาพให้กับวัสดุ วัสดุเสริมฐานฟันเทียมชนิด UG กับ TR การแช่ด้วยสารละลายเอทานอลที่ความเข้มข้น 10% และ UT ที่ความเข้มข้น 30% ในเครื่องล้างอัลตราโซนิกส์ไม่มีผลต่อคุณสมบัติทางกายภาพของวัสดุเสริมฐานฟันเทียม

ภาควิชา ทันตกรรมประดิษฐ์ ลายมือชื่อนิสิต .....

สาขาวิชา ทันตกรรมประดิษฐ์ ลายมือชื่อ อ.ที่ปรึกษาหลัก .....

ปีการศึกษา 2559

5775827132 : MAJOR PROSTHODONTICS
KEYWORDS: AUTO-POLYMERIZING ACRYLIC HARD DENTURE RELINE / FLEXURAL STRENGTH / RESIDUAL MONOMER / ULTRASONIC CLEANER / WATER SORPTION

SUTASINEE SOONTORNWIPATH: THE PHYSICAL PROPERTIES OF AUTO-POLYMERIZING HARD DENTURE RELINING MATERIALS AFTER ULTRASONICAL CLEANED WITH ETHANOL SOLUTIONS. ADVISOR: ASSOC. PROF. CHAIRAT WIWATWARRAPAN, pp.

Hard reline resin materials are commonly used to improve the fit of denture bases caused from resorption of the residual alveolar ridge. Major problem of resin is the presence of residual monomer after polymerization which had an adverse effect on the toxic and physical properties of the reline resin. Ethanol solutions in ultrasonic cleaner used been proven to reduce the residual monomer effectively in auto-polymerizing hard reline resins. The purpose of this study was to evaluate the flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of four auto-polymerizing hard reline resins [Unifast Trad (UT), Kooliner (KL), Ufi Gel Hard (UG), and Tukuso Rebase II (TR)] after ultrasonic treatment with difference ethanol concentrations. For each material, ninety specimens were prepared and divided into 9 groups: negative control (NC): no treatment, positive controls: treated by immersion in water at 50°C for 1 hour (PC1), immersion in water at 55°C for 10 minutes (PC2) and the ultrasonic group, treated by immersion in 0%, 10%, 20%, 30%, 40% or 50% ethanol solution at 55°C for 5 minutes. The flexural strength and flexural modulus were determined using a three point transverse test. After that, one fragment of each specimen was analyzed using the Vickers microhardness test. The water sorption and solubility tests were performed per ISO No.20795-1. The data were analyzed by one-way ANOVA, Tukey's test, and Dunnett's test at a 95% confidence level. The results demonstrated significant differences in the level of flexural strength, flexural modulus, water sorption, and water solubility between the groups ($p<0.05$) within each specific auto-polymerizing hard reline resin, whereas there is no significant differences in the level of surface hardness between the groups of each specific material ($p>0.05$). Comparing in four types of auto-polymerizing hard reline resin after the same treatment method showed significant differences in level of flexural strength, flexural modulus, surface hardness, water sorption, and water solubility ($p<0.05$). UT demonstrated significantly higher flexural strength, flexural modulus, surface hardness, and water sorption than the other hard reline resins, whereas TR showed significantly higher water solubility than other materials. In conclusion, for KL, the 10% ethanol solution in ultrasonic cleaner should be the treatment of choice which effectively increases the physical properties. In UG and TR, the 10% ethanol solution in ultrasonic cleaner maintained the physical properties. In UT, the 30% ethanol solution in ultrasonic cleaner had appropriate physical properties.

Department: Prosthodontics
Field of Study: Prosthodontics
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Student's Signature .....
Advisor's Signature .....

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CHAPTER I
INTRODUCTION

Research background and rationale

Hard reline resin materials have been used to improve the fit of denture bases because of resorption of the residual alveolar ridge and providing better retention and stability for removable prostheses (1). Auto-polymerizing acrylic reline resin is still one of the most commonly denture reline materials in daily prosthodontic because they can be easily completed at room temperature in a short period of time without the use of any additional equipment (2, 3). It can be divided into two main types that the composition is based on polymethyl methacrylate (PMMA) in the powder and methyl methacrylate in liquid component (MMA-based). Another type is polyethyl methacrylate (PEMA) that contained high molecular weight methacrylate monomers in the liquids (Non-MMA based). The non-MMA based hard reline resins are less irritating to the patient (1, 4). During the polymerization reaction of the hard reline resin, the conversion of monomer into polymer is not complete and varying amounts of free or unreacted monomer remain in the polymerized resin (3). This residual monomer has adverse effect on physical properties and allergic reactions in oral mucosa (3, 5, 6).

Thus, there is a need for effective post-polymerization treatment methods that decrease the residual monomer content in acrylic resin (7-9). It has been reported that a decrease in the residual monomer content could be achieved by immersion conventional acrylic resins in hot water or using microwave irradiation (7, 9-11). Currently, water can be used immersion medium of post-polymerization for reduce residual monomers (8, 9). Apart from water, ethanol solutions can be used to increase and accelerate compounds solubility, indicating the importance of the solvent in leaching processes (12, 13). Neves et al showed that higher concentrations of ethanol promoted lower residual monomer in acrylic reline resin compared to immersion in hot water (14). Other methods have been proposed to minimize residual monomer. Ultrasonic cleaners was first used to increase the efficacy of

residual monomer reduction in dental acrylic resin by Charasseanpaisarn et al (15, 16). They recommended using an ultrasonic cleaner in water 50^{O}C for 5 min to reduce residual monomer similarly to hot water, however it required less chair time (15, 16). Post-polymerization treatment in ethanol solutions by an ultrasonic cleaner also reduced the residual monomer in an auto-polymerizing hard reline resin (17). When determining the most effective post-polymerization treatment, the treatment that reduces the residual monomer content more effectively using the least time should be chosen. However, there is not research to study the effect of ethanol solutions in ultrasonic treatment the physical properties of dental polymeric resins.

Research question

Do the flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of auto-polymerizing hard denture reline resins change after reducing the residual monomer in various ethanol concentrations by an ultrasonic cleaner?

Objectives

1. To determine and compare flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of auto-polymerizing hard denture reline resins after reducing the residual monomer in various ethanol concentrations by an ultrasonic cleaner.

2. To determine and compare flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of four types of auto-polymerizing hard denture reline resin after reducing the residual monomer of the same method.

Research hypotheses

Hypothesis 1

Null hypothesis: The flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of auto-polymerizing hard denture reline resins after ultrasonic treatment with various ethanol concentrations do not significantly differ from those of the non-treatment at the 95% confidence level.

Alternative hypothesis: The flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of auto-polymerizing hard denture reline resins after ultrasonic treatment with ethanol at least one concentration will significantly differ the non-treatment at the 95% confidence level.

Hypothesis 2

Null hypothesis: The flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of auto-polymerizing hard denture reline resins after ultrasonic treatment with various ethanol concentrations do not significantly differ from those of the group of immersion in water at 50 °C for 1 hour at the 95% confidence level.

Alternative hypothesis: The flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of auto-polymerizing hard denture reline resins after ultrasonic treatment with ethanol at least one concentration will significantly differ of the immersion in water at 50 °C for 1 hour at the 95% confidence level.

Hypothesis 3

Null hypothesis: The flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of auto-polymerizing hard denture reline resins after ultrasonic treatment with various ethanol concentrations do not significantly differ from those of the group of immersion in water at 55 °C for 10 minutes at the 95% confidence level.

Alternative hypothesis: The flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of auto-polymerizing hard denture reline resins after ultrasonic treatment with ethanol at least one concentration will significantly

differ of the group of immersion in water at 55 ℃ for 10 minutes at the 95% confidence level

Hypothesis 4

Null hypothesis: Type of auto-polymerizing hard denture reline resins do not significantly affect the level of flexural strength, flexural modulus, surface hardness, water sorption, and water solubility after the same treatment methods at the 95% confidence level.

Alternative hypothesis: Type of auto-polymerizing hard denture reline resins will significantly affect the level of flexural strength, flexural modulus, surface hardness, water sorption, and water solubility after the same treatment methods at the 95% confidence level.

Scope of the Reserch

1. The research is an in vitro study
2. The four types of commercial acrylic hard denture relining material used in this study are UnifastTrad, Kooliner, Ufi Gel Hard, and Tokuso Rebase II
3. A single investigator performed this study.

Keywords

Auto-polymerizing acrylic hard denture reline resin

Flexural strength

Residual monomer

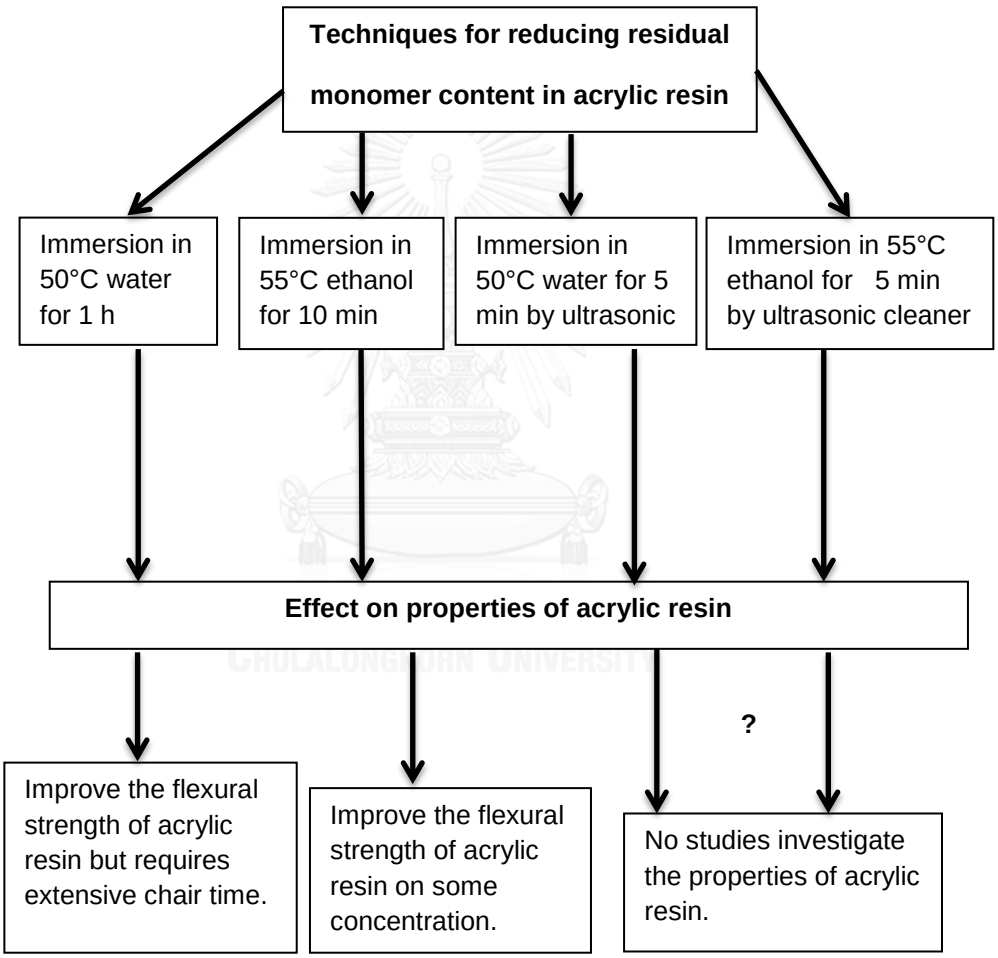
Ultrasonic cleaner

Water sorption

Expected benefit

Flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of auto-polymerizing hard denture reline resin could be improved after reducing the residual monomer in ethanol solution by an ultrasonic cleaner.

Conceptual framework



CHAPTER II

LITERATURE REVIEW

Denture base polymers

The International Standards Organization's (ISO) 20795-1:2013 document classifies denture base polymers into 5 types (18).

Type 1: Heat-polymerizable materials

Class 1: Powder and liquid

Class 2: Plastic cake

Type 2: Auto-polymerizable materials

Class 1: Powder and liquid

Class 2: Powder and liquid for pour-type resins

Type 3: Thermoplastic blank or powder

Type 4: Light-activated materials

Type 5: Microwave cured materials

Acrylic resin (2)

Acrylic resins are derivatives of ethylene and contain a vinyl (-CH=CH2) group in their structural formula: $H_2C=CHR$.

There are at least two acrylic resin series of dental interest. One series is derived from acrylic acid, $CH_2=CHCOOH$, and the other from methacrylic acid, $CH_2C(CH_3)COOH$. Both polymerize by addition. Although the poly-acids are hard and translucent, their polarity related to the carboxyl group causes them to imbibe water. Water tends to separate the chains and causes a general softening and loss of strength.

Polymethyl methacrylate, PMMA (2)

Polymethyl methacrylate (PMMA) is a transparent resin of water-like clarity. It is a hard resin with a Knoop hardness number of 18 to 20 KHN. It has a tensile strength approximately of 60 MPa, a density of 1.18 g/cm^3, and a modulus of elasticity of approximately 2.4 GPa. It is easy to handle, tough, wear-resistant, able to be pigmented to a lifelike appearance, able to be sterilized, easily cleaned, biologically safe, and very durable.

Methyl methacrylate, MMA (2)

PMMA by itself is not used in dentistry to a great extent in molding procedures. Rather, the liquid monomer MMA is mixed with the polymer which is supplied in a powdered form. The monomer partially dissolves a polymer to form a plastic dough-like material. This dough-like is packed into a mold and the monomer is polymerized.

MMA is a transparent liquid at room temperature. It has a melting point of -48 °C, a boiling point of 100.8 °C, a density of 0.945 g/mL at 20 °C, a heat of polymerization value of approximately 12.9 kcal/mol and is an excellent organic solvent. The polymerization of MMA can be initiated by visible light, ultraviolet light, heat, or chemically; most dental products are initiated chemically.

Auto-polymerizing materials

Auto-polymerizing materials are known as self-curing, cold-curing, or chemically curing. The difference from heat-curing materials are the method of activation of benzoyl peroxide and the degree of polymerization, which is not high as that of heat-polymerizing materials (19).

Auto-polymerizing materials consist of powder and liquid components, which are mixed together just like heat-polymerizing products. Mixing is followed by a gradual increase in viscosity until a dough-like stage is reached. This increase in viscosity is due to a combination of physical and chemical changes occurring in the

mix. Generally, materials reach the dough-like stage quickly and remain workable for only a short period of time. Within a few minute of attaining a dough-like consistency, the rate of polymerization increases rapidly causing a large temperature rise resulting in the material becoming hard and unmanageable. The polymerization is never as complete as that of the heat-polymerizing type (20).

The powder of the auto-polymerizing type contains beads of polymer, which have a lower molecular weight than the heat-polymerizing type. In addition, the liquid contains a chemical activator to initiate the additional polymerization resulting in the greater amount of residual monomer compared to that of the heat-polymerizing type (21). These residual monomers create major disadvantages. It serves as a potential tissue irritant. In addition, it acts as a plasticizer resulting in decreased transverse strength of the auto-polymerizing type (2). The auto-polymerizing types also have advantages such as easily fabrication for minor denture repair and chairside denture relining (22).

Requirements of auto-polymerizable polymers

The requirements of auto-polymerizable dental base polymers according to ISO 20795-1:2013 are: (18)

1. Un-polymerized material

1.1 Liquid component consists essentially of monomeric material compatible with the powder and free of deposit or sediment that can be observed by visual inspection.

1.2 Solid component shall be free of extraneous material that can be observed by visual inspection.

2. Polymerized material

2.1 Biocompatibility

2.2 Surface characteristic; specimens processed in the manner recommended by the manufacturer should have a smooth, hard, and glossy surface.

2.3 Color; the specimen shall provide a shade guide on request. The colored denture base polymer shall contain translucent pigments and fibers shall be evenly distributed.

2.4 Color stability; test specimens shall not show more than a slight change in color.

2.5 Translucency; the shadow of the illuminated opaque disc shall be visible from the opposite side of the test specimen plate.

2.6 Freedom from porosity; specimen strip shall not show voids that can be observed by visual inspection.

2.7 Flexural strength; the flexural strength shall be not less than 60 MPa when tested in water at 37±1°C.

2.8 Flexural modulus; the flexural modulus shall be at least 1500 MPa when tested in water at 37±1°C.

2.9 Residual methyl methacrylate monomer; the maximum limit for residual methyl methacrylate is 4.5% mass fraction.

2.10 Sorption; water sorption shall not exceed 32 μg/mm^3.

2.11 Solubility; water solubility shall not exceed 8.0 μg/mm^3.

Denture lining materials (1)

Denture lining materials can be divided into three groups: permanent hard reline materials, semi-permanent soft liners and tissue conditioners. Hard reline materials are used to replace the fitting surface of a denture base because of reduced resorption of the residual alveolar ridge and improved retention of the denture. The criteria for relining are:

- poor retention or stability,
- collapse of the vertical dimension of the occlusion,
- degradation of the denture base,
- lack of denture extension into mucobuccal fold areas.

The reline can be achieved either with an auto-polymerizing reline resins at the chairside, or the denture is sent to a dental laboratory for relining with a heat-

polymerizing acrylic resin. The auto-polymerizing hard denture reline resin come in two types, with constituents as listed in table 1. The reason for using the second type of reline material is that MMA can be very irritated to soft tissue and can sensitize the patient. Polyethyl methacrylate (PEMA) and butyl methacrylate are less irritating to the patient, but have the disadvantage that they cause a reduction in the glass transition temperature (T_g) which increases the possibility of dimensional instability.

Table 1 Two types of auto-polymerizing hard denture reline resin

	Type 1	Type 2
Powder	Polymethyl methacrylate Benzoyl peroxide Pigments	Polyethyl methacrylate Benzoyl peroxide Pigments
Liquid	Methyl methacrylate Di-*n*-butylphthalate Amine	Butyl methacrylate Amine

Residual monomers

For auto-polymerizing resins, a level of residual monomer as 3-5% can appear, in comparison with 0.2-0.5% for heat-polymerizing acrylic resins (22). Levels of residual monomer vary with the conditions and the methods of polymerization (5, 23).

During polymerization of acrylic resins, the conversion of monomer into polymers is not complete, and varying amounts of free or unreacted monomer remain in the polymerized resin (3). Residual monomer have an adverse effect on the physical and mechanical properties of the acrylic resins (5, 6). In addition, it can cause allergic reactions or chemical burns (3). There has been a search for effective post-polymerization treatment methods that decrease the residual monomer content (7-9). It has been reported that a decrease in the residual monomer content could be achieved by immersing conventional acrylic resins in hot water or using

microwave irradiation (7, 9, 10). Water can be used as a post-polymerization immersion medium for reducing residual monomer (9, 14). In addition, ethanol solutions can be used to increase and accelerate a compound's solubility, indicating the importance of the solvent in terms of leaking (12, 13). Neves et al showed that higher concentrations of ethanol solution resulted in a lower residual monomer content in acrylic reline resin (14).

Ethanol

Ethanol (CH_3CH_2OH) is an alcohol, a group of chemical compounds whose molecules contain a hydroxyl group (OH), bonded to a carbon atom (24). It has a molar mass of 46.07 g mol^{-1}, a melting point of −114.1°C, a boiling point of 78.5°C, and a density of 0.789 g/ml at 20 °C (24). Ethanol is soluble in water. This is due to the hydroxyl group in the alcohol which is able to form a hydrogen bond with a water molecule (25). Ethanol is a clear, colorless liquid with a wine-like odor. It is used in alcoholic beverages, as a solvent, and in making other chemicals (25). In dentistry, ethanol can improve the biocompatibility of resins (14). In additional, ethanol solutions can be used to reduce residual monomer. Neves showed that higher concentrations of ethanol resulted in a lower residual monomer content in acrylic reline resins (14). Other studies found that immersion in ethanol reduced the amount of residual compounds in acrylic polymers as denture base resins (12) and temporary restorative resins (13).

Ultrasonic cleaner

Ultrasonic waves are pressure waves above the limits of human audibility, with a frequency above 20 kHz. Ultrasonic are used extensively in industry (e.g. underwater acoustics (SONAR), cleaning and for medical purposes such as medical imaging (ultrasound) (26).

In dentistry, ultrasonic cleaning has been recommended because it removes a variety of contaminants, reduces the direct handling of instruments and has superior cleaning ability compared with other cleaning techniques (27, 28).

The principle of ultrasonic cleaning is based on the cavitation effect caused by high-frequency ultrasonic wave vibration in the fluid. Microscopic bubbles are formed which grow and eventually implode. These bubbles are generated when the negative pressure during the rarefaction phase of the sound wave is sufficiently large to disrupt the liquid. The implosion of the bubbles can locally produce extreme temperatures and pressures. These hot-spots can lead to irreversible changes. The cavitation creates an intense scrubbing action on the surface of the item being cleaned. The majority of the ultrasonic cleaning that is done in industrial applications uses a frequency of 40 kHz. Ultrasonic treatment can also reduce residual monomer in acrylic temporary restorations because it increases the flow rate of water in the tank, which may affect the elution of the residual monomer into the environment. In addition, implosive bubbles release energy to the surface of the specimens and may cause polymerization of the remaining monomer (15).

Flexural strength (19)

Flexural strength is an important mechanical property of acrylic resins. The flexural strength test is especially useful in comparing denture base materials in which a stress is applied to a specimen of denture acrylic with masticatory loads. This test determines the strength of the material and indicated amount of distortion expected.

The flexural strength is calculated from the following equation:

$$\text{Flexural strength (FS)} = \frac{3 * F * L}{2 * b * h^2}$$

Where F is the maximum load (N)

L is the distance between the supports (mm)

b is the width of the specimen (mm)

h is the height of the specimen (mm)

$$\text{Flexural modulus (E)} = \frac{F1 * L^3}{4 * b * h^3 * d}$$

Where F1 is the load (N) at a convenient point in the straight-line portion of the trace

d is the deflection at load F1 (mm)

Several investigators studied the relationship between residual monomer and mechanical properties of acrylic resin (5, 6, 23). They agreed that a high amount of residual monomer adversely affects the mechanical properties of the acrylic resin. Dogan concluded that entrapped residual monomer in a polymer matrix will leave voids, which increases porosity and this affects the mechanical properties (23).

Surface hardness

Hardness is a property used to predict the wear resistance of the material (2). The properties that are related to the hardness of materials are strength, proportional limit, and ductility to permanent surface indentation (19). There are several types of surface hardness tests. Most are based on the ability of the surface of a material to resist penetration by a diamond point or steel ball under a specified load. The choice of a hardness test depends on the material of interest, the expected hardness range and the desired degree of localization. Common methods used for hardness evaluation include Vickers, Knoop, Brinell, and Rockwell (2).

The Vickers hardness test used a pyramid-shaped diamond with a square base that it is suitable for testing the surface hardness of dental materials. The Vickers test is useful in measuring the hardness of small areas and for very hard or brittle materials (2, 29). Hardness has been found to be sensitive to the residual monomer content in the polymerized resin, and is a simple and effective way to assess the degree of conversion of dental polymers (30).

Lee showed that provisional PMMA resins cured in hot water reduced residual MMA elution and increased the microhardness values (31).

Water sorption and solubility

PMMA slowly absorbs water, due to the polar nature of the resin molecules (1). This water sorption is typically of the order of 1.0-2.0% by weight. In practice, this helps to compensate for the slight processing shrinkage (1). However, water sorption is related to the dimensional stability and long-term durability. When the material takes up water, its dimension and structural integrity may be affected (2). The water solubility of acrylic monomer is related to the water sorption (19). The solubility of the acrylic resin represents the amount of water-soluble ingredients, unreacted monomers, plasticizers, and initiators that leaked out (32). In general, acrylic resins have a low solubility, which is a result of the leaking out of traces of unreacted monomers and water-soluble additives into the oral fluids (19).

There is a relation between the level of the residual monomer and the percentage water absorption because the solubility of acrylic resins is the result of the leakage of unreacted monomer (32). Dogan et al reported a correlation between the amount of residual monomer and water absorption (23). In additional, Takahashi et al showed that the mechanical properties of denture base materials decreased if the solubility increased (33).

CHAPTER III

METHODOLOGY

Material and instruments use in this study

1. Four types of auto-polymerizing hard reline resins:
   - Unifast Trad (GC Corporation, Tokyo, Japan)
   - Kooliner (GC America Inc, Alsip, IL, USA)
   - Ufi Gel Hard (Voco, Cuxhaven, Germany)
   - Tokuso Rebase II (Tokuyama Dental Corporation, Tsukuba, Japan)
2. Ethanol (EMD Millipore Corporation, Billerica, USA)
3. Vaseline
4. Distilled water
5. Metallographic grinding paper No.500 (TOA Corporation, Samut Prakarn, Thailand)
6. Polishing machine (Pace Technologies, USA)
7. Universal testing machine (SHIMADZU EZ-X, Kyoto, Japan)
8. Microhardness testing machine (FM-810, Future-Tech, Japan)
9. Stainless steel mould according to ISO 20795-1:2013
10. Ultrasonic cleaner (GT-SONIC, China)
11. Racks for holding specimens
12. Desiccator with silica gel
13. Thermometer
14. Digital vernier caliper (Mitutoyo, Japan)
15. Analytical balance (Precisa, Switzerland)
16. Incubator (Contherm Scientific, New Zealand)
17. Volumetric pipettes (Labnet, USA)

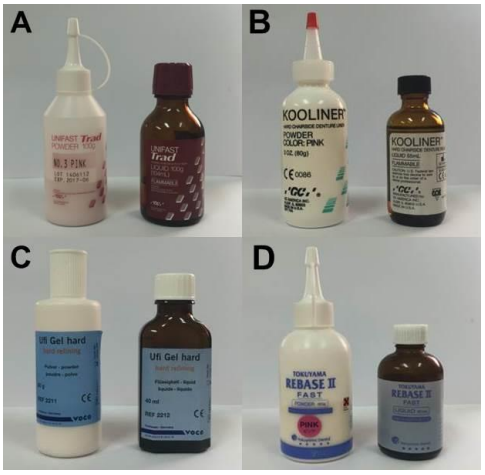


Figure 1 Auto-polymerizing hard reline resins

A UnifastTrad, **B** Kooliner, **C** Ufi Gel Hard, **D** Tokuso Rebase II.

Figure 2 The polishing machine (Pace Technologies, USA)

Figure 3 The analytical balance (Precisa, Switzerland)

Figure 4 Ultrasonic cleaner (GT-SONIC, China)

Figure 5 Mould for flexural strength and modulus tests: stainless steel mould with 64 mm in length, 10 mm in width, 3.3 mm in height, ISO 20795-1

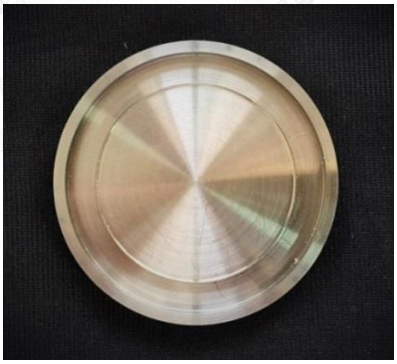


Figure 6 Mould for water sorption and solubility tests: stainless steel mould with 50 mm in diameter and 0.5 mm in thickness, ISO 20795-1

Table 2 Auto-polymerizing acrylic resins used in this study

Product	Code	Manufacturer	Powder/ liquid ratio	Polymerization condition	Composition
Unifast Trad	UT	GC Corporation, Tokyo, Japan	2.0 g 1 ml	5 min at room temperature	P: PMMA L: MMA
Kooliner	KL	GC America Inc, Alsip, IL, USA	1.4 g 1 ml	10 min at room temperature	P: PEMA L: IBMA
Ufi Gel Hard	UG	Voco, Cuxhaven, Germany	1.8 g 1 ml	7 min at room temperature	P: PEMA L: 1,6-HDMA
Tokuso Rebase II	TR	Tokuyama Dental Corp, Tsukuba, Japan	2.0 g 1 ml	5.5 min at room temperature	P: PEMA L: MAOP, 1,6-HDMA

P: powder; L: liquid; PMMA, polymethyl methacrylate; MMA, methyl methacrylate; PEMA, polyethyl methacrylate; IBMA, isobutyl methacrylate; 1,6-HDMA, 1,6-hexanediol dimethacrylate; MAOP, ß-methacryloyl oxyethyl propionate.

Four auto-polymerizing hard reline resins were selected for comparison (Table 2). Unifast Trad (UT) was selected as representative of a MMA-based hard reline resin which is commonly used for relining of denture bases. Tokuso Rebase II (TR), Ufi Gel Hard (UG), and Kooliner (KL) were representative of a non MMA-based hard reline resins. TR and UG contain high percentages of cross-linking agents. The liquid composition of KL is isobutyl methacrylate (IBMA) without a cross-linking agent.

Part 1: Preparation of specimens for flexural strength and modulus test

A total of 360 specimens (90 specimens per type of auto-polymerizing hard reline resin) were fabricated in a stainless steel mould of dimension 64 × 10 × 3.3 mm according to ISO 20795-1 (2013). Each material was proportioned and manipulated following the manufacturer's instructions and packed within the mould. The specimens were finished with a 500-grit silicon carbide paper (TOA, Thailand) to remove irregularities. The accuracy of the dimension was verified with a digital vernier caliper, at three locations of each dimension to within 0.2 mm tolerance.

All specimens were divided into 9 groups (n= 10 per group): negative control (NC): no treatment, positive controls (PC): treated by immersion in water at 50°C for 60 minutes (PC1), immersion in water at 55°C for 10 minutes (PC2) and the ultrasonic group, treated by immersion in 0%, 10%, 20%, 30%, 40% or 50% ethanol (E) solution while undergoing 40 kHz ultrasonic cleaner at 55°C for 5 minutes. All specimens were stored in water at 37±1°C for 48 h before testing.

Flexural strength and modulus test

All the specimens were subjected to the flexural strength test in a Universal testing machine (Shimadzu EZ-S, Japan) using three-point loading. A crosshead speed of 5±1 mm/min was used. The specimen was laid on the support of the flexural test rig. The distance between the center of the supports was 50±1 mm. The loading plunger was placed midway (±0.1 mm) between the support. The force was increased uniformly on the loading plunger until the specimen was fractured, and the fracture load was recorded in Newton (N). The flexural strength (FS, MPa) was calculated using the formula:

$$\text{Flexural strength (FS)} = \frac{3 * F * L}{2 * b * h^2}$$

Where F = the maximum load (N)

L = the distance between the supports (mm)

b = the width of the specimen (mm)

h = the height of the specimen (mm)

$$\text{Flexural modulus (E)} = \frac{F1 * L^3}{4 * b * h^3 * d}$$

Where F1= the load (N) at a convenient point in the straight-line portion of the trace

d = the deflection at load F1 (mm)

Figure 7 Specimens strip for flexural strength and modulus tests

(64±0.2 mm x 10±0.2 mm x 3.3±0.2 mm)

Figure 8 The universal testing machine (SHIMADZU EZ-X, Kyoto, Japan)

Part 2: Surface hardness test

After flexural strength test, one fragment of each specimen was selected for microhardness test by the technique described by ISO 6507-1(E): Vickers hardness test (2005) (34). The specimens were dried and tested with a microhardness testing machine with a Vickers pyramid diamond indenter and a square base with a 136° angle at vertex. A force of 50 g was applied in a direction perpendicular to the specimen surface, without shock or vibration for 15 s at a speed of 50 μm/sec. The specimens were indented at five positions/specimen at room temperature (23±2°C).

After unloading, the diagonals of indentation were measured using a microscope and the results were analyzed using the FT_ARS program for automatic hardness testing systems.

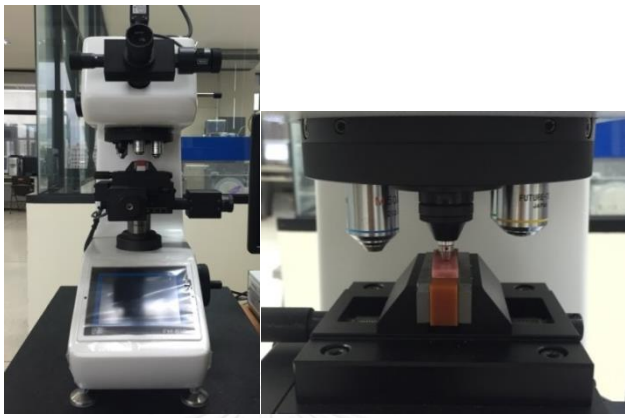


Figure 9 The microhardness testing machine (FM-810, Future-Tech, Japan)

Part 3: Preparation of specimens for water sorption and solubility test

A total of 360 specimens (90 specimens per type of auto-polymerizing hard reline resin) were fabricated in a stainless steel mould 50 mm in diameter and 0.5 mm in thickness according to ISO 20795-1 (2013). Each material was proportioned and manipulated following the manufacturer's instructions and packed within the mould. The specimens were finished with a 500-grit silicon carbide paper (TOA, Thailand) to remove irregularities. The accuracy of the dimension was verified with a digital vernier caliper, at three locations of each dimension to within 1 mm tolerance in diameter and 0.1 mm tolerance in thickness.

All specimens were divided into 9 groups (n= 10) and prepared using the same procedure as in Part 1.

Water sorption and solubility test

Conditioned specimens

The specimens were stored for 23±1 h in a rack inside the desiccator, which was placed in an oven at 37±1 °C. After that, they were left at ambient temperature

for 60±10 min. Each specimen was weighed using an electric balance (Precisa, Switzerland) to an accuracy of ±0.2 mg. The desiccator was kept sealed except for the shortest possible period required for removing and replacing specimens. After weighing all the specimens, the silica gel in the desiccator was replaced with freshly dried one.

The weighing procedure was repeated for each specimen until a constant mass m_1, called the "conditioned mass", was reached i.e. when the difference in weight between each specimen was not more than 0.2 mg. At this point, the volume *V* of each specimen was calculated using the mean of three diameter measurements and the mean of five thickness measurements.

$$V = \pi * r^2 * h$$

Where V = the volume (N)

r = the radius of the specimen (mm)

h = the height of the specimen (mm)

Wet specimens

The conditioned specimens were immersed in water at 37±1 ℃ for 7 days ± 2 h. After that, the discs were removed from water, wiped with a clean dry towel until they were free from visible moisture, waved in the air for 15±1 s and weighed within 60±10 s after removal from water. The mass was recorded as m_2.

Reconditioned specimens

After weighing the wet specimens, the specimens were reconditioned to a constant mass in the desiccator. The mass of the "reconditioned" specimens was recorded as m_3.

Calculation of water sorption and water solubility

Water sorption (Wsp)

$$Wsp = \frac{m_2 - m_3}{V} \qquad (\mu g/mm^3)$$

Where m_2 = the mass of the specimens after immersion in water (μg)

m_3 = the reconditioned mass of the specimens (μg)

V = the volume of the specimens (mm^3)

Water solubility (Wsl)

$$Wsl = \frac{m_1 - m_3}{V} \qquad (\mu g/mm^3)$$

Where m_1 = the conditioned mass of the specimens (μg)

m_3 = the reconditioned mass of the specimens (μg)

V = the volume of the specimens (mm^3)

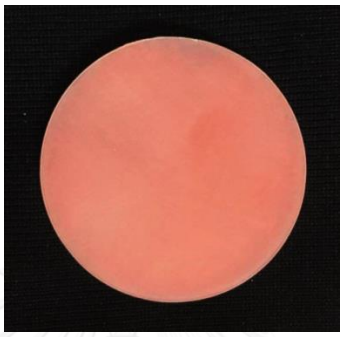


Figure 10 Specimens disc for water sorption and solubility tests (50.0±1.0 mm in diameter and 0.5±0.1 mm in thickness)

Figure 11 Desiccator with specimen disc

Statistical analysis

The mean and standard deviation of each group are calculated. The one-sample Kolmogorov-Smirnov test is performed to assess the normal distribution and Levene's test is performed to assess the variance. The data were analyzed using one-way analysis of variance (ANOVA) followed by Tukey's HSD test at a 95% confidence level to determine statistically significant differences in levels of flexural strength, flexural modulus, surface hardness, water sorption, and water solubility among groups of each auto-polymerizing hard reline resins. One-way ANOVA followed by Dunnett T3 was used to compare statistically significant differences in levels of flexural strength, flexural modulus, surface hardness, water sorption, and water solubility in four auto-polymerizing hard reline resins after reducing residual monomer by the same method. All statistics were performed with SPSS version 19.0 for Windows (SPSS Inc, USA).

CHAPTER IV
RESULTS

The values of flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of each auto-polymerizing acrylic hard denture reline materials are shown in Table 3-6, respectively.

Table 3 Mean and standard deviation of flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of Unifast Trad

Groups	FS (MPa)	FM (MPa)	SH* (VHN)	Wsp ($\mu g/mm^3$)	Wsl ($\mu g/mm^3$)
1) NC	58.72 ± 4.14^{AB}	$1,496.39\pm66.63^{A}$	15.10 ± 0.11	15.07 ± 0.36^{B}	0.86 ± 0.36^{C}
2) PC1	56.11 ± 3.74^{B}	$1,544.75\pm58.37^{A}$	14.95 ± 0.28	15.44 ± 0.51^{B}	0.76 ± 0.29^{C}
3) PC2	59.40 ± 2.54^{AB}	$1,566.88\pm42.07^{A}$	15.16 ± 0.14	15.05 ± 0.47^{B}	0.91 ± 0.34^{C}
4) 0%E	61.28 ± 1.95^{A}	$1,525.75\pm32.64^{A}$	15.02 ± 0.20	15.01 ± 0.43^{B}	0.82 ± 0.35^{C}
5) 10%E	61.18 ± 3.09^{A}	$1,494.03\pm54.49^{A}$	15.01 ± 0.25	15.44 ± 0.38^{B}	0.70 ± 0.18^{C}
6) 20%E	60.60 ± 3.83^{A}	$1,486.64\pm62.15^{A}$	15.01 ± 0.12	15.03 ± 0.40^{B}	0.70 ± 0.35^{C}
7) 30%E	60.63 ± 3.33^{A}	$1,568.49\pm96.60^{A}$	15.01 ± 0.11	15.29 ± 0.41^{B}	0.78 ± 0.18^{C}
8) 40%E	57.25 ± 2.72^{AB}	$1,517.81\pm40.68^{A}$	14.90 ± 0.16	16.04 ± 0.33^{A}	1.42 ± 0.44^{B}
9) 50%E	49.70 ± 2.27^{C}	$1,351.46\pm58.25^{B}$	15.03 ± 0.23	16.36 ± 0.35^{A}	2.37 ± 0.45^{A}

Same superscript letter in the same column indicates no significant difference of the property between the groups ($p>0.05$). * No significant differences between the groups in column.

Table 4 Mean and standard deviation of flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of Kooliner

Groups	FS (MPa)	FM (MPa)	SH* (VHN)	Wsp ($\mu g/mm^3$)	Wsl ($\mu g/mm^3$)
1) NC	33.59 ± 1.03^{C}	535.31 ± 48.71^{D}	9.52±0.14	15.66 ± 1.58^{A}	3.55 ± 0.95^{A}
2) PC1	40.02 ± 1.19^{A}	759.62 ± 72.19^{A}	9.49±0.22	15.89 ± 1.14^{A}	3.49 ± 0.55^{A}
3) PC2	38.28 ± 1.82^{B}	662.00 ± 50.99^{C}	9.60±0.21	15.26 ± 0.94^{A}	4.31 ± 0.72^{A}
4) 0%E	37.45 ± 0.84^{B}	680.16 ± 56.09^{BC}	9.68±0.21	16.73 ± 1.35^{A}	4.20 ± 0.87^{A}
5) 10%E	41.20 ± 1.17^{A}	761.17 ± 55.08^{A}	9.69±0.20	16.39 ± 0.89^{A}	4.38 ± 0.90^{A}
6) 20%E	40.90 ± 1.08^{A}	745.66 ± 60.37^{AB}	9.71±0.18	16.24 ± 0.80^{A}	4.10 ± 0.94^{A}
7) 30%E	37.55 ± 1.38^{B}	623.25 ± 47.26^{C}	9.72±0.15	12.41 ± 0.84^{B}	1.46 ± 0.47^{B}
8) 40%E	37.22 ± 0.71^{B}	616.79 ± 42.94^{C}	9.60±0.21	12.97 ± 1.26^{B}	1.35 ± 0.64^{B}
9) 50%E	35.06 ± 0.65^{C}	626.04 ± 56.93^{C}	9.56±0.25	12.76 ± 0.81^{B}	1.32 ± 0.43^{B}

Same superscript letter in the same column indicates no significant difference of the property between the groups ($p>0.05$). * No significant differences between the groups in column.

Table 5 Mean and standard deviation flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of Ufi Gel Hard

Groups	FS (MPa)	FM (MPa)	SH* (VHN)	Wsp ($\mu g/mm^3$)	Wsl ($\mu g/mm^3$)
1) NC	38.20 ± 4.02^{AB}	$1,385.83\pm79.37^{AB}$	13.56±0.22	9.85 ± 0.36^{AB}	4.09 ± 0.71^{A}
2) PC1	35.36 ± 4.23^{BC}	$1,358.57\pm56.47^{AB}$	13.59±0.28	10.38 ± 0.43^{A}	4.08 ± 1.07^{A}
3) PC2	37.53 ± 2.91^{ABC}	$1,364.97\pm72.95^{AB}$	13.43+0.12	10.09 ± 0.68^{A}	4.17 ± 1.04^{A}
4) 0%E	38.57 ± 2.56^{AB}	$1,419.07\pm56.66^{A}$	13.58+0.15	9.24 ± 0.54^{B}	3.21 ± 0.34^{AB}
5) 10%E	40.95 ± 3.71^{A}	$1,425.70\pm69.57^{A}$	13.41+0.22	9.23 ± 0.57^{B}	2.98 ± 1.05^{AB}
6) 20%E	38.41 ± 2.15^{AB}	$1,418.46\pm59.34^{A}$	13.35±0.19	9.23 ± 0.48^{B}	3.04 ± 1.21^{AB}
7) 30%E	37.44 ± 2.21^{ABC}	$1,419.60\pm57.64^{A}$	13.56+0.26	9.24 ± 0.37^{B}	2.77 ± 1.33^{AB}
8) 40%E	32.89 ± 3.43^{C}	$1,318.58\pm70.08^{B}$	13.40±0.20	9.73 ± 0.46^{AB}	2.54 ± 1.29^{B}
9) 50%E	32.83 ± 4.13^{C}	$1,307.52\pm58.03^{B}$	13.35±0.19	9.95 ± 0.63^{AB}	2.50 ± 0.64^{B}

Same superscript letter in the same column indicates no significant difference of the property between the groups ($p>0.05$). * No significant differences between the groups in column.

Table 6 Mean and standard deviation of flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of Tokuso Rebase II

Groups	FS (MPa)	FM (MPa)	SH* (VHN)	Wsp ($\mu g/mm^3$)	Wsl ($\mu g/mm^3$)
1) NC	46.72 ± 1.06^{AB}	718.89 ± 34.57^{AB}	10.09 ± 0.17	13.84 ± 1.71^{AB}	7.87 ± 1.37^{A}
2) PC1	44.95 ± 0.72^{BC}	694.71 ± 28.74^{AB}	9.99 ± 0.11	14.37 ± 1.69^{A}	8.37 ± 1.56^{A}
3) PC2	45.82 ± 1.22^{BC}	704.23 ± 31.84^{AB}	10.03 ± 0.14	13.51 ± 1.44^{AB}	6.97 ± 1.13^{A}
4) 0%E	44.58 ± 1.05^{C}	683.93 ± 25.81^{B}	10.14 ± 0.12	13.67 ± 0.92^{AB}	6.68 ± 1.66^{A}
5) 10%E	48.18 ± 1.08^{A}	739.48 ± 54.07^{A}	9.97 ± 0.10	13.55 ± 1.83^{AB}	6.97 ± 1.47^{A}
6) 20%E	47.78 ± 1.27^{A}	727.73 ± 43.85^{AB}	10.06 ± 0.06	13.74 ± 1.32^{AB}	6.90 ± 1.59^{A}
7) 30%E	45.79 ± 1.40^{BC}	699.96 ± 45.65^{AB}	10.06 ± 0.18	13.74 ± 1.34^{AB}	6.54 ± 1.16^{A}
8) 40%E	45.08 ± 1.14^{BC}	694.95 ± 26.11^{AB}	10.07 ± 0.13	12.15 ± 0.75^{B}	2.97 ± 1.18^{B}
9) 50%E	44.66 ± 2.01^{C}	696.77 ± 40.85^{AB}	9.97 ± 0.18	12.04 ± 1.06^{B}	2.00 ± 0.54^{B}

Same superscript letter in the same column indicates no significant difference of the property between the groups ($p>0.05$). * No significant differences between the groups in column.

The values of flexural strength, flexural modulus, surface hardness, water sorption, and water solubility in four types of auto-polymerizing hard denture reline resin after reducing residual monomers by the same method are shown in Figure 12-16, respectively.

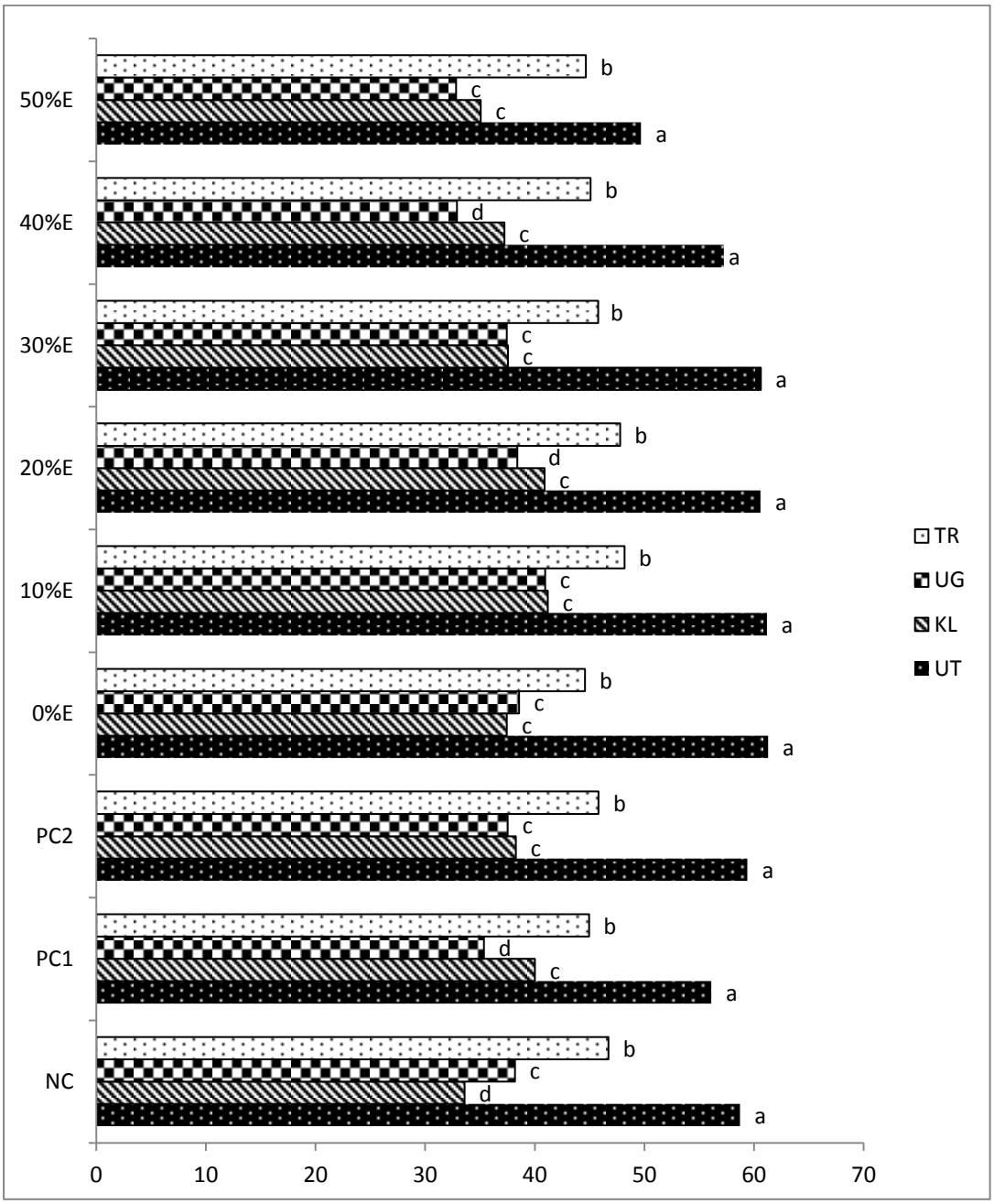


Figure 12 Mean of flexural strength in four types of auto-polymerizing hard denture reline resin (MPa). Same lowercase letter indicates no significant difference of the property between types of reline after treatment by the same method ($p>0.05$).

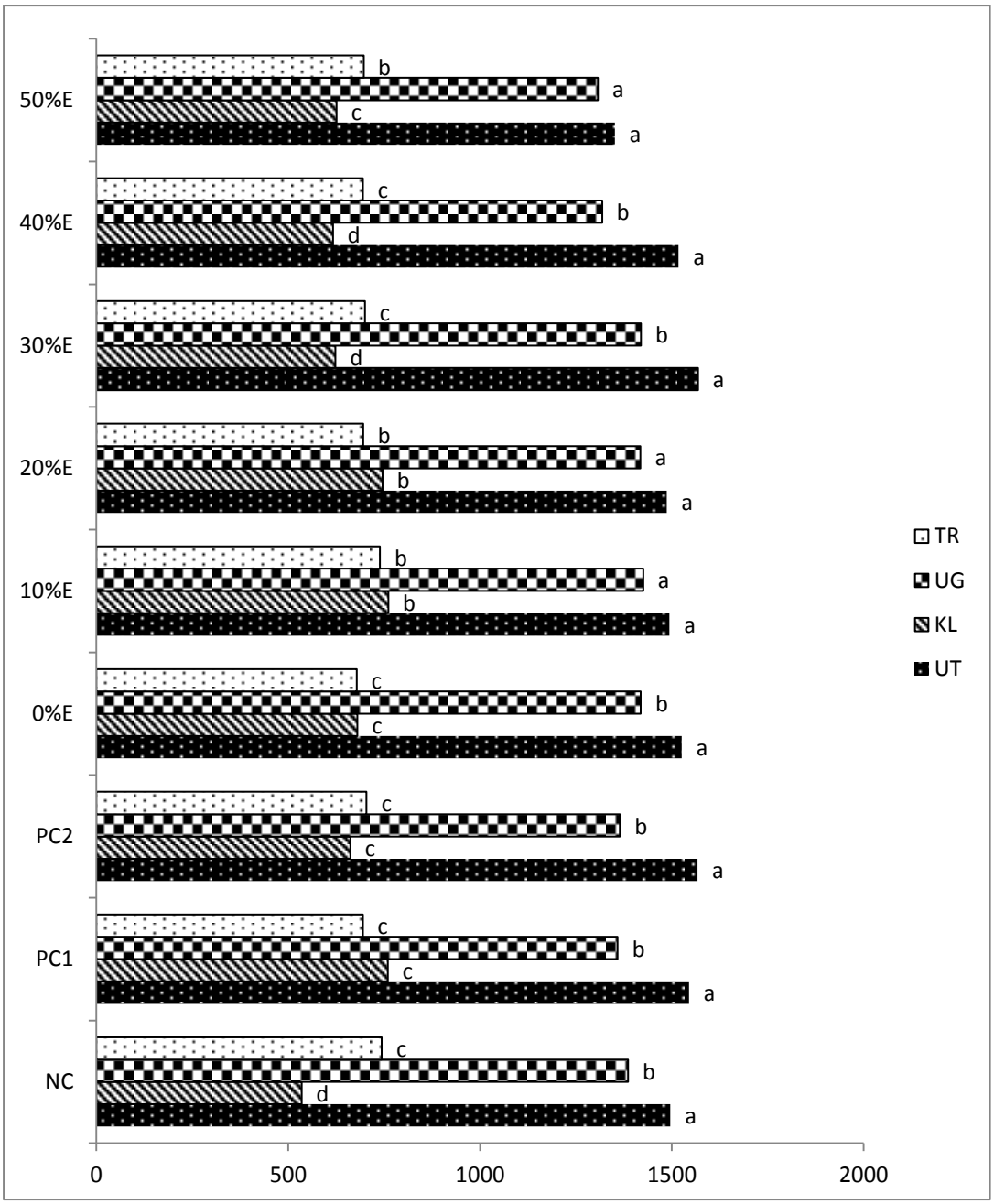


Figure 13 Mean of flexural modulus in four types of auto-polymerizing hard denture reline resin (MPa). Same lowercase letter indicates no significant difference of the property between types of reline after treatment by the same method ($p>0.05$).

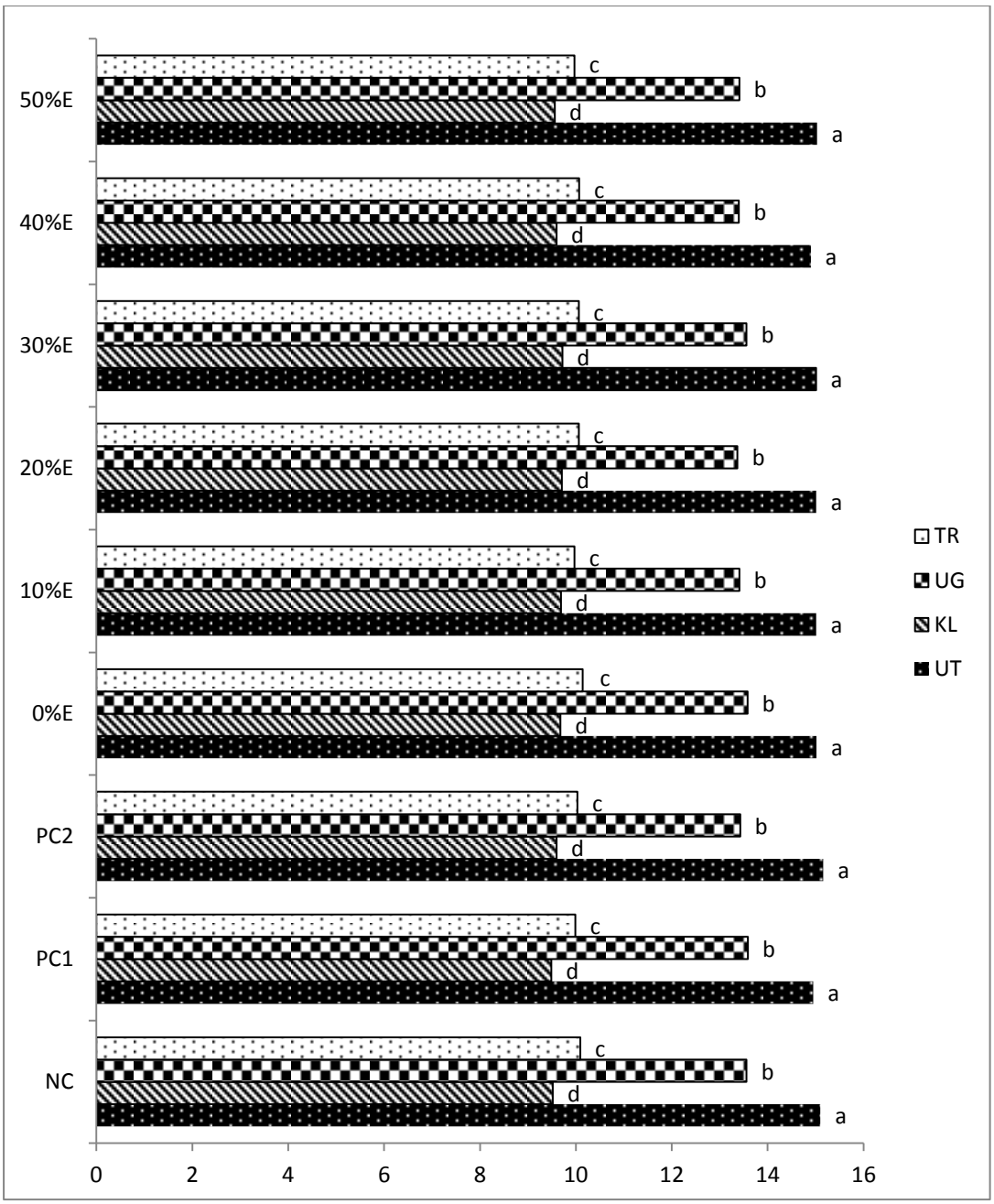


Figure 14 Mean of surface hardness in four types of auto-polymerizing hard denture reline resin (VHN). Same lowercase letter indicates no significant difference of the property between types of reline after treatment by the same method ($p>0.05$).

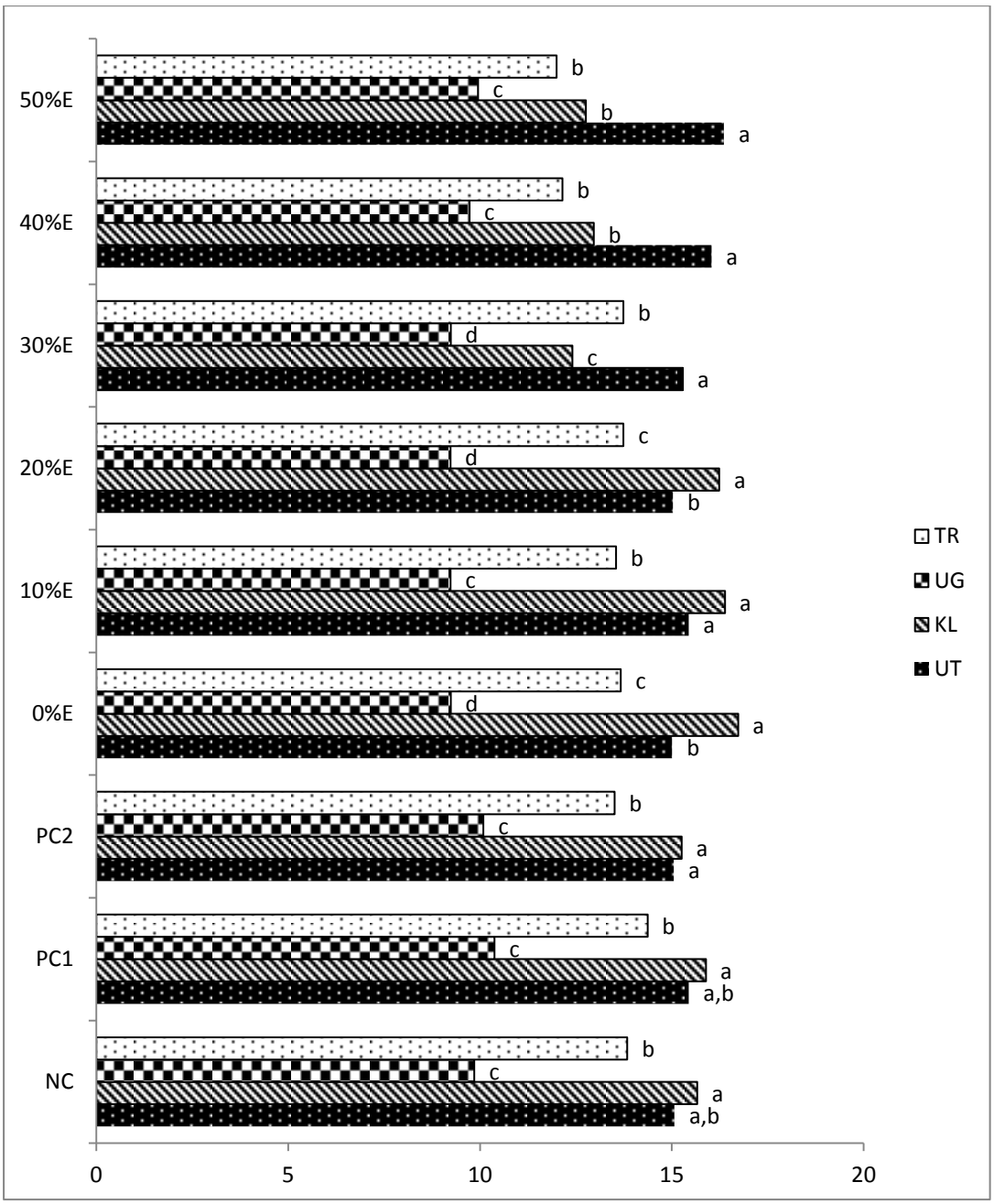


Figure 15 Mean of water sorption in four types of auto-polymerizing hard denture reline resin ($\mu g/mm^3$). Same lowercase letter indicates no significant difference of the property between types of reline after treatment by the same method ($p>0.05$).

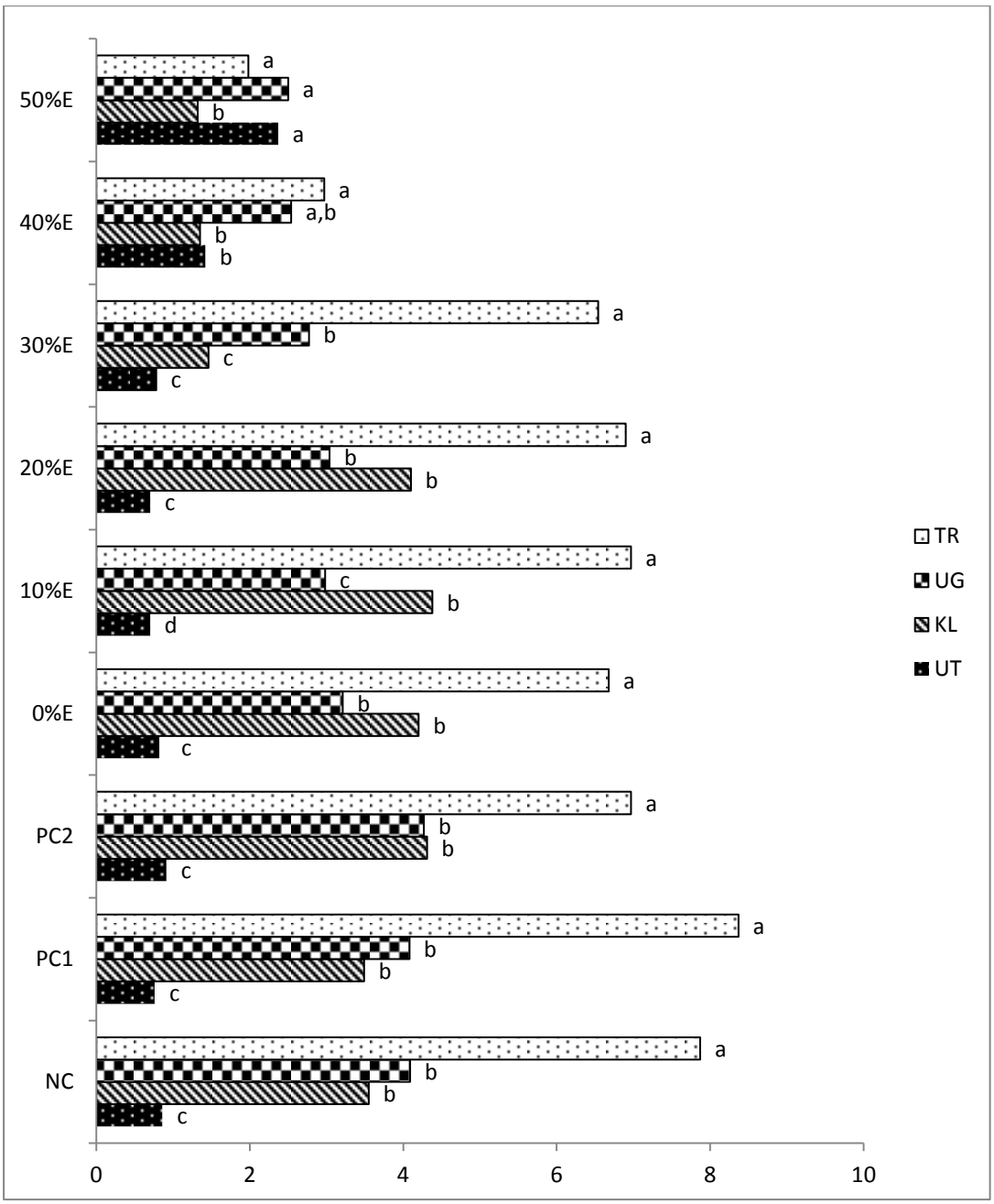


Figure 16 Mean of water solubility in four types of auto-polymerizing hard denture reline resin (μg/mm^3). Same lowercase letter indicates no significant difference of the property between types of reline after treatment by the same method ($p>0.05$).

The data of each group was analyzed using the one-sample Kolmogorov-Smirnov test to determine data distribution and Levene's test for homogeneity of variance. The resulted showed the data were normal distribution in all group and equal variance ($p>0.05$) as shown in Table 7 and 8 (Appendix).

Then data of each auto-polymerizing acrylic hard denture reline materials were separated analyzed by One-way ANOVA to compare the level of flexural strength, flexural modulus, surface hardness, water sorption, and water solubility. The results showed significant differences in the level of flexural strength, flexural modulus, water sorption, and water solubility between the groups ($p<0.05$) of each auto-polymerizing acrylic hard denture reline materials whereas no significant differences in the level of surface hardness between the groups ($p>0.05$) of each auto-polymerizing acrylic hard denture reline materials as shown in Table 9 (Appendix), multiple comparison of each auto-polymerizing acrylic hard denture reline material was analyzed by Tukey HSD to identify the differences between the groups as shown in Table 10 (Appendix).

One-way ANOVA analysis was used to compare level of flexural strength, flexural modulus, surface hardness, water sorption, and water solubility in four types of auto-polymerizing hard denture reline resin after reducing residual monomers by the same method. The results showed significant differences in the level of flexural strength, flexural modulus, surface hardness, water sorption, and water solubility between the type ($p<0.05$) of auto-polymerizing acrylic hard denture reline material as shown in Table 11 (Appendix), multiple comparison was analyzed by Dunnett T3 to identify the differences between the type of auto-polymerizing acrylic hard denture reline material as shown in Table 12 (Appendix).

Flexural strength and modulus

For UT material (Table 3), almost treatment groups presented no significant less flexural strength and modulus than the NC and PC2 group ($p>0.05$) except the 50%E group. Comparing to PC1, the groups of ultrasonic treatment in 0%E, 10%E, 20%E, and 30%E had a significantly higher flexural strength ($p<0.05$); the 40%E group

had no differences ($p>0.05$), and the 50%E group had lower values flexural strength ($p<0.05$).

For KL material (Table 4), almost treatment groups showed significant higher flexural strength than NC group ($p<0.05$) except the 50%E group. Compared with the PC1 group, the groups of ultrasonic treatment in 0%E, 30%E, 40%E, and 50%E had a significantly lower flexural strength ($p<0.05$); the 10%E and 20%E groups had no differences ($p>0.05$). Both 10%E and 20%E had a significant higher values of flexural strength and modulus than the PC2 group ($p<0.05$). For flexural modulus, all treatment groups revealed significant higher flexural modulus than NC group ($p<0.05$). Comparing to PC1, the groups of ultrasonic treatment in 0%E, 30%E, 40%E, and 50%E had a significantly lower flexural strength ($p<0.05$); the 10%E and 20%E group had no differences ($p>0.05$).

For UG material (Table 5), almost treatment groups exhibited no significant less flexural strength with the NC group ($p>0.05$) except the 40%E and 50%E groups. Comparing to PC1, the groups of ultrasonic treatment in 10%E had a significantly higher flexural strength ($p<0.05$); the 0%E, 20%E, 30%E, 40%E, and 50%E group had no differences ($p>0.05$). All treatment groups presented no significant flexural strength with the PC2 group ($p>0.05$). The flexural modulus, the groups of ultrasonic treatment presented no significant differences compared with NC, PC1, and PC2 groups ($p>0.05$). The 40%E and 50%E groups had a significantly lower flexural modulus compared with the 0%E, 10%E, 20%E, and 30%E groups ($p<0.05$).

For TR material (Table 6), almost treatment groups revealed no significant less flexural strength with the NC group ($p>0.05$) except the 0%E and 50%E groups. Ultrasonic treatment groups presented no significant more flexural strength compared the PC1 and PC2 groups ($p>0.05$) except the 10%E and 20%E groups. Considering flexural modulus, all treatment groups presented no significant in flexural modulus with NC group ($p>0.05$). The 10%E had a significant higher than 0%E group ($p<0.05$).

Comparing of four auto-polymerizing hard reline resins found that all experimental groups of UT presented significant higher values of flexural strength

than the other auto-polymerizing hard reline resins after reducing residual monomers by the same method ($p<0.05$). In NC group showed indicated UT higher significant differences in flexural strength than TR, UG, and KL, respectively ($p<0.05$). Considering PC1, 20%E, and 40%E groups indicated UT higher significant differences in flexural strength than TR, KL, and UG, respectively ($p<0.05$). However, the PC2, 0%E, 10%E, and 30%E, and 50%E groups presented no significant difference in flexural strength between KL and UG ($p>0.05$) as shown in figure 12. Flexural modulus, the NC, 30%E, and 40%E groups of UT presented significant higher flexural modulus than the UG, TR, and KL, respectively ($p<0.05$). The PC1, PC2, and 0%E groups showed no significant difference in flexural modulus between KL and TR ($p>0.05$). The 10%E and 20%E groups of UT and UG showed significant higher flexural modulus than KL and TR ($p<0.05$). The 50%E group of UT and UG presented significant higher flexural modulus than TR and KL, respectively ($p<0.05$) as shown in figure 13.

Surface hardness

All specimens of each auto-polymerizing hard reline resins no significant differences in surface hardness between the groups of each auto-polymerizing acrylic hard denture reline materials as shown in table 3-6. Considering the same method of reducing residual monomer between auto-polymerizing hard reline resins showed significant differences in the level of surface hardness. UT material showed significant higher values of surface hardness than UG, TR and KL, respectively as shown in figure 14.

Water sorption and solubility

For UT material (Table 3), almost treatment groups showed no significant more water sorption and solubility than the NC group ($p>0.05$) except 40%E and 50%E groups. Comparing to PC1 and PC2, the group of ultrasonic treatment in 0%E, 10%E, 20%E, and 30%E presented no significant of water sorption and solubility ($p>0.05$); the 40%E and 50%E groups had higher values of water sorption and solubility ($p<0.05$). The 50%E group showed significant more water solubility than 40%E group ($p<0.05$).

For KL material (Table 4), the 30%E, 40%E, and 50%E groups exhibited significant lower water sorption and solubility than those of the other groups ($p<0.05$). The 0%E, 10%E, and 20%E had no significant water sorption and solubility than the NC, PC1, and PC2 groups ($p>0.05$).

For UG material (Table 5), all treatment groups showed no significant different water sorption with the NC group ($p>0.05$). The 0%E, 10%E, 20%E, and 30%E groups presented significant lower water sorption than the PC1 and PC2 groups ($p<0.05$). The water solubility, the 40%E and 50%E groups displayed significant lower values than NC, PC1, and PC2 groups ($p<0.05$). The 0%E, 10%E, 20%E, and 30%E groups exhibited no significant water solubility compare the NC, PC1, and PC2 groups ($p<0.05$).

For TR material (Table 6), all treatment groups exhibited no significant different water sorption with the NC group ($p>0.05$). The 40%E and 50%E groups presented significant lower water sorption than PC1 group ($p<0.05$). The 40%E and 50%E groups displayed significant lower water solubility than the other groups ($p<0.05$).

Comparing in four auto-polymerizing hard reline resins found that the NC and PC1 groups presented the UT material no significant differences in water sorption with KL and TR materials ($p>0.05$) whereas UG material presented significant lower water sorption than the other materials ($p<0.05$). The PC2 and 10%E groups showed the UT and KL materials significant higher water sorption than TR and UG materials, respectively ($p<0.05$). The 0%E and 20%E of KL material exhibited significant higher water sorption than UT, TR, and UG, respectively ($p<0.05$). The 30%E group showed the UT material significant higher water sorption than TR, KL, and UG, respectively ($p<0.05$). The 40%E and 50%E groups presented the UT material significant higher water sorption than the other materials ($p<0.05$) but the KL material no significant differences with TR ($p>0.05$) shown in figure 15.

Considering NC, PC1, PC2, 0%E, and 20%E groups, TR material showed significant differences higher water solubility than the UG, KL, and UT materials ($p<0.05$) whereas KL material presented no significant difference with UG material ($p>0.05$). The 10%E group revealed the TR material significant higher water solubility

than the KL, UG, and UT materials, respectively ($p<0.05$). The 30%E group showed the UT and KL material significant lower water solubility than the UG and TR materials, respectively ($p<0.05$). The 40%E group presented the TR material significant higher water solubility than the UT and KL ($p<0.05$) but the TR material no significant differences with UG ($p>0.05$). The 50%E group showed the KL material significant lower water solubility than the UT, UG, and TR ($p<0.05$) as shown in figure 16.

CHAPTER V
DISCUSSION AND CONCLUSION

The present study investigated the effect of reducing residual monomer using a range of ethanol concentrations in an ultrasonic cleaner on the physical properties of four auto-polymerizing hard reline resins. The results of our study indicated the reduction of residual monomer in resin treated by ethanol solutions in an ultrasonic cleaner does not adversely affect its physical properties of the auto-polymerizing hard reline resins.

Flexural strength and modulus

Tsuchiya et al suggested the immersion of acrylic resin dentures in 50°C water for 60 minutes before insertion, especially for auto-polymerized acrylic resins, significantly decreased residual monomer (11). In our study, the immersion in water at 50°C for 60 minutes groups of UT, UG, and TR had a significantly lower flexural strength compared with the 10% ethanol in an ultrasonic cleaner. This study may be different solvents of post-polymerization treatment, it influences reduction of residual monomer (14). Some researchers observed post-polymerization of acrylic reline resins and reported that efficiency reduction of the residual monomer content at 55°C due to ethanol solutions treatment when compared with water (14, 17). The Hildebrand solubility parameter (δ) provides a numerical value estimate of the degree of interaction between materials which similar values of δ are likely to be miscible. The δ of the ethanol is 26.2 $MPa^{1/2}$ that is closer to the monomer (δ = 18.0 $MPa^{1/2}$) than the water (δ = 48.0 $MPa^{1/2}$) (35). Several investigators studied the relationship between residual monomers and mechanical properties of acrylic resin (5, 6, 23). They agreed that a high amount of residual monomers adversely affects the mechanical properties of the acrylic resin. Therefore, flexural strength is improved by lower amounts of residual monomer in material.

UT demonstrated the flexural strength and flexural modulus were maintained after being ultrasonically cleaned the 0%, 10%, 20%, 30%, and 40% ethanol in an

ultrasonic cleaner compared with the NC and PC2 groups. Whereas, KL material produced increase on the flexural strength after 10% and 20% ethanol in ultrasonic treatment compared with the NC and PC2 groups. After post-polymerization treatment of KL material demonstrated a significant increase in flexural modulus compared with the NC group. These results are in accordance with previous studies that described KL to be the material with the highest level of residual monomer (9, 14). Therefore, post-polymerization treatment to reduce the RM content of KL. As a result, flexural strength and modulus are improved. Compared with the other materials in NC group, KL had flexible characteristics such as significantly lower flexural modulus than the other materials. Because of the presence of the isobutyl methacrylate (IBMA) that the principle ingredient of the liquid without a cross-linking agent. Similar result has been found by Arima et al to compare the properties of several reline acrylic resins in which KL had significantly lower transverse bend strength and modulus of elasticity than highly cross linking reline acrylic resins (4). The flexural strength of UG and TR was not improved by ethanol solution in ultrasonic post-polymerization compared with the NC group. For UG material, this study showed did not significant differences from the NC group on flexural strength after 0%, 10%, 20%, and 30% ethanol treatment in an ultrasonic cleaner. Moreover, for the 40% and 50% ethanol solution in an ultrasonic cleaner presented significant lower flexural strength than the NC group. These findings are in agreement with those reported by Neves and Lopes who observed that ethanol treatment of an auto-polymerizing acrylic resin, UG specimens did not show any differences on flexural strength after 20% ethanol treatment. Moreover, the 50% and 70% ethanol solution produced a reduction on the flexural strength of the resin (14). The result of UG are similar TR because both materials which contained a cross-linking agent 1,6-hexanediol dimethacrylate (1,6 HDMA). The long distance between the 2 methacrylate groups of 1,6-HDMA probably enhanced the reactivity of the second double bond, thereby resulting in a more complete polymerization and lower of residual monomer (14, 36). Therefore, post-polymerization treatment is not increase flexural strength and modulus in UG and TR materials.

Comparing in four auto-polymerizing hard reline resins found that all experimental groups of UT presented significant higher values of flexural strength and modulus than the other reline acrylic resins after reducing residual monomers by the same method. This result may be explained by the chemical composition of the UT material based on polymethyl methacrylate (PMMA) polymer and having methyl methacrylate (MMA) as the monomer. Similar result was already found in earlier studies since PMMA based reline resin yielded higher transverse bend strength and modulus of elasticity to PMMA based denture base resin than PEMA based reline resin (4). For the NC group, KL had a significantly lower flexural strength compared with the UG. In this study may be explained due to amount residual monomer that KL to be the material with the highest level of residual monomer (14). However, the 20% and 40% ethanol solution in an ultrasonic cleaner that KL shows significantly higher flexural strength than the UG. Post-polymerization with ethanol solution in ultrasonic cleaner promote a more effective reduction in residual monomer led to increase flexural strength in KL material.

Surface hardness

Our Vickers hardness value results indicated that there were no significant differences between the groups within each specific auto-polymerizing acrylic hard denture reline materials after reducing residual monomer by the different methods. Similar results was found by Seo and Vergani, who demonstrated that auto-polymerizing acrylic resin had no difference in hardness before and after post-polymerization treatment (36). These results are combined with flexural tests, indicated that the effects of post-polymerization treatments was more pronounced in the bulk of the specimens rather than in their superficial layer. This is supported by Mello et al (2003), in which higher hardness values were recorded at greater specimen depths after post-polymerization using microwave energy or hot water (37). Comparing in four auto-polymerizing hard denture reline resins within each treatment group found that UT was significantly harder than those made with UG, TR, and KL materials, respectively. Finally, it was not surprising that KL showed the lowest Vickers hardness values of all materials. The isobutyl methacrylate molecules, it is

principle ingredient of KL increase the backbone separation of the polymer molecules, decreasing the intermolecular interaction. Thus, the isobutyl possesses a lower surface hardness (2). Although UG and TR contain the same cross-linking agent in the liquid, it was interesting to note that TR showed significantly lower hardness values than UG material. This may be explained by the use of the monofunctional monomer, 39.8% ß-methacryloyloxyethyl propionate (MAOP) that constitutes the liquid component of TR material. The MAOP molecule contains 2 esteric bonds that form flexible polymer chains upon polymerization (4). The flexibility of the polymer chain may account for the relatively lower hardness of TR in comparison to UG.

Water sorption and solubility

In our study, the measure of water sorption and solubility used the method that recommended by ISO. The water sorption was determined according to increase in mass per unit volume. Also water solubility was determined according to loose of mass from polymers (18). Polymethy methacrylate is not soluble in water (4). Therefore, the water solubility of UT had significantly lower than the other hard reline resins. UT material was maintained of water sorption and solubility compared the NC, PC1, and PC2 groups after 0%, 10%, 20%, and 30% ethanol solution in an ultrasonic treatment, whereas 40% and 50% ethanol solution increased water sorption and solubility than the other groups. This result may be explained that the higher concentration of ethanol had effect the porosity of material, leading to enhanced water uptake. This was confirmed by a reduction of the flexural strength of the UT specimens, when compared to lower concentration of ethanol. The water solubility of acrylic is related to its water sorption (22), therefore water solubility was increased. KL had effective decrease after 30%, 40%, and 50% ethanol solution in an ultrasonic treatment on the water sorption and solubility than the NC, PC1, and PC2 group. UG and TR are materials that contain 1,6-hexanediol dimethacrylate in the liquids which is a cross-linking agents. UG and TR showed the same lower water solubility after 40% and 50% ethanol solution in an ultrasonic treatment than the NC, PC1, and PC2 group. The result of UT opposite with the KL, UG, and TR because of difference resin matrix composition. Water is absorbed into polymer by the

polarity of the molecules in the polymers by unsaturated bonds of the molecules or unbalanced intermolecular forces in the polymers (38). It may be explained higher proportion of ethanol in the solution leads to a more activity in polymer. The hydroxyl group in ethanol is also responsible for the hydrogen bonds that form between adjacent molecules. The MMA as the monomer in UT material contained only one pair double bond, while non-MMA base monomer appeared long hydrocarbon chains and functional groups (terminal two pairs double bond). Therefore, hydrogen bonding in water molecule is slightly attracted to the oxygen atoms of non-MMA base molecules. As a result, water sorption and solubility of non-MMA based hard reline materials decreased with higher proportion of ethanol.

In the part of water sorption compare between four auto-polymerizing hard reline resins receiving the same treatment, the study found that UG had a significant lower values of water sorption than those made with TR, UT, and KL materials. According to Arima et al, the highly cross-linked reline acrylic resin had lower water sorption than non-crosslinked reline acrylic resin (4). Importantly, the tested groups complied with the requirements of International Standards Organization 20795-1:2013, i.e. water sorption and water solubility shall not exceed 32 μg/mm^3 and 8 μg/mm^3, respectively, for auto-polymerizing acrylic resins (18).

The most effective post polymerization treatment should be chosen that reduces the residual monomer content and techniques must not change material properties. The ethanol solution in ultrasonic treatment presented more effective than hot water on the reduction of the residual monomer (17). The higher proportion of ethanol in the solution leads to a more significant reduction of residual monomer content (14). Our experiments combine the possible benefits to reduction of residual monomer that interaction between aqueous ethanol solution and ultrasonic cleaner. This study proved that the reduction of residual monomer by ethanol solution in an ultrasonic cleaner did not adversely affect the physical properties of auto-polymerizing hard reline resins. It could be described ultrasonic treatment reduced residual monomer because increased the flow rate of the water in the tank, which increases the elution of the residual monomer. Moreover, implosive cavitation

bubbles were generated by the ultrasonic cleaner release energy to the surface of the specimens causing polymerization of the remaining monomer (9, 15, 16). In the part of ethanol aqueous solutions have been used in order to increase and accelerate compounds solubility that enhances the leaching processes (12, 13).

Conclusions

Under our experimental conditions, we found that the reduction of residual monomer in auto-polymerizing hard reline resins based on a combination approach of ethanol solutions and ultrasonic treatment at 55°C during 5 minutes does not adversely affect its physical properties. Specifically, for KL, the 10% ethanol in an ultrasonic cleaner should be the treatment of choice which effective increase the physical properties. In UG and TR, the 10% ethanol solution maintained the physical properties. In UT, the 30% ethanol solution had appropriate physical properties.

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APPENDIX

Table 7 One-Sample Kolmogorov-Smirnov tests analysis of the data distribution of auto-polymerizing acrylic hard denture reline materials.

Materials/Groups	Flexural strength	Flexural modulus	Surface hardness	Water sorption	Water solubility
Unifast Trad					
1) NC	.200	.200	.200	.098	.200
2) PC1	.200	.200	.200	.200	.200
3) PC2	.200	.200	.126	.200	.086
4) 0%E	.155	.200	.060	.200	.200
5) 10%E	.200	.200	.200	.200	.200
6) 20%E	.200	.200	.200	.200	.150
7) 30%E	.200	.200	.200	.200	.200
8) 40%E	.200	.200	.200	.200	.200
9) 50%E	.200	.200	.200	.200	.200
Kooliner					
1) NC	.200	.156	.200	.200	.200
2) PC1	.200	.200	.200	.200	.060
3) PC2	.200	.200	.200	.200	.200
4) 0%E	.200	.200	.051	.162	.200
5) 10%E	.200	.200	.059	.142	.200
6) 20%E	.200	.075	.200	.200	.200
7) 30%E	.200	.200	.200	.200	.200
8) 40%E	.119	.200	.153	.200	.200
9) 50%E	.200	.200	.200	.200	.200

Materials/Groups	Flexural strength	Flexural modulus	Surface hardness	Water sorption	Water solubility
Ufi Gel Hard					
1) NC	.200	.200	.200	.200	.138
2) PC1	.069	.200	.200	.178	.164
3) PC2	.200	.121	.200	.200	.200
4) 0%E	.200	.200	.200	.080	.200
5) 10%E	.200	.053	.068	.084	.200
6) 20%E	.200	.200	.200	.066	.060
7) 30%E	.200	.200	.200	.200	.200
8) 40%E	.089	.065	.200	.200	.089
9) 50%E	.200	.071	.200	.200	.200
Tokuso Rebase II					
1) NC	.0.67	.200	.200	.200	.200
2) PC1	.200	.123	.200	.200	.051
3) PC2	.059	.200	.200	.117	.200
4) 0%E	.200	.050	.127	.200	.200
5) 10%E	.069	.200	.200	.091	.200
6) 20%E	.200	.055	.122	.172	.200
7) 30%E	.200	.181	.111	.200	.200
8) 40%E	.200	.200	.072	.200	.200
9) 50%E	.200	.200	.200	.053	.200

Table 8 The Levene statistical analysis of auto-polymerizing acrylic hard denture reline materials.

Types of hard denture reline materials.	Flexural strength	Flexural modulus	Surface hardness	Water sorption	Water solubility
Unifast Trad	.111	.249	.081	.485	.188
Kooliner	.102	.721	.344	.540	.120
Ufi Gel Hard	.509	.927	.212	.331	.163
Tokuso Rebase II	.329	.277	.114	.061	.077

Table 9 One-way analysis of variance (ANOVA) test of flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of each auto-polymerizing acrylic hard denture reline material.

Types of hard denture reline materials.	Flexural strength	Flexural modulus	Surface hardness	Water sorption	Water solubility
Unifast Trad	.000	.000	.153	.000	.000
Kooliner	.000	.000	.086	.000	.000
Ufi Gel Hard	.000	.000	.056	.000	.000
Tokuso Rebase II	.000	.031	.093	.003	.000

Table 10 Tukey's HSD test of flexural strength, flexural modulus, surface hardness, water sorption, and water solubility of each auto-polymerizing acrylic hard denture reline material.

Materials/Groups	Flexural strength	Flexural modulus	Water sorption	Water solubility
Unifast Trad				
Group 1 – Group 2	.646	.671	.506	.999
Group 1 – Group 3	1.000	.184	1.000	1.000
Group 1 – Group 4	.672	.972	1.000	1.000
Group 1 – Group 5	.715	1.000	.539	.983
Group 1 – Group 6	.917	1.000	1.000	.982
Group 1 – Group 7	.910	.161	.948	1.000
Group 1 – Group 8	.980	.996	.000	.009
Group 1 – Group 9	.000	.000	.000	.000
Group 2 – Group 3	.333	.996	.459	.983
Group 2 – Group 4	.012	.998	.321	1.000
Group 2 – Group 5	.015	.612	1.000	1.000
Group 2 – Group 6	.049	.427	.374	1.000
Group 2 – Group 7	.047	.993	.995	1.000
Group 2 – Group 8	.996	.984	.041	.001
Group 2 – Group 9	.001	.000	.000	.000
Group 3 – Group 4	.918	.831	1.000	.999
Group 3 – Group 5	.938	.152	.490	.904
Group 3 – Group 6	.995	.079	1.000	.898
Group 3 – Group 7	.994	1.000	.929	.992
Group 3 – Group 8	.841	.653	.000	.028
Group 3 – Group 9	.000	.000	.000	.000
Group 4 – Group 5	1.000	.956	.349	.998

Materials/Groups	Flexural strength	Flexural modulus	Water sorption	Water solubility
Unifast Trad				
Group 4 – Group 6	1.000	.866	1.000	.998
Group 4 – Group 7	1.000	.799	.841	1.000
Group 4 – Group 8	.115	1.000	.000	.004
Group 4 – Group 9	.000	.000	.000	.000
Group 5 – Group 6	1.000	1.000	.403	1.000
Group 5 – Group 7	1.000	.132	.997	1.000
Group 5 – Group 8	.134	.993	.036	.000
Group 5 – Group 9	.000	.000	.000	.000
Group 6 – Group 7	1.000	.067	.882	1.000
Group 6 – Group 8	.309	.960	.000	.000
Group 6 – Group 9	.000	.000	.000	.000
Group 7 – Group 8	.299	.613	.003	.002
Group 7 – Group 9	.000	.000	.000	.000
Group 8 – Group 9	.000	.000	.721	.000
Kooliner				
Group 1 – Group 2	.000	.000	1.000	1.000
Group 1 – Group 3	.000	.000	.996	.364
Group 1 – Group 4	.000	.000	.432	.577
Group 1 – Group 5	.000	.000	.854	.256
Group 1 – Group 6	.000	.000	.958	.783
Group 1 – Group 7	.000	.017	.000	.000
Group 1 – Group 8	.000	.036	.000	.000
Group 1 – Group 9	.111	.012	.000	.000
Group 2 – Group 3	.028	.005	.936	.261
Group 2 – Group 4	.000	.045	.740	.451

Materials/Groups	Flexural strength	Flexural modulus	Water sorption	Water solubility
Kooliner				
Group 2 – Group 5	.357	1.000	.982	.175
Group 2 – Group 6	.732	1.000	.998	.667
Group 2 – Group 7	.000	.000	.000	.000
Group 2 – Group 8	.000	.000	.000	.000
Group 2 – Group 9	.000	.000	.000	.000
Group 3 – Group 4	.794	.998	.085	1.000
Group 3 – Group 5	.000	.004	.352	1.000
Group 3 – Group 6	.000	.028	.556	.999
Group 3 – Group 7	.884	.817	.000	.000
Group 3 – Group 8	.506	.959	.000	.000
Group 3 – Group 9	.000	.871	.000	.000
Group 4 – Group 5	.000	.038	.999	1.000
Group 4 – Group 6	.000	.180	.985	1.000
Group 4 – Group 7	1.000	.349	.000	.000
Group 4 – Group 8	1.000	.215	.000	.000
Group 4 – Group 9	.000	.418	.000	.000
Group 5 – Group 6	1.000	.999	1.000	.995
Group 5 – Group 7	.000	.000	.000	.000
Group 5 – Group 8	.000	.000	.000	.000
Group 5 – Group 9	.000	.000	.000	.000
Group 6 – Group 7	.000	.000	.000	.000
Group 6 – Group 8	.000	.000	.000	.000
Group 6 – Group 9	.000	.000	.000	.000
Group 7 – Group 8	.999	1.000	.964	1.000
Group 7 – Group 9	.000	1.000	.990	1.000

Materials/Groups	Flexural strength	Flexural modulus	Water sorption	Water solubility
Kooliner				
Group 8 – Group 9	.002	1.000	1.000	1.000
UFi Gel Hard				.
Group 1 – Group 2	.620	.990	.370	1.000
Group 1 – Group 3	1.000	.998	.979	1.000
Group 1 – Group 4	1.000	.965	.177	.579
Group 1 – Group 5	.659	.905	.157	.270
Group 1 – Group 6	1.000	.969	.164	.343
Group 1 – Group 7	1.000	.962	.179	.100
Group 1 – Group 8	.018	.346	1.000	.025
Group 1 – Group 9	.016	.166	1.000	.019
Group 2 – Group 3	.876	1.000	.949	1.000
Group 2 – Group 4	.455	.492	.000	.602
Group 2 – Group 5	.010	.348	.000	.288
Group 2 – Group 6	.528	.507	.000	.363
Group 2 – Group 7	.901	.480	.000	.108
Group 2 – Group 8	.774	.903	.128	.028
Group 2 – Group 9	.752	.709	.638	.021
Group 3 – Group 4	.999	.641	.011	.470
Group 3 – Group 5	.365	.487	.009	.196
Group 3 – Group 6	1.000	.655	.010	.256
Group 3 – Group 7	1.000	.629	.011	.067
Group 3 – Group 8	.064	.804	.808	.015
Group 3 – Group 9	.058	.563	.999	.012
Group 4 – Group 5	.808	1.000	1.000	1.000
Group 4 – Group 6	1.000	1.000	1.000	1.000

Materials/Groups	Flexural strength	Flexural modulus	Water sorption	Water solubility
UFi Gel Hard				
Group 4 – Group 7	.998	1.000	1.000	.988
Group 4 – Group 8	.008	.023	.463	.861
Group 4 – Group 9	.007	.007	.068	.819
Group 5 – Group 6	.745	1.000	1.000	1.000
Group 5 – Group 7	.329	1.000	1.000	1.000
Group 5 – Group 8	.000	.012	.427	.987
Group 5 – Group 9	.000	.003	.059	.977
Group 6 – Group 7	.999	1.000	1.000	1.000
Group 6 – Group 8	.012	.025	.440	.970
Group 6 – Group 9	.011	.008	.062	.954
Group 7 – Group 8	.075	.022	.466	1.000
Group 7 – Group 9	.068	.007	.068	1.000
Group 8 – Group 9	1.000	1.000	.989	1.000
Tokuso Rebase II				
Group 1 – Group 2	.055	.885	.994	.995
Group 1 – Group 3	.803	.994	1.000	.849
Group 1 – Group 4	.008	.508	1.000	.555
Group 1 – Group 5	.214	.952	1.000	.850
Group 1 – Group 6	.631	1.000	1.000	.792
Group 1 – Group 7	.771	.970	1.000	.402
Group 1 – Group 8	.101	.891	.155	.000
Group 1 – Group 9	.012	.928	.105	.000
Group 2 – Group 3	.826	1.000	.901	.326
Group 2 – Group 4	.999	.999	.967	.122
Group 2 – Group 5	.000	.189	.923	.327

Materials/Groups	Flexural strength	Flexural modulus	Water sorption	Water solubility
Tokuso Rebase II				
Group 2 – Group 6	.000	.586	.983	.268
Group 2 – Group 7	.854	1.000	.983	.069
Group 2 – Group 8	1.000	1.000	.016	.000
Group 2 – Group 9	1.000	1.000	.009	.000
Group 3 – Group 4	.411	.955	1.000	1.000
Group 3 – Group 5	.002	.497	1.000	1.000
Group 3 – Group 6	.022	.901	1.000	1.000
Group 3 – Group 7	1.000	1.000	1.000	.998
Group 3 – Group 8	.924	1.000	.414	.000
Group 3 – Group 9	.502	1.000	.313	.000
Group 4 – Group 5	.000	.040	1.000	1.000
Group 4 – Group 6	.000	.212	1.000	1.000
Group 4 – Group 7	.448	.990	1.000	1.000
Group 4 – Group 8	.993	.999	.271	.000
Group 4 – Group 9	1.000	.998	.194	.000
Group 5 – Group 6	.999	.999	1.000	1.000
Group 5 – Group 7	.002	.339	1.000	.998
Group 5 – Group 8	.000	.194	.372	.000
Group 5 – Group 9	.000	.241	.278	.000
Group 6 – Group 7	.019	.782	1.000	1.000
Group 6 – Group 8	.000	.595	.216	.000
Group 6 – Group 9	.000	.667	.151	.000
Group 7 – Group 8	.941	1.000	.217	.000
Group 7 – Group 9	.540	1.000	.152	.000
Group 8 – Group 9	.998	1.000	1.000	.628

Table 11 One-way analysis of variance (ANOVA) test of flexural strength, flexural modulus, surface hardness, water sorption, and water solubility in four types of auto-polymerizing hard denture reline resins after reducing residual monomers by the same method.

Group	Flexural strength	Flexural modulus	Surface hardness	Water sorption	Water solubility
1) NC	.000	.000	.000	.000	.000
2) PC1	.000	.000	.000	.000	.000
3) PC2	.000	.000	.000	.000	.000
4) 0%E	.000	.000	.000	.000	.000
5) 10%E	.000	.000	.000	.000	.000
6) 20%E	.000	.000	.000	.000	.000
7) 30%E	.000	.000	.000	.000	.000
8) 40%E	.000	.000	.000	.000	.001
9) 50%E	.000	.000	.000	.000	.000

Table 12 Dunnett T3 test of flexural strength, flexural modulus, surface hardness, water sorption, and water solubility in four types of auto-polymerizing hard denture reline resins after reducing residual monomers by the same method.

Materials/Groups	Flexural strength	Flexural modulus	Surface hardness	Water sorption	Water solubility
NC group					
UT – KL	.000	.000	.000	.685	.000
UT – UG	.000	.020	.000	.000	.000
UT – TR	.000	.000	.000	.115	.000
KL – UG	.029	.000	.000	.000	.559
KL – TR	.000	.000	.000	.008	.000
UG – TR	.000	.000	.000	.000	.000

Materials/Groups	Flexural strength	Flexural modulus	Surface hardness	Water sorption	Water solubility
PC1 group					
UT – KL	.000	.000	.000	.792	.000
UT – UG	.000	.000	.000	.000	.000
UT – TR	.000	.000	.000	.132	.000
KL – UG	.037	.000	.000	.000	.553
KL – TR	.000	.112	.000	.016	.000
UG – TR	.000	.000	.000	.000	.000
PC2 group					
UT – KL	.000	.000	.000	.961	.000
UT – UG	.000	.000	.000	.000	.000
UT – TR	.000	.000	.000	.005	.000
KL – UG	.979	.000	.000	.000	.982
KL – TR	.000	.209	.000	.001	.000
UG – TR	.000	.000	.000	.000	.000
0%E group					
UT – KL	.000	.000	.000	.001	.000
UT – UG	.000	.001	.000	.000	.000
UT – TR	.000	.000	.000	.009	.000
KL – UG	.715	.000	.000	.000	.118
KL – TR	.000	1.000	.000	.000	.000
UG – TR	.000	.000	.000	.000	.000

Materials/Groups	Flexural strength	Flexural modulus	Surface hardness	Water sorption	Water solubility
10% group					
UT – KL	.000	.000	.000	.208	.000
UT – UG	.000	.135	.000	.000	.000
UT – TR	.000	.000	.000	.002	.000
KL – UG	1.000	.000	.000	.000	.020
KL – TR	.000	.933	.008	.000	.000
UG – TR	.001	.000	.000	.000	.000
20% group					
UT – KL	.000	.000	.000	.013	.000
UT – UG	.000	.117	.000	.000	.000
UT – TR	.000	.000	.000	.007	.000
KL – UG	.032	.000	.000	.000	.169
KL – TR	.000	.966	.000	.000	.000
UG – TR	.000	.000	.000	.000	.000
30%E group					
UT – KL	.000	.000	.000	.000	.355
UT – UG	.000	.005	.000	.000	.000
UT – TR	.000	.000	.000	.001	.000
KL – UG	1.000	.000	.000	.000	.014
KL – TR	.000	.010	.001	.006	.000
UG – TR	.000	.000	.000	.000	.000

Materials/Groups	Flexural strength	Flexural modulus	Surface hardness	Water sorption	Water solubility
40% group					
UT – KL	.000	.000	.000	.000	.998
UT – UG	.000	.000	.000	.000	.061
UT – TR	.000	.000	.000	.000	.005
KL – UG	.017	.000	.000	.000	.043
KL – TR	.000	.001	.000	.105	.003
UG – TR	.000	.000	.000	.000	.748
50% group					
UT – KL	.000	.000	.000	.000	.000
UT – UG	.000	.466	.000	.000	.941
UT – TR	.000	.000	.000	.000	.417
KL – UG	.490	.000	.000	.000	.000
KL – TR	.000	.031	.004	.169	.028
UG – TR	.000	.000	.000	.000	.166

Table 13 The flexural strength of auto-polymerizing hard denture reline in each specimen.

Materials/ Groups	No	Flexural strength (MPa)	Mean	SD
UT/NC	1	57.15		
	2	63.66		
	3	51.02		
	4	63.38		
	5	57.95		
	6	59.92		
	7	63.17		
	8	54.24		
	9	59.85		
	10	56.85	58.72	4.14
UT/PC1	1	59.74		
	2	52.88		
	3	56.15		
	4	62.76		
	5	53.78		
	6	57.37		
	7	51.20		
	8	51.59		
	9	57.17		
	10	58.42	56.11	3.74
UT/PC2	1	62.37		
	2	57.09		
	3	60.92		
	4	56.32		
	5	57.13		
	6	59.85		
	7	58.99		
	8	56.80		
	9	60.93		
	10	63.59	59.40	2.54

Materials/ Groups	No	Flexural strength (MPa)	Mean	SD
UT/0%E	1	59.13		
	2	56.86		
	3	62.62		
	4	62.41		
	5	61.91		
	6	62.47		
	7	60.57		
	8	63.26		
	9	62.16		
	10	61.38	61.28	1.95
UT/10%E	1	61.17		
	2	62.90		
	3	58.14		
	4	60.77		
	5	56.93		
	6	65.97		
	7	63.55		
	8	59.34		
	9	58.10		
	10	64.95	61.18	3.09
UT/20%E	1	63.51		
	2	62.72		
	3	57.52		
	4	64.14		
	5	58.90		
	6	55.82		
	7	58.55		
	8	66.31		
	9	63.24		
	10	55.33	60.60	3.83

Materials/ Groups	No	Flexural strength (MPa)	Mean	SD
UT/30%E	1	56.68		
	2	58.55		
	3	64.98		
	4	57.92		
	5	60.59		
	6	60.92		
	7	61.41		
	8	65.68		
	9	56.18		
	10	63.40	60.63	3.33
UT/40%E	1	56.48		
	2	59.60		
	3	59.64		
	4	56.93		
	5	53.53		
	6	55.18		
	7	59.72		
	8	52.76		
	9	58.53		
	10	60.15	57.25	2.72
UT/50%E	1	47.39		
	2	50.75		
	3	47.61		
	4	49.85		
	5	52.02		
	6	52.92		
	7	48.51		
	8	49.94		
	9	51.97		
	10	46.06	49.70	2.27

Materials/ Groups	No	Flexural strength (MPa)	Mean	SD
KL/NC	1	35.04		
	2	32.92		
	3	32.33		
	4	34.62		
	5	34.42		
	6	33.50		
	7	34.40		
	8	32.41		
	9	33.85		
	10	32.38	33.59	1.03
KL/PC1	1	41.30		
	2	38.69		
	3	39.70		
	4	40.30		
	5	38.18		
	6	38.71		
	7	40.05		
	8	41.69		
	9	40.57		
	10	41.04	40.02	1.19
KL/PC2	1	37.85		
	2	40.46		
	3	39.49		
	4	39.60		
	5	37.14		
	6	37.88		
	7	37.70		
	8	38.51		
	9	39.97		
	10	34.17	38.28	1.82

Materials/ Groups	No	Flexural strength (MPa)	Mean	SD
KL/0%E	1	38.52		
	2	36.11		
	3	37.16		
	4	37.65		
	5	37.33		
	6	38.01		
	7	36.86		
	8	37.76		
	9	36.42		
	10	38.66	37.45	0.84
KL/10%E	1	42.15		
	2	40.63		
	3	42.06		
	4	39.89		
	5	39.85		
	6	43.20		
	7	40.41		
	8	42.17		
	9	41.53		
	10	40.11	41.20	1.17
KL/20%E	1	40.03		
	2	41.17		
	3	39.07		
	4	41.68		
	5	41.50		
	6	42.40		
	7	41.40		
	8	41.59		
	9	39.36		
	10	40.85	40.90	1.08

Materials/ Groups	No	Flexural strength (MPa)	Mean	SD
KL/30%E	1	37.81		
	2	36.51		
	3	40.65		
	4	35.87		
	5	37.27		
	6	36.79		
	7	37.98		
	8	38.63		
	9	36.34		
	10	37.60	37.55	1.38
KL/40%E	1	38.00		
	2	35.59		
	3	37.00		
	4	37.07		
	5	36.92		
	6	37.46		
	7	37.66		
	8	37.06		
	9	37.30		
	10	38.12	37.22	0.71
KL/50%E	1	35.45		
	2	33.99		
	3	34.82		
	4	35.26		
	5	34.97		
	6	34.52		
	7	34.96		
	8	34.85		
	9	35.31		
	10	36.48	35.06	0.65

Materials/ Groups	No	Flexural strength (MPa)	Mean	SD
UG/NC	1	43.51		
	2	36.61		
	3	33.99		
	4	31.92		
	5	39.07		
	6	40.11		
	7	35.62		
	8	35.77		
	9	43.11		
	10	42.29	38.20	4.02
UG/PC1	1	35.73		
	2	35.54		
	3	31.95		
	4	34.08		
	5	45.93		
	6	34.39		
	7	32.54		
	8	37.07		
	9	30.53		
	10	35.86	35.36	4.23
UG/PC2	1	34.06		
	2	38.98		
	3	40.90		
	4	35.49		
	5	35.92		
	6	42.61		
	7	34.12		
	8	36.02		
	9	39.58		
	10	37.63	37.53	2.91

Materials/ Groups	No	Flexural strength (MPa)	Mean	SD
UG/0%E	1	37.65		
	2	37.36		
	3	35.54		
	4	41.96		
	5	40.26		
	6	36.35		
	7	38.76		
	8	39.91		
	9	35.33		
	10	42.59	38.57	2.56
UG/10%E	1	40.41		
	2	36.82		
	3	45.39		
	4	39.12		
	5	46.89		
	6	36.50		
	7	38.81		
	8	43.69		
	9	38.10		
	10	43.81	40.95	3.71
UG/20%E	1	41.37		
	2	38.66		
	3	39.33		
	4	40.07		
	5	39.96		
	6	35.71		
	7	34.22		
	8	39.33		
	9	37.85		
	10	37.56	38.41	2.15

Materials/ Groups	No	Flexural strength (MPa)	Mean	SD
UG/30%E	1	34.87		
	2	38.84		
	3	38.85		
	4	38.41		
	5	38.81		
	6	36.86		
	7	35.05		
	8	34.68		
	9	41.45		
	10	36.55	37.44	2.21
UG/40%E	1	31.56		
	2	34.05		
	3	39.54		
	4	31.53		
	5	32.93		
	6	26.14		
	7	33.21		
	8	31.68		
	9	32.30		
	10	35.96	32.89	3.43
UG/50%E	1	35.06		
	2	30.15		
	3	36.91		
	4	36.67		
	5	30.87		
	6	33.09		
	7	32.05		
	8	37.14		
	9	23.52		
	10	32.87	32.83	4.13

Materials/ Groups	No	Flexural strength (MPa)	Mean	SD
TR/NC	1	46.27		
	2	46.58		
	3	46.41		
	4	45.43		
	5	48.13		
	6	48.48		
	7	46.02		
	8	45.97		
	9	46.03		
	10	47.93	46.72	1.06
TR/PC1	1	44.27		
	2	45.51		
	3	45.72		
	4	43.91		
	5	45.57		
	6	44.44		
	7	45.33		
	8	44.92		
	9	44.07		
	10	45.72	44.95	0.72
TR/PC2	1	46.00		
	2	44.13		
	3	46.62		
	4	46.70		
	5	47.10		
	6	43.22		
	7	46.15		
	8	46.37		
	9	45.61		
	10	46.33	45.82	1.22

Materials/ Groups	No	Flexural strength (MPa)	Mean	SD
TR/0%E	1	43.81		
	2	43.86		
	3	43.16		
	4	43.62		
	5	45.26		
	6	46.32		
	7	45.19		
	8	44.03		
	9	45.90		
	10	44.63	44.58	1.05
TR/10%E	1	48.93		
	2	47.75		
	3	47.17		
	4	47.33		
	5	50.72		
	6	48.54		
	7	47.38		
	8	48.66		
	9	47.65		
	10	47.62	48.18	1.08
TR/20%E	1	48.65		
	2	50.43		
	3	47.13		
	4	45.85		
	5	48.07		
	6	46.90		
	7	47.83		
	8	47.92		
	9	48.45		
	10	46.62	47.78	1.27

Materials/ Groups	No	Flexural strength(MPa)	Mean	SD
TR/30%E	1	44.20		
	2	47.66		
	3	47.50		
	4	44.10		
	5	44.36		
	6	45.60		
	7	45.96		
	8	45.07		
	9	45.90		
	10	47.56	45.79	1.40
TR/40%E	1	43.97		
	2	44.11		
	3	44.67		
	4	46.17		
	5	46.22		
	6	45.21		
	7	42.93		
	8	46.25		
	9	46.03		
	10	45.24	45.08	1.14
TR/50%E	1	44.68		
	2	41.79		
	3	45.16		
	4	43.33		
	5	46.66		
	6	45.71		
	7	45.70		
	8	44.82		
	9	41.23		
	10	47.50	44.66	2.01

Table 14 The flexural modulus of auto-polymerizing hard denture reline in each specimen.

Materials/ Groups	No	Flexural modulus (MPa)	Mean	SD
UT/NC	1	1,534.31		
	2	1,401.35		
	3	1,490.62		
	4	1,531.17		
	5	1,367.00		
	6	1,537.85		
	7	1,472.59		
	8	1,510.32		
	9	1,535.97		
	10	1,582.74	1,496.39	66.63
UT/PC1	1	1,420.00		
	2	1,543.22		
	3	1,523.61		
	4	1,495.98		
	5	1,574.74		
	6	1,516.78		
	7	1,596.57		
	8	1,573.25		
	9	1,619.16		
	10	1,584.25	1,544.75	58.37
UT/PC2	1	1,655.24		
	2	1,580.44		
	3	1,588.53		
	4	1,513.72		
	5	1,564.37		
	6	1,578.92		
	7	1,586.24		
	8	1,520.59		
	9	1,557.26		
	10	1,523.53	1,566.88	42.07

Materials/ Groups	No	Flexural modulus (MPa)	Mean	SD
UT/0%E	1	1,528.01		
	2	1,473.13		
	3	1,507.31		
	4	1,528.82		
	5	1,587.57		
	6	1,534.35		
	7	1,494.74		
	8	1,540.39		
	9	1,557.29		
	10	1,505.85	1,525.75	32.64
UT/10%E	1	1,434.31		
	2	1,451.73		
	3	1,511.88		
	4	1,477.70		
	5	1,597.39		
	6	1,481.68		
	7	1,466.54		
	8	1,444.09		
	9	1,499.35		
	10	1,575.66	1,494.03	54.49
UT/20%E	1	1,522.74		
	2	1,602.04		
	3	1,504.47		
	4	1,539.15		
	5	1,420.79		
	6	1,443.57		
	7	1,400.06		
	8	1,513.94		
	9	1,484.34		
	10	1,435.32	1,486.64	62.15

Materials/ Groups	No	Flexural modulus (MPa)	Mean	SD
UT/30%E	1	1,589.02		
	2	1,705.33		
	3	1,520.49		
	4	1,571.01		
	5	1,591.86		
	6	1,541.60		
	7	1,580.22		
	8	1,473.28		
	9	1,716.87		
	10	1,395.18	1,568.49	96.60
UT/40%E	1	1,580.97		
	2	1,544.60		
	3	1,520.90		
	4	1,528.94		
	5	1,507.62		
	6	1,470.06		
	7	1,506.11		
	8	1,440.11		
	9	1,556.21		
	10	1,522.56	1,517.81	40.68
UT/50%E	1	1,271.90		
	2	1,348.38		
	3	1,322.57		
	4	1,360.59		
	5	1,407.67		
	6	1,454.58		
	7	1,274.30		
	8	1,348.23		
	9	1,403.50		
	10	1,322.88	1,351.46	58.25

Materials/ Groups	No	Flexural modulus (MPa)	Mean	SD
KL/NC	1	560.83		
	2	454.20		
	3	635.66		
	4	548.11		
	5	539.45		
	6	476.08		
	7	533.43		
	8	550.54		
	9	526.31		
	10	528.52	535.31	48.71
KL/PC1	1	873.22		
	2	772.48		
	3	657.84		
	4	783.00		
	5	780.83		
	6	712.36		
	7	854.50		
	8	656.10		
	9	738.32		
	10	767.56	759.62	72.19
KL/PC2	1	598.97		
	2	680.58		
	3	641.53		
	4	725.76		
	5	665.92		
	6	745.77		
	7	596.74		
	8	613.86		
	9	693.21		
	10	657.68	662.00	50.99

Materials/ Groups	No	Flexural modulus (MPa)	Mean	SD
KL/0%E	1	679.46		
	2	601.24		
	3	679.25		
	4	631.51		
	5	667.12		
	6	623.85		
	7	772.09		
	8	767.48		
	9	682.28		
	10	697.27	680.16	56.09
KL/10%E	1	708.18		
	2	864.84		
	3	813.33		
	4	731.25		
	5	724.20		
	6	693.74		
	7	792.36		
	8	727.71		
	9	750.18		
	10	805.89	761.17	55.08
KL/20%E	1	686.79		
	2	772.10		
	3	814.46		
	4	704.81		
	5	696.44		
	6	668.26		
	7	702.89		
	8	774.40		
	9	836.01		
	10	800.43	745.66	60.37

Materials/ Groups	No	Flexural modulus (MPa)	Mean	SD
KL/30%E	1	645.60		
	2	589.73		
	3	699.24		
	4	630.00		
	5	572.59		
	6	576.36		
	7	616.63		
	8	695.10		
	9	572.72		
	10	634.55	623.25	47.26
KL/40%E	1	628.97		
	2	583.07		
	3	588.27		
	4	693.17		
	5	562.85		
	6	610.05		
	7	664.49		
	8	565.45		
	9	629.79		
	10	641.80	616.79	42.94
KL/50%E	1	707.52		
	2	603.33		
	3	552.14		
	4	614.35		
	5	569.34		
	6	716.29		
	7	623.67		
	8	617.24		
	9	678.11		
	10	578.44	626.04	56.93

Materials/ Groups	No	Flexural modulus (MPa)	Mean	SD
UG/NC	1	1,362.13		
	2	1,378.66		
	3	1,369.75		
	4	1,288.76		
	5	1,402.08		
	6	1,323.01		
	7	1,289.93		
	8	1,527.92		
	9	1,492.49		
	10	1,423.60	1,385.83	79.37
UG/PC1	1	1,344.88		
	2	1,369.35		
	3	1,434.59		
	4	1,390.15		
	5	1,405.53		
	6	1,399.84		
	7	1,362.18		
	8	1,298.13		
	9	1,339.46		
	10	1,241.62	1,358.57	56.47
UG/PC2	1	1,328.82		
	2	1,499.34		
	3	1,279.02		
	4	1,328.58		
	5	1,307.18		
	6	1,339.58		
	7	1,430.58		
	8	1,309.70		
	9	1,456.89		
	10	1,369.99	1,364.97	72.95

Materials/ Groups	No	Flexural modulus (MPa)	Mean	SD
UG/0%E	1	1,426.88		
	2	1,444.35		
	3	1,325.85		
	4	1,332.94		
	5	1,454.65		
	6	1,462.48		
	7	1,482.30		
	8	1,382.58		
	9	1,476.64		
	10	1,402.05	1,419.07	56.66
UG/10%E	1	1,382.25		
	2	1,369.79		
	3	1,370.81		
	4	1,534.05		
	5	1,367.00		
	6	1,428.46		
	7	1,373.30		
	8	1,396.91		
	9	1,519.19		
	10	1,515.27	1,425.70	69.57
UG/20%E	1	1,347.57		
	2	1,302.31		
	3	1,463.29		
	4	1,439.51		
	5	1,435.19		
	6	1,392.83		
	7	1,439.09		
	8	1,398.35		
	9	1,500.04		
	10	1,466.39	1,418.46	59.34

Materials/ Groups	No	Flexural modulus (MPa)	Mean	SD
UG/30%E	1	1,367.87		
	2	1,477.74		
	3	1,414.51		
	4	1,488.73		
	5	1,325.85		
	6	1,441.34		
	7	1,480.60		
	8	1,405.99		
	9	1,347.57		
	10	1,445.79	1,419.60	57.64
UG/40%E	1	1,363.56		
	2	1,335.30		
	3	1,326.04		
	4	1,309.38		
	5	1,355.66		
	6	1,292.46		
	7	1,355.11		
	8	1,134.21		
	9	1,330.42		
	10	1,383.62	1,318.58	70.08
UG/50%E	1	1,287.87		
	2	1,347.04		
	3	1,356.76		
	4	1,294.47		
	5	1,361.78		
	6	1,351.97		
	7	1,350.05		
	8	1,298.64		
	9	1,237.86		
	10	1,188.76	1,307.52	58.03

Materials/ Groups	No	Flexural modulus (MPa)	Mean	SD
TR/NC	1	700.45		
	2	709.49		
	3	708.14		
	4	795.66		
	5	730.10		
	6	706.25		
	7	706.17		
	8	718.89		
	9	749.22		
	10	664.59	718.89	34.57
TR/PC1	1	626.57		
	2	703.92		
	3	731.82		
	4	698.08		
	5	687.73		
	6	672.19		
	7	711.44		
	8	697.28		
	9	714.62		
	10	703.48	694.71	28.74
TR/PC2	1	637.94		
	2	740.34		
	3	697.07		
	4	714.79		
	5	732.33		
	6	689.79		
	7	718.65		
	8	737.04		
	9	673.07		
	10	701.24	704.23	31.84

Materials/ Groups	No	Flexural modulus (MPa)	Mean	SD
TR/0%E	1	673.91		
	2	636.52		
	3	686.17		
	4	672.51		
	5	678.77		
	6	683.82		
	7	694.22		
	8	684.99		
	9	742.01		
	10	686.40	683.93	25.81
TR/10%E	1	744.01		
	2	733.57		
	3	692.92		
	4	765.56		
	5	748.01		
	6	624.64		
	7	805.05		
	8	811.53		
	9	717.18		
	10	752.28	739.48	54.07
TR/20%E	1	686.40		
	2	721.18		
	3	781.65		
	4	668.69		
	5	718.91		
	6	716.44		
	7	718.38		
	8	798.26		
	9	777.45		
	10	689.99	727.73	43.85

Materials/ Groups	No	Flexural modulus (MPa)	Mean	SD
TR/30%E	1	627.65		
	2	697.52		
	3	668.30		
	4	693.00		
	5	689.80		
	6	665.40		
	7	781.90		
	8	687.79		
	9	729.79		
	10	758.40	699.96	45.65
TR/40%E	1	687.68		
	2	650.66		
	3	686.83		
	4	694.21		
	5	714.27		
	6	686.05		
	7	696.28		
	8	708.59		
	9	749.87		
	10	675.03	694.95	26.11
TR/50%E	1	704.00		
	2	738.90		
	3	653.84		
	4	703.13		
	5	666.68		
	6	659.74		
	7	750.73		
	8	735.35		
	9	633.50		
	10	721.87	696.77	40.85

Table 15 The surface hardness of auto-polymerizing hard denture reline in each specimen.

Materials/ Groups	No	Surface hardness (VHN)	Mean	SD
UT/NC	1	15.09		
	2	15.06		
	3	15.04		
	4	15.34		
	5	14.97		
	6	15.12		
	7	15.11		
	8	15.16		
	9	14.93		
	10	15.15	15.10	0.11
UT/PC1	1	14.48		
	2	14.63		
	3	14.94		
	4	14.77		
	5	15.39		
	6	15.11		
	7	14.99		
	8	14.97		
	9	15.02		
	10	15.25	14.95	0.28
UT/PC2	1	15.00		
	2	14.88		
	3	15.20		
	4	15.38		
	5	15.15		
	6	15.24		
	7	15.15		
	8	15.13		
	9	15.19		
	10	15.23	15.16	0.14

Materials/ Groups	No	Surface hardness (VHN)	Mean	SD
UT/0%E	1	14.61		
	2	15.12		
	3	15.07		
	4	15.06		
	5	15.00		
	6	15.06		
	7	15.29		
	8	15.00		
	9	14.77		
	10	15.25	15.02	0.20
UT/10%E	1	14.85		
	2	15.07		
	3	15.22		
	4	14.58		
	5	15.30		
	6	15.22		
	7	15.14		
	8	14.67		
	9	14.88		
	10	15.20	15.01	0.25
UT/20%E	1	15.08		
	2	14.86		
	3	15.16		
	4	15.01		
	5	15.13		
	6	14.98		
	7	14.85		
	8	14.96		
	9	14.89		
	10	15.16	15.01	0.12

Materials/ Groups	No	Surface hardness (VHN)	Mean	SD
UT/30%E	1	15.01		
	2	14.90		
	3	14.98		
	4	15.11		
	5	14.99		
	6	15.23		
	7	15.10		
	8	14.94		
	9	14.90		
	10	14.93	15.01	0.11
UT/40%E	1	14.90		
	2	14.85		
	3	14.80		
	4	14.70		
	5	14.80		
	6	15.18		
	7	14.74		
	8	14.96		
	9	14.91		
	10	15.17	14.90	0.16
UT/50%E	1	14.92		
	2	15.11		
	3	14.95		
	4	14.83		
	5	14.76		
	6	15.02		
	7	15.20		
	8	14.92		
	9	15.04		
	10	15.57	15.03	0.23

Materials/ Groups	No	Surface hardness (VHN)	Mean	SD
KL/NC	1	9.28		
	2	9.52		
	3	9.56		
	4	9.67		
	5	9.56		
	6	9.71		
	7	9.42		
	8	9.59		
	9	9.58		
	10	9.31	9.52	0.14
KL/PC1	1	9.68		
	2	9.54		
	3	9.60		
	4	9.79		
	5	9.18		
	6	9.34		
	7	9.23		
	8	9.77		
	9	9.31		
	10	9.50	9.49	0.22
KL/PC2	1	9.45		
	2	9.97		
	3	9.57		
	4	9.77		
	5	9.28		
	6	9.45		
	7	9.81		
	8	9.64		
	9	9.54		
	10	9.47	9.60	0.21

Materials/ Groups	No	Surface hardness (VHN)	Mean	SD
KL/0%E	1	9.78		
	2	9.89		
	3	9.87		
	4	9.84		
	5	9.49		
	6	9.54		
	7	9.77		
	8	9.85		
	9	9.36		
	10	9.43	9.68	0.21
KL/10%E	1	9.78		
	2	9.43		
	3	9.26		
	4	9.79		
	5	9.77		
	6	9.64		
	7	9.72		
	8	9.80		
	9	9.91		
	10	9.78	9.69	0.20
KL/20%E	1	9.76		
	2	9.79		
	3	9.73		
	4	9.83		
	5	9.84		
	6	9.47		
	7	9.63		
	8	9.70		
	9	9.37		
	10	9.95	9.71	0.18

Materials/ Groups	No	Surface hardness (VHN)	Mean	SD
KL/30%E	1	9.82		
	2	9.80		
	3	9.75		
	4	9.64		
	5	9.66		
	6	9.51		
	7	9.71		
	8	9.83		
	9	9.52		
	10	9.99	9.72	0.15
KL/40%E	1	9.48		
	2	9.71		
	3	9.79		
	4	9.74		
	5	9.68		
	6	9.34		
	7	9.28		
	8	9.67		
	9	9.38		
	10	9.90	9.60	0.21
KL/50%E	1	9.82		
	2	9.53		
	3	9.89		
	4	9.22		
	5	9.60		
	6	9.74		
	7	9.80		
	8	9.43		
	9	9.30		
	10	9.26	9.56	0.25

Materials/ Groups	No	Surface hardness (VHN)	Mean	SD
UG/NC	1	13.21		
	2	13.36		
	3	13.27		
	4	13.69		
	5	13.81		
	6	13.71		
	7	13.61		
	8	13.84		
	9	13.56		
	10	13.56	13.56	0.22
UG/PC1	1	13.51		
	2	13.43		
	3	13.59		
	4	13.61		
	5	13.48		
	6	13.22		
	7	13.79		
	8	13.24		
	9	13.97		
	10	14.03	13.59	0.28
UG/PC2	1	13.52		
	2	13.35		
	3	13.55		
	4	13.31		
	5	13.23		
	6	13.46		
	7	13.49		
	8	13.29		
	9	13.43		
	10	13.61	13.43	0.12

Materials/ Groups	No	Surface hardness (VHN)	Mean	SD
UG/0%E	1	13.72		
	2	13.55		
	3	13.55		
	4	13.87		
	5	13.68		
	6	13.31		
	7	13.53		
	8	13.46		
	9	13.62		
	10	13.55	13.58	0.15
UG/10%E	1	13.70		
	2	13.10		
	3	13.60		
	4	13.31		
	5	13.77		
	6	13.21		
	7	13.36		
	8	13.27		
	9	13.38		
	10	13.36	13.41	0.22
UG/20%E	1	13.52		
	2	13.44		
	3	13.74		
	4	13.18		
	5	13.19		
	6	13.22		
	7	13.34		
	8	13.20		
	9	13.17		
	10	13.46	13.35	0.19

Materials/ Groups	No	Surface hardness (VHN)	Mean	SD
UG/30%E	1	13.71		
	2	13.78		
	3	13.75		
	4	13.98		
	5	13.60		
	6	13.62		
	7	13.32		
	8	13.36		
	9	13.15		
	10	13.30	13.56	0.26
UG/40%E	1	13.49		
	2	13.24		
	3	13.17		
	4	13.58		
	5	13.16		
	6	13.55		
	7	13.49		
	8	13.42		
	9	13.21		
	10	13.73	13.40	0.20
UG/50%E	1	13.10		
	2	13.42		
	3	13.60		
	4	13.33		
	5	13.38		
	6	13.39		
	7	13.54		
	8	13.10		
	9	13.26		
	10	13.39	13.35	0.16

Materials/ Groups	No	Surface hardness (VHN)	Mean	SD
TR/NC	1	9.75		
	2	10.29		
	3	10.22		
	4	10.04		
	5	9.91		
	6	9.96		
	7	10.12		
	8	10.13		
	9	10.23		
	10	10.23	10.09	0.17
TR/PC1	1	10.02		
	2	10.19		
	3	9.81		
	4	10.00		
	5	10.04		
	6	9.89		
	7	9.89		
	8	9.95		
	9	10.00		
	10	10.08	9.99	0.11
TR/PC2	1	9.96		
	2	10.34		
	3	9.91		
	4	9.84		
	5	9.98		
	6	10.00		
	7	9.98		
	8	10.17		
	9	10.10		
	10	10.04	10.03	0.14

Materials/ Groups	No	Surface hardness (VHN)	Mean	SD
TR/0%E	1	10.26		
	2	10.33		
	3	10.23		
	4	10.20		
	5	10.01		
	6	10.05		
	7	10.02		
	8	10.01		
	9	10.06		
	10	10.21	10.14	0.12
TR/10%E	1	9.97		
	2	10.04		
	3	9.85		
	4	10.04		
	5	9.99		
	6	10.06		
	7	10.06		
	8	9.92		
	9	9.75		
	10	10.00	9.97	0.10
TR/20%E	1	10.04		
	2	10.08		
	3	10.04		
	4	10.00		
	5	10.04		
	6	10.10		
	7	9.98		
	8	10.12		
	9	10.18		
	10	10.01	10.06	0.06

Materials/ Groups	No	Surface hardness (VHN)	Mean	SD
TR/30%E	1	9.74		
	2	9.91		
	3	9.96		
	4	10.20		
	5	10.18		
	6	10.06		
	7	9.88		
	8	10.23		
	9	10.27		
	10	10.18	10.06	0.18
TR/40%E	1	9.76		
	2	10.17		
	3	10.09		
	4	10.16		
	5	10.06		
	6	10.05		
	7	10.05		
	8	9.98		
	9	10.12		
	10	10.22	10.07	0.13
TR/50%E	1	10.04		
	2	10.22		
	3	10.28		
	4	9.92		
	5	9.93		
	6	9.87		
	7	9.71		
	8	9.75		
	9	9.96		
	10	10.00	9.97	0.18

Table 16 The water sorption of auto-polymerizing hard denture reline in each specimen.

Materials/ Groups	No	Water sorption ($\mu g/mm^3$)	Mean	SD
UT/NC	1	15.62		
	2	14.97		
	3	14.47		
	4	14.91		
	5	14.80		
	6	15.03		
	7	15.57		
	8	14.90		
	9	15.02		
	10	15.39	15.07	0.36
UT/PC1	1	15.98		
	2	16.09		
	3	15.72		
	4	15.87		
	5	14.78		
	6	14.82		
	7	15.58		
	8	15.15		
	9	15.65		
	10	14.79	15.44	0.51
UT/PC2	1	15.68		
	2	14.55		
	3	15.07		
	4	14.82		
	5	14.61		
	6	14.70		
	7	15.21		
	8	15.69		
	9	14.57		
	10	15.64	15.05	0.47

Materials/ Groups	No	Water sorption ($\mu g/mm^3$)	Mean	SD
UT/0%E	1	14.35		
	2	14.95		
	3	14.39		
	4	15.65		
	5	15.10		
	6	14.70		
	7	15.05		
	8	15.23		
	9	15.42		
	10	15.29	15.01	0.43
UT/10%E	1	15.58		
	2	15.77		
	3	15.76		
	4	15.63		
	5	15.44		
	6	15.80		
	7	14.86		
	8	15.52		
	9	14.71		
	10	15.28	15.44	0.38
UT/20%E	1	14.64		
	2	15.24		
	3	15.05		
	4	14.57		
	5	15.04		
	6	14.88		
	7	15.60		
	8	14.42		
	9	15.46		
	10	15.39	15.03	0.40

Materials/ Groups	No	Water sorption ($\mu g/mm^3$)	Mean	SD
UT/30%E	1	15.42		
	2	15.57		
	3	15.36		
	4	15.67		
	5	14.80		
	6	15.73		
	7	15.79		
	8	14.69		
	9	14.93		
	10	14.95	15.29	0.41
UT/40%E	1	15.75		
	2	16.42		
	3	16.37		
	4	16.55		
	5	15.85		
	6	15.86		
	7	15.71		
	8	16.19		
	9	15.64		
	10	16.05	16.04	0.33
UT/50%E	1	16.21		
	2	16.38		
	3	16.58		
	4	16.02		
	5	16.43		
	6	16.48		
	7	16.79		
	8	16.76		
	9	16.32		
	10	15.61	16.36	0.35

Materials/ Groups	No	Water sorption ($\mu g/mm^3$)	Mean	SD
KL/NC	1	15.26		
	2	14.13		
	3	13.66		
	4	15.00		
	5	16.00		
	6	15.03		
	7	17.68		
	8	14.80		
	9	18.80		
	10	16.21	15.66	1.58
KL/PC1	1	15.27		
	2	14.54		
	3	15.70		
	4	14.66		
	5	16.29		
	6	18.28		
	7	16.63		
	8	15.41		
	9	16.84		
	10	15.26	15.89	1.14
KL/PC2	1	14.97		
	2	16.15		
	3	16.48		
	4	13.75		
	5	15.37		
	6	15.67		
	7	15.20		
	8	13.66		
	9	16.03		
	10	15.33	15.26	0.94

Materials/ Groups	No	Water sorption ($\mu g/mm^3$)	Mean	SD
KL/0%E	1	17.13		
	2	16.60		
	3	18.10		
	4	18.32		
	5	17.17		
	6	17.13		
	7	15.62		
	8	13.59		
	9	16.47		
	10	17.15	16.73	1.35
KL/10%E	1	16.95		
	2	16.02		
	3	15.17		
	4	16.33		
	5	17.72		
	6	15.86		
	7	15.71		
	8	15.99		
	9	16.21		
	10	17.97	16.39	0.89
KL/20%E	1	16.54		
	2	15.24		
	3	16.26		
	4	16.46		
	5	14.86		
	6	17.24		
	7	16.49		
	8	16.59		
	9	15.50		
	10	17.21	16.24	0.80

Materials/ Groups	No	Water sorption ($\mu g/mm^3$)	Mean	SD
KL/30%E	1	13.13		
	2	12.89		
	3	10.45		
	4	12.88		
	5	12.83		
	6	12.17		
	7	12.22		
	8	13.41		
	9	12.14		
	10	11.98	12.41	0.84
KL/40%E	1	13.43		
	2	10.46		
	3	12.50		
	4	13.61		
	5	14.56		
	6	14.14		
	7	13.91		
	8	12.81		
	9	11.46		
	10	12.87	12.97	1.26
KL/50%E	1	13.93		
	2	12.77		
	3	12.90		
	4	12.70		
	5	13.42		
	6	12.20		
	7	11.81		
	8	13.94		
	9	12.24		
	10	11.66	12.76	0.81

Materials/ Groups	No	Water sorption ($\mu g/mm^3$)	Mean	SD
UG/NC	1	9.81		
	2	9.37		
	3	9.45		
	4	10.37		
	5	9.79		
	6	10.22		
	7	10.02		
	8	10.04		
	9	10.11		
	10	9.36	9.85	0.36
UG/PC1	1	10.53		
	2	10.60		
	3	9.99		
	4	10.54		
	5	10.40		
	6	9.60		
	7	10.75		
	8	10.44		
	9	11.03		
	10	9.89	10.38	0.43
UG/PC2	1	10.31		
	2	9.70		
	3	10.10		
	4	9.0220		
	5	9.63		
	6	10.59		
	7	11.28		
	8	9.34		
	9	10.72		
	10	10.26	10.09	0.68

Materials/ Groups	No	Water sorption ($\mu g/mm^3$)	Mean	SD
UG/0%E	1	9.09		
	2	9.61		
	3	8.34		
	4	9.71		
	5	8.91		
	6	8.90		
	7	8.60		
	8	9.91		
	9	9.64		
	10	9.72	9.24	0.54
UG/10%E	1	8.87		
	2	8.26		
	3	9.61		
	4	8.26		
	5	9.30		
	6	9.36		
	7	9.71		
	8	9.55		
	9	9.72		
	10	9.65	9.23	0.57
UG/20%E	1	9.41		
	2	9.32		
	3	8.49		
	4	8.76		
	5	9.37		
	6	9.21		
	7	9.15		
	8	10.30		
	9	9.38		
	10	8.93	9.23	0.48

Materials/ Groups	No	Water sorption ($\mu g/mm^3$)	Mean	SD
UG/30%E	1	8.96		
	2	8.88		
	3	9.53		
	4	9.04		
	5	9.95		
	6	9.31		
	7	8.88		
	8	9.02		
	9	9.69		
	10	9.17	9.24	0.37
UG/40%E	1	10.15		
	2	10.63		
	3	9.79		
	4	9.51		
	5	9.52		
	6	9.05		
	7	9.32		
	8	9.66		
	9	10.15		
	10	9.51	9.73	0.46
UG/50%E	1	10.66		
	2	9.77		
	3	10.70		
	4	9.06		
	5	9.90		
	6	10.22		
	7	8.97		
	8	10.42		
	9	9.45		
	10	10.31	9.95	0.63

Materials/ Groups	No	Water sorption ($\mu g/mm^3$)	Mean	SD
TR/NC	1	15.89		
	2	13.38		
	3	13.81		
	4	11.39		
	5	16.54		
	6	13.56		
	7	11.71		
	8	12.81		
	9	15.52		
	10	13.77	13.84	1.71
TR/PC1	1	16.97		
	2	14.37		
	3	15.60		
	4	15.34		
	5	14.83		
	6	13.42		
	7	13.10		
	8	11.51		
	9	15.93		
	10	12.62	14.37	1.69
TR/PC2	1	16.50		
	2	14.29		
	3	13.01		
	4	14.44		
	5	14.76		
	6	11.98		
	7	12.18		
	8	12.33		
	9	12.86		
	10	12.77	13.51	1.44

Materials/ Groups	No	Water sorption ($\mu g/mm^3$)	Mean	SD
TR/0%E	1	13.01		
	2	12.36		
	3	12.97		
	4	14.09		
	5	14.74		
	6	13.76		
	7	12.48		
	8	13.89		
	9	14.96		
	10	14.42	13.67	0.92
TR/10%E	1	14.79		
	2	11.99		
	3	16.54		
	4	11.74		
	5	12.10		
	6	12.87		
	7	12.69		
	8	12.13		
	9	14.32		
	10	16.38	13.55	1.83
TR/20%E	1	14.31		
	2	12.20		
	3	15.02		
	4	12.47		
	5	15.24		
	6	13.31		
	7	14.96		
	8	15.11		
	9	12.06		
	10	12.72	13.74	1.32

Materials/ Groups	No	Water sorption ($\mu g/mm^3$)	Mean	SD
TR/30%E	1	15.40		
	2	14.95		
	3	12.26		
	4	13.20		
	5	15.42		
	6	11.89		
	7	14.53		
	8	13.43		
	9	12.22		
	10	14.08	13.74	1.34
TR/40%E	1	11.92		
	2	12.59		
	3	12.30		
	4	11.19		
	5	12.76		
	6	11.87		
	7	11.89		
	8	13.53		
	9	12.47		
	10	10.97	12.15	0.75
TR/50%E	1	12.13		
	2	11.39		
	3	13.69		
	4	11.61		
	5	11.24		
	6	13.42		
	7	11.50		
	8	11.15		
	9	13.40		
	10	10.92	12.04	1.06

Table 17 The water solubility of auto-polymerizing hard denture reline in each specimen.

Materials/ Groups	No	Water solubility ($\mu g/mm^3$)	Mean	SD
UT/NC	1	0.84		
	2	1.19		
	3	1.25		
	4	1.52		
	5	0.67		
	6	0.56		
	7	0.40		
	8	0.74		
	9	0.85		
	10	0.55	0.86	0.36
UT/PC1	1	0.90		
	2	1.07		
	3	0.71		
	4	0.71		
	5	0.73		
	6	0.56		
	7	0.36		
	8	0.88		
	9	0.37		
	10	1.27	0.76	0.29
UT/PC2	1	1.50		
	2	1.48		
	3	0.45		
	4	0.96		
	5	0.82		
	6	0.69		
	7	0.93		
	8	0.86		
	9	0.67		
	10	0.77	0.91	0.34

Materials/ Groups	No	Water solubility ($\mu g/mm^3$)	Mean	SD
UT/0%E	1	0.46		
	2	0.26		
	3	0.37		
	4	0.90		
	5	0.80		
	6	1.13		
	7	0.78		
	8	1.25		
	9	1.03		
	10	1.17	0.82	0.35
UT/10%E	1	0.56		
	2	0.36		
	3	1.05		
	4	0.77		
	5	0.67		
	6	0.67		
	7	0.83		
	8	0.68		
	9	0.66		
	10	0.79	0.70	0.18
UT/20%E	1	0.80		
	2	0.74		
	3	0.78		
	4	1.45		
	5	0.62		
	6	0.20		
	7	0.47		
	8	0.31		
	9	0.78		
	10	0.86	0.70	0.35

Materials/ Groups	No	Water solubility ($\mu g/mm^3$)	Mean	SD
UT/30%E	1	0.97		
	2	0.96		
	3	0.79		
	4	0.90		
	5	0.78		
	6	0.97		
	7	0.75		
	8	0.46		
	9	0.59		
	10	0.58	0.78	0.18
UT/40%E	1	0.91		
	2	0.83		
	3	1.35		
	4	1.24		
	5	1.55		
	6	1.27		
	7	1.71		
	8	1.79		
	9	2.32		
	10	1.26	1.42	0.44
UT/50%E	1	1.79		
	2	2.48		
	3	1.99		
	4	1.99		
	5	2.66		
	6	3.24		
	7	2.53		
	8	2.37		
	9	2.67		
	10	1.94	2.37	0.45

Materials/ Groups	No	Water solubility ($\mu g/mm^3$)	Mean	SD
KL/NC	1	2.75		
	2	2.68		
	3	3.35		
	4	3.21		
	5	3.91		
	6	2.86		
	7	5.17		
	8	2.69		
	9	5.12		
	10	3.78	3.55	0.95
KL/PC1	1	3.82		
	2	3.71		
	3	3.39		
	4	3.83		
	5	3.07		
	6	3.17		
	7	4.11		
	8	2.22		
	9	3.74		
	10	3.84	3.49	0.55
KL/PC2	1	4.33		
	2	3.75		
	3	3.57		
	4	3.92		
	5	4.56		
	6	5.02		
	7	3.60		
	8	5.73		
	9	4.88		
	10	3.79	4.31	0.72

Materials/ Groups	No	Water solubility ($\mu g/mm^3$)	Mean	SD
KL/0%E	1	2.80		
	2	4.82		
	3	3.16		
	4	5.32		
	5	4.01		
	6	4.13		
	7	3.75		
	8	5.38		
	9	4.84		
	10	3.84	4.20	0.87
KL/10%E	1	3.24		
	2	3.68		
	3	3.37		
	4	5.76		
	5	4.86		
	6	4.32		
	7	3.88		
	8	5.78		
	9	4.12		
	10	4.79	4.38	0.90
KL/20%E	1	3.56		
	2	4.23		
	3	4.53		
	4	4.12		
	5	3.03		
	6	5.35		
	7	5.88		
	8	3.68		
	9	3.06		
	10	3.52	4.10	0.94

Materials/ Groups	No	Water solubility ($\mu g/mm^3$)	Mean	SD
KL/30%E	1	1.60		
	2	1.95		
	3	0.55		
	4	1.71		
	5	1.25		
	6	1.42		
	7	1.77		
	8	2.09		
	9	1.24		
	10	1.01	1.46	0.47
KL/40%E	1	1.13		
	2	0.53		
	3	0.96		
	4	1.94		
	5	1.93		
	6	1.20		
	7	2.51		
	8	1.72		
	9	0.66		
	10	0.95	1.35	0.64
KL/50%E	1	1.64		
	2	1.94		
	3	1.22		
	4	1.10		
	5	0.91		
	6	1.12		
	7	1.32		
	8	1.90		
	9	0.56		
	10	1.50	1.32	0.43

Materials/ Groups	No	Water solubility ($\mu g/mm^3$)	Mean	SD
UG/NC	1	3.79		
	2	3.86		
	3	4.92		
	4	4.63		
	5	4.29		
	6	3.83		
	7	3.34		
	8	3.29		
	9	3.56		
	10	5.44	4.09	0.71
UG/PC1	1	4.15		
	2	3.80		
	3	5.90		
	4	3.84		
	5	3.37		
	6	3.20		
	7	4.28		
	8	3.23		
	9	3.02		
	10	5.99	4.08	1.07
UG/PC2	1	5.01		
	2	4.76		
	3	5.54		
	4	4.26		
	5	3.72		
	6	2.06		
	7	5.14		
	8	3.43		
	9	4.40		
	10	3.36	4.17	1.04

Materials/ Groups	No	Water solubility ($\mu g/mm^3$)	Mean	SD
UG/0%E	1	3.09		
	2	2.73		
	3	3.35		
	4	3.40		
	5	3.30		
	6	3.85		
	7	3.26		
	8	2.92		
	9	3.40		
	10	2.79	3.21	0.34
UG/10%	1	2.99		
	2	2.04		
	3	2.89		
	4	2.21		
	5	4.18		
	6	1.89		
	7	4.76		
	8	4.20		
	9	1.97		
	10	2.71	2.98	1.05
UG/20%E	1	5.93		
	2	1.61		
	3	1.78		
	4	2.49		
	5	2.68		
	6	2.64		
	7	3.39		
	8	3.47		
	9	2.97		
	10	3.49	3.04	1.21

Materials/ Groups	No	Water solubility ($\mu g/mm^3$)	Mean	SD
UG/30%E	1	3.44		
	2	2.62		
	3	3.34		
	4	5.59		
	5	3.35		
	6	3.25		
	7	1.54		
	8	1.04		
	9	1.96		
	10	1.60	2.77	1.33
UG/40%	1	2.23		
	2	2.39		
	3	3.84		
	4	3.58		
	5	1.64		
	6	2.22		
	7	1.21		
	8	5.24		
	9	1.81		
	10	1.23	2.54	1.29
UG/50%E	1	2.33		
	2	2.97		
	3	3.54		
	4	2.68		
	5	1.67		
	6	1.72		
	7	2.89		
	8	2.45		
	9	1.73		
	10	3.02	2.50	0.64

Materials/ Groups	No	Water solubility ($\mu g/mm^3$)	Mean	SD
TR/NC	1	7.35		
	2	7.21		
	3	5.05		
	4	7.42		
	5	8.64		
	6	10.51		
	7	7.97		
	8	8.37		
	9	7.86		
	10	8.33	7.87	1.37
TR/PC1	1	9.38		
	2	9.21		
	3	10.10		
	4	9.02		
	5	10.05		
	6	9.34		
	7	7.01		
	8	6.47		
	9	5.85		
	10	7.30	8.37	1.56
TR/PC2	1	8.67		
	2	7.40		
	3	6.46		
	4	6.39		
	5	8.81		
	6	5.54		
	7	6.14		
	8	5.87		
	9	7.63		
	10	6.76	6.97	1.13

Materials/ Groups	No	Water solubility ($\mu g/mm^3$)	Mean	SD
TR/0%E	1	4.07		
	2	4.75		
	3	5.27		
	4	7.91		
	5	7.46		
	6	7.11		
	7	5.43		
	8	7.57		
	9	8.28		
	10	8.93	6.68	1.66
TR/10%E	1	7.54		
	2	6.38		
	3	5.69		
	4	7.19		
	5	5.70		
	6	5.00		
	7	6.32		
	8	7.68		
	9	8.17		
	10	10.01	6.97	1.47
TR/20%E	1	7.99		
	2	4.13		
	3	7.75		
	4	5.69		
	5	8.26		
	6	8.22		
	7	8.83		
	8	6.81		
	9	4.87		
	10	6.47	6.90	1.59

Materials/ Groups	No	Water solubility ($\mu g/mm^3$)	Mean	SD
TR/30%E	1	6.00		
	2	7.87		
	3	5.64		
	4	5.89		
	5	8.33		
	6	5.00		
	7	8.01		
	8	6.43		
	9	6.71		
	10	5.51	6.54	1.16
TR/40%E	1	2.49		
	2	3.35		
	3	2.03		
	4	1.42		
	5	4.19		
	6	3.51		
	7	1.67		
	8	5.03		
	9	3.79		
	10	2.21	2.97	1.18
TR/50%E	1	2.23		
	2	2.34		
	3	2.61		
	4	1.18		
	5	1.09		
	6	2.66		
	7	2.09		
	8	2.18		
	9	2.01		
	10	1.64	2.00	0.54

VITA

Miss Sutasinee Sootornwipath was born on Friday 27th December 1985 in Suphanburi, Thailand. She graduated high-school from Sa-Nguan Ying school, Suphanburi. Then, she graduated the degree in Doctor of Dental Surgery (D.D.S.) from Faculty of Dentistry, Naresuan University in 2011. After that, she has worked as a dentist at Khok Samrong Hospital, Lopburi for 2 years and U-Thong Hospital, Suphanburi for 1 year. Then she is permitted to attend the course of Master of Science in Prosthodontics, Department of Prosthodontics, Faculty of Dentistry, Chulalongkorn University.