

Research Article

The Mechanical Properties of a Poly(methyl methacrylate) Denture Base Material Modified with Dimethyl Itaconate and Di-*n*-butyl Itaconate

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This study investigates a wide range of clinically relevant mechanical properties of poly(methyl methacrylate) (PMMA) denture base materials modified with di-methyl itaconate (DMI) and di-*n*-butyl itaconate (DBI) in order to compare them to a commercial PMMA denture base material. The commercial denture base formulation was modified with DMI and DBI by replacing up to 10 wt% of methyl methacrylate (MMA) monomer. The specimens were prepared by standard bath curing process. The influence of the itaconate content on hardness, impact strength, tensile, and thermal and dynamic mechanical properties was investigated. It is found that the addition of di-*n*-alkyl itaconates gives homogenous blends that show decreased glass transition temperature, as well as decrease in storage modulus, ultimate tensile strength, and impact fracture resistance with increase in the itaconate content. The mean values of surface hardness show no significant change with the addition of itaconates. The magnitude of the measured values indicates that the poly(methyl methacrylate) (PMMA) denture base material modified with itaconates could be developed into a less toxic, more environmentally and patient friendly product than commercial pure PMMA denture base material.

1. Introduction

One of the most important unmodified acrylic materials for dental applications is poly(methyl methacrylate) (PMMA). Discovered and commercialized many years ago, PMMA is one of the most widely used industrial polymeric materials and still remains an active material for research at the cutting edges of science. Because of its good biocompatibility, reliability, dimensional stability, absence of taste, odor, tissue irritation and toxicity [1], teeth adhesion [2], insolubility in body fluids, relative ease of manipulation, good aesthetic appearance [3], and color stability [4], PMMA based materials are widely used as biomaterials. Nowadays, PMMA finds applications not only in dentistry but also in areas such as transparent glass substitutes, interior design, transparent dielectric films [5], acrylic paints [6], and microcellular foams

[7]. Still, one of the most attractive applications of PMMA based materials is in various biomedical applications such as intraocular lenses [8], bone cement in orthopedic surgery [9], and removable partial denture [10].

Although PMMA denture base materials have a lot of qualities, they are often subject to intense criticism because of their inherent drawbacks such as residual monomer toxicity and its effect on the oral tissues (Figure 1) [11–15], mechanical properties that are not always perfect [16], and susceptibility to distortion. The cumulative effect of these properties may lead to residual monomer leakage into adjacent oral tissues and the generation of cracks and the other structural damage of denture base that, apart from leading to denture base breakage, can also form a point of entry for various bacteria, yeasts, and moulds.



FIGURE 1: Inflammation of the oral mucosal tissues after removal of the prosthesis.

In order to overcome the drawbacks of pure PMMA denture base material we have investigated the possibility of modifying the commercial formulations with the derivatives of itaconic acid. The similarity of di-itaconates and corresponding methacrylates makes polymers based on itaconic acid an interesting alternative to the methacrylic acid based materials. The advantage of itaconic over methacrylic acid is the fact that the itaconic acid is a natural product, and its industrial sized production is based on renewable resources [17, 18] rendering it significantly less toxic [19]. The replacement of methacrylates, partly or completely, results also in significantly less toxic residual monomer, which in turn would not cause the inflammation of the tissue that comes into contact with the denture. As a dibasic acid, itaconic acid gives a larger scope of possible ester substituents than methacrylic acid. Itaconic acid derivatives are cocomponents in various formulations in controlled-drug release systems [20–24], paints and coatings [25–27], resist compositions [28, 29], contact lenses [30] ocular lenses [31], and personal care compositions [32]. In dentistry, itaconic acid and itaconates have been used in glass-ionomer cements [33–35].

The copolymers of methyl methacrylate and dialkyl itaconates have been investigated in many studies because of their structural similarity. The kinetics of copolymerization has been thoroughly examined [36, 37]. Fernández-García and Madruga examined the influence of copolymer composition on the glass transition temperature [38]. They have come to the conclusion that the glass transition temperature of copolymer decreased with the increase in the amount of itaconates in the copolymer as well as in the chain length of ester alkyl groups. The same authors have examined the thermal stability of methyl methacrylate copolymers with dialkyl itaconates and found that the relative thermal stability increased with the increase in methyl methacrylate copolymer molar fraction, following a trend similar to the glass transition temperature variation [39].

In our previous study we have investigated water sorption and residual monomer content in the PMMA denture base materials modified with DMI and DBI [40]. We have found that addition of itaconates reduced water sorption and greatly decreased residual MMA content. The highest reduction was for the material where MMA was replaced by 10 wt% of DMI and reduction was 88% while in the other

cases the reduction ranged from 49% to 84%. As residual MMA has been recognized as the main cause of allergic sensitization, contact dermatitis, and tissue inflammation, its reduction significantly improved the applicative properties and biocompatibility of the PMMA denture base material. It gave us a reason to continue investigation of such materials.

To the best of our knowledge, there are no literature data that evaluate mechanical properties of copolymers of dialkyl itaconates and methyl methacrylate. Taken that it is not only the lesser toxicity and the “greener” properties of the material that are important for the enhanced qualities of denture base, in this paper we investigated the mechanical properties of commercial denture base material modified with dimethyl itaconate (DMI) and di-*n*-butyl itaconate (DBI).

2. Experimental

2.1. Materials. The commercial dental resin Biocryl was kindly supplied by Galenika AD, Belgrade, Serbia, and was used as received. The liquid component consists of MMA as monomer and ethylene glycol dimethacrylate as a crosslinker. The powder component consists of prepolymerized beads of PMMA coated with dibenzoyl peroxide as an initiator. In our previous study the powder component of Biocryl has been characterized and its basic physicochemical properties are $M_n = 1.27 \times 10^5 \text{ g mol}^{-1}$, $M_w = 3.82 \times 10^5 \text{ g mol}^{-1}$, $PI = 3.01$, monomer content 4.3 wt%, and average particle size of $55 \mu\text{m}$ [41]. DMI and itaconic acid were obtained from Fluka. Butyl alcohol, sulphuric acid, and hydroquinone were supplied by Sigma-Aldrich. DMI was purified by conventional method [36]. DBI was synthesized as described previously [42] and characterized by infrared spectroscopy.

2.2. Sample Preparation. The liquid component of the denture base material, Biocryl, was mixed with selected di-*n*-alkyl itaconate. The amounts of DMI and DBI in the mixture were 0, 2.5, 5.0, 7.5, and 10 wt%, with respect to the total mass of formulation used (Table 1).

The mixed resins were packed into teflon moulds designed to produce a bar specimens measuring $60 \text{ mm} \times 12 \text{ mm} \times 3 \text{ mm}$ for the dynamic mechanical thermal analysis (DMTA) measurements and $80 \text{ mm} \times 10 \text{ mm} \times 4 \text{ mm}$ for the tensile (Figure 2) and the impact strength (Figure 3)

TABLE I: Feed composition of synthesized denture base materials.

Sample	Feed composition, wt%			
	Biocryl, powder	Biocryl, liquid	DMI	DBI
PMMA	66.0	34.0	/	/
2.5 DMI	64.5	33.0	2.5	/
5.0 DMI	63.0	32.0	5.0	/
7.5 DMI	61.5	31.0	7.5	/
10 DMI	59.5	30.5	10	/
2.5 DBI	65.0	32.5	/	2.5
5.0 DBI	64.0	31.0	/	5.0
7.5 DBI	63.0	29.5	/	7.5
10 DBI	62.5	27.5	/	10

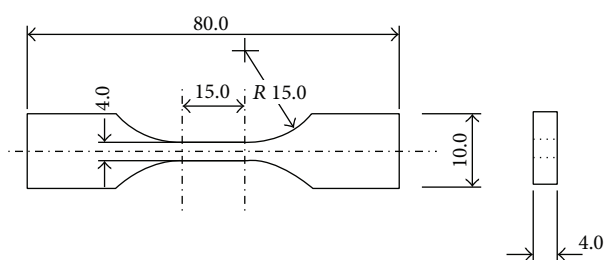


FIGURE 2: Geometry of the tensile test specimen used in this investigation according to ASTM D882.

measurements. The discs measuring 25 mm in diameter and 4 mm thickness were prepared for the hardness measurement. The moulds were then clamped tightly and placed in a thermostatically controlled water bath (NEIB-8, Progen Scientific, London, UK). The denture base acrylic resins were polymerized for 30 minutes at 100°C by immersing the moulds into water bath. Specimens were examined visually without magnification and were required to demonstrate no porosity. Five samples were prepared under the same polymerization conditions and were used for all measurements.

The homopolymers of poly(dimethyl itaconate) (PDMI) and poly(di-*n*-butyl itaconate) (PDBI) were prepared at 50°C in toluene solution (3 mol dm⁻³) using AIBN (1.5 × 10⁻² mol dm⁻³) as initiator.

2.3. Fourier Transform Infrared Spectroscopy (FTIR). The spectra were recorded on Bomem MB 100 Fourier transform infrared spectrometer (FTIR), Hartmann & Braun, Canada. The 10 mg from each polymerized sample was finely ground with 100 mg of KBr. The mixtures were transferred to the pneumatic pellet-making press and left for three minutes under pressure. The KBr discs with the polymer sample were placed in the IR cell and the spectrum was recorded. All spectra were obtained from 10 scans at resolution of 4 cm⁻¹.

2.4. Scanning Electron Microscopy (SEM). Representative SEM micrographs were taken from the fractured surfaces. The fractured surfaces were carefully cut from the denture

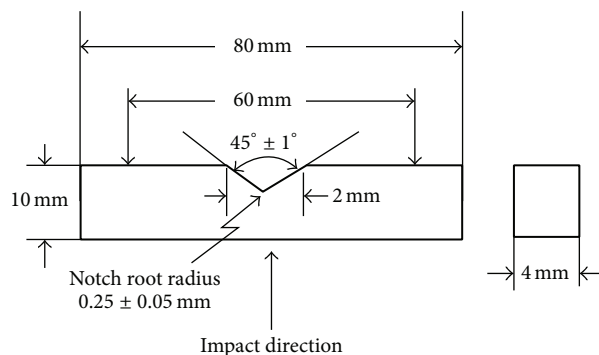


FIGURE 3: Geometry of the Charpy V-notch impact specimen used in this investigation [43].

teeth, stuck to a specimen holder (Plano, Wetzlar, Germany), and sputtered with platinum. SEM evaluation was performed with a scanning electron microscope (type FEI XL30 ESEM FEG, FEI Company, Eindhoven, Netherlands) at an accelerating voltage of 5–10 kV.

2.5. Differential Scanning Calorimetry (DSC). The thermal properties of copolymers were studied by differential scanning calorimetry (DSC) using Perkin Elmer DSC-2 thermal analyzer under nitrogen (flow rate 26 mL min⁻¹). For measurement of the glass transition temperature (T_g), 10–20 mg of sample was placed in an aluminum holder and heated from 50°C to 200°C. The heating rate was set to 20°C min⁻¹. In order to erase the thermal history of the sample all calculations were done using the data acquired from second cycle. The glass transition temperature was estimated as the temperature at midpoint of the line drawn between the temperature of intersection of the initial tangent with the tangent drawn through the point of inflection of the trace and the temperature of intersection of the tangent drawn through the point of inflection with the final tangent.

2.6. Dynamic Mechanical Thermal Analysis (DMTA). Dynamic mechanical thermal analysis tests were performed on a mechanical spectrometer Rheometrics 605 in bending mode. A frequency of 1 Hz and strain of 0.3% were applied. A temperature range of 25°C–180°C and a heating rate of 3°C min⁻¹ were selected to cover mouth temperature and the materials likely glass transition temperature (T_g). Storage modulus (E'), loss modulus (E''), and damping factor ($\tan \delta$) were plotted against temperature over this period. Glass transition temperatures were recorded as a maximum value of $\tan \delta$.

2.7. Tensile Analysis. Tensile testing was conducted on an Instron Testing Machine (model number 6025) with a clip-on extensometer to measure specimen extension. The employed cross-head speed was 0.5 mm min⁻¹, and the maximum force recorded was used to obtain the ultimate tensile strength, σ_{ult} . Five specimens were tested, and the standard deviation is reported for all the samples.

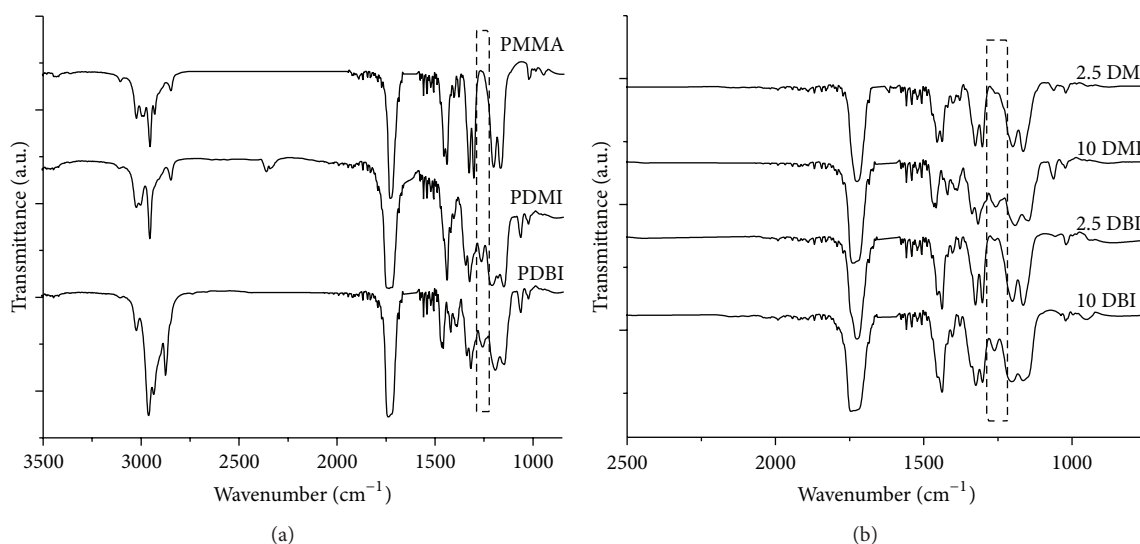


FIGURE 4: FTIR spectra of (a) PMMA, PDMI, and PDBI and (b) PMMA denture base materials modified with 2.5 wt% and 10 wt% of DMI and DBI.

2.8. Hardness Measurements. Hardness was measured on Instron durometer D-XD (Instron, Norwood). The measurements were taken at 10 measuring points at each sample, and the mean values and standard deviations were calculated.

2.9. Charpy Impact Strength. Charpy impact tests were conducted on a Charpy impact tester using the specimens with a V-notch (Figure 3) with the edgewise blow direction. The tests were carried out with striker energy of 8.73 J and a span length of 60 mm at 23°C. The average values of notched Charpy impact energy were obtained from each group of five samples.

3. Results and Discussion

This study evaluates the possibility of using itaconates, renewable, nonpetrochemical materials, as cocomponents for the synthesis of denture base materials. Two esters of itaconic acid (DMI and DBI) were used and four samples (2.5; 5.0; 7.5; and 10 wt%) of denture base materials modified with each of these esters of itaconic acid were prepared. The synthesized samples of PMMA denture base materials modified with DMI and DBI, as well as the homopolymers of PMMA, PDMI, and PDBI, were characterized by FTIR analysis. Figure 4(a) shows FTIR spectra of PMMA, PDMI, and PDBI, while Figure 4(b) shows FTIR spectra of PMMA denture base materials modified with 2.5 wt% and 10 wt% of DMI, that is, DBI.

FTIR spectra of PMMA, PDMI, and PDBI are very similar, as shown in Figure 4(a). Each spectrum shows a sharp intense peak at 1731 cm^{-1} , which is attributed to the presence of C=O stretching vibrations. The broad peak ranging from 1260 cm^{-1} to 1000 cm^{-1} is due to C–O (ester bond) stretching vibrations. The broad band from 950 cm^{-1} to 650 cm^{-1} is due to C–H bending. The peaks at 3025 cm^{-1} , 2950 cm^{-1} , and 2845 cm^{-1} are due to the presence of C–H

stretching vibrations. In spite of being very similar, some subtle differences between FTIR spectrum of PMMA and itaconates are observed because of the presence of the second ester group in itaconates. The peak at 1266 cm^{-1} (C–O–C asymmetric stretching vibration) is presented in PDMI and PDBI FTIR spectra, while for PMMA this band is shifted to lower wave numbers. The increase in itaconate content in denture base material leads to the increase in the intensity of peak at 1266 cm^{-1} (Figure 4(b)).

Figures 5(a) and 5(b) show the DSC thermograms of the initial PMMA denture base material and PMMA denture base material modified with DMI (a) and DBI (b).

The initial PMMA denture base material shows single T_g around 123°C. Single T_g is also observed with the PMMA denture base materials modified with itaconates (Table 2), which indicates that these copolymers are homogeneous in the range of 10–30 nm [44]. The difference between the chemical structures of MMA and DMI is that the one hydrogen of the α -methyl group is replaced by a $-\text{COOCH}_3$ group. It can be seen from the data presented in Table 2 that this substitution caused the lowering of T_g presumably owing to the plasticizing effect of the additional flexible ester group. The insertion of methylene units to lengthen the ester side chains increases their flexibility and leads to internal plasticization of the polymer. This results in a progressive decrease in T_g as the side chains lengthen, so, decrease in T_g was more pronounced in samples modified with DBI than DMI (Table 2).

The values of storage modulus (E'), loss modulus (E'') at 37°C, damping factor ($\tan \delta$), and glass transition temperatures calculated from DSC ($T_{g,DSC}$) and DMTA ($T_{g,DMTA}$) measurements for PMMA denture base materials modified with DMI and DBI, are listed in Table 2. The obtained results for T_g indicated that the T_g values derived from the maximum of the dependence $\tan \delta$ versus temperature. That is, DMTA are higher than those obtained by DSC. Although the values

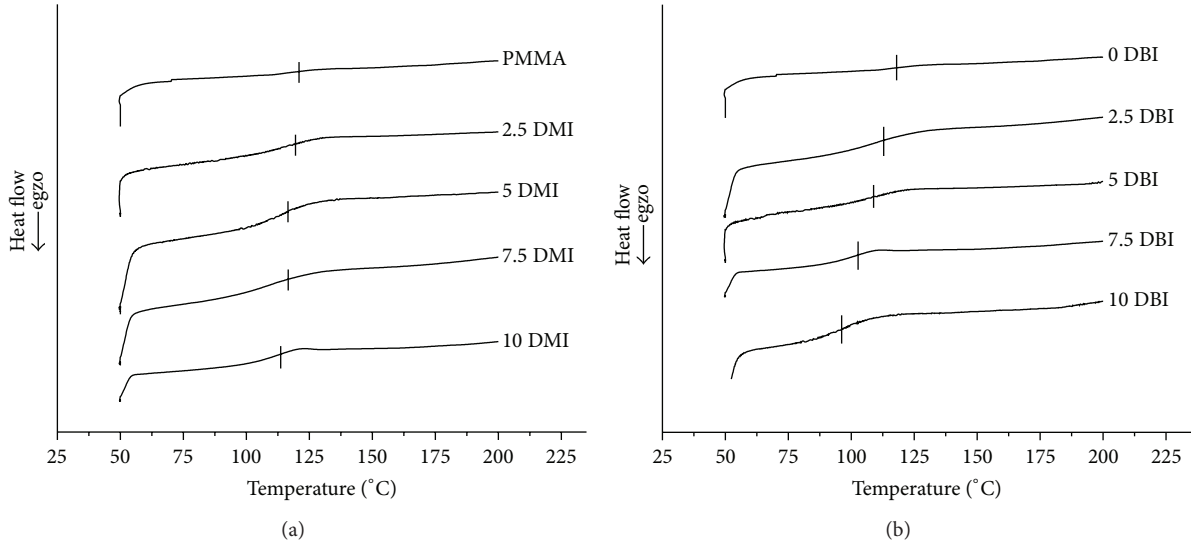


FIGURE 5: DSC curves of PMMA denture base materials modified with (a) DMI and (b) DBI.

TABLE 2: Storage modulus (E'), loss modulus (E'') and damping factor ($\tan \delta_{37}$) at 37°C , maximal value for damping factor ($\tan \delta$), and glass transition temperatures obtained by DSC ($T_{g,\text{DSC}}$) and DMTA ($T_{g,\text{DMTA}}$) measurements for PMMA denture base materials modified with DMI and DBI.

Sample	E' , GPa	E'' , MPa	$\tan \delta_{37}$	$\tan \delta_{\text{max}}$	$T_{g,\text{DSC}}$, $^\circ\text{C}$	$T_{g,\text{DMTA}}$, $^\circ\text{C}$
PMMA	4.03	359	0.089	1.31	123.2	141.5
2.5 DMI	3.99	375	0.094	1.34	122.5	139.1
5.0 DMI	3.83	440	0.115	1.46	121.1	136.2
7.5 DMI	3.65	464	0.127	1.52	119.7	134.5
10 DMI	3.41	481	0.141	1.65	117.5	133.5
2.5 DBI	3.86	417	0.108	1.41	111.0	127.9
5.0 DBI	3.71	460	0.124	1.53	106.4	123.7
7.5 DBI	3.47	475	0.137	1.63	103.8	117.3
10 DBI	3.13	513	0.164	1.81	93.4	107.4

of $T_{g,\text{DMTA}}$ are higher than those of $T_{g,\text{DSC}}$, the trend of T_g variation is similar for both methods.

Figures 6(a) and 6(b) show the effect of copolymer composition on storage modulus (E') dependence over temperature.

An “ideal” hard denture reline material would demonstrate a high stiffness (E') and a moderate deformation with stress (E'') but a low $\tan \delta$, as it is preferable that the material returns to its original shape shortly after the load is removed. As presented in Figures 6(a) and 6(b), between 25°C and 100°C , the modified denture base materials behaved as a homogeneous rigid solids, while the increase in temperature was firstly followed by a dramatic fall in modulus over a short temperature span when the material became rubbery beyond its T_g and afterwards E' entry into the plateau. It is also clear that the storage modulus decreased with increasing itaconate concentration while at the same time loss modulus and $\tan \delta$ increased as could be seen from their values presented in Table 2. The presented values are obtained at 37°C because that is the material’s operational temperature. These results indicated that the addition of

itaconates in PMMA denture base material reduced material’s stiffness and increased its deformation with stress due to the plasticizing effect of the aliphatic side chains of itaconates. The decrease in E' and increase in E'' and $\tan \delta$ were more pronounced in case of DBI compared to DMI which further confirmed the previous statements. However, we can note that, in case of addition of a small amount of DMI (2.5 wt%) the decrease in E' was negligible (less than 1%) which means that with this amount of itaconate, the denture base material had practically the same dynamic-mechanical properties as the commercial material. The T_g values obtained by DMTA measurements (i.e., temperature corresponding to $\tan \delta$ maximum) are higher compared to the T_g values measured by DSC (Table 2) which is in accordance with the literature data [45]. The T_g obtained by DSC method represents the temperature at which the material undergoes the maximum change in polymer chain mobility that corresponds to the chemical definition of the T_g . The T_g obtained as peak of the $\tan \delta$ dependence over temperature describes the damping characteristics of material and has historical significance, since it was the first DMTA property quantified and much

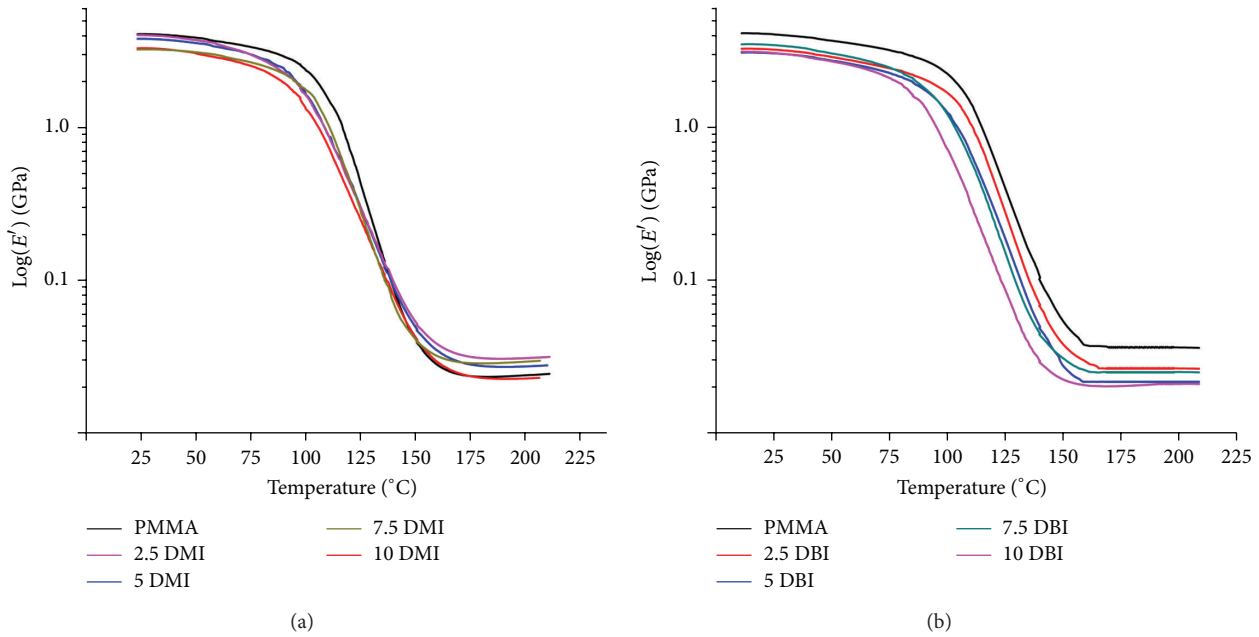


FIGURE 6: Changes in storage modulus (E') over temperature for PMMA denture base materials modified with (a) DMI and (b) DBI.

TABLE 3: Ultimate tensile strength (σ_{ult}), elongation at break (ϵ), Shore D hardness, and Charpy impact strength of PMMA denture base materials modified with DMI and DBI.

Sample	σ_{ult} , MPa	ϵ	Shore D	Charpy, J/cm ²
PMMA	63.8	6.18	96.0	0.315
2.5 DMI	61.8	5.21	96.3	0.317
5.0 DMI	58.1	4.91	97.0	0.307
7.5 DMI	55.8	4.43	96.1	0.301
10 DMI	52.7	5.08	94.9	0.294
2.5 DBI	58.5	5.23	91.5	0.309
5.0 DBI	56.3	4.7	95.5	0.295
7.5 DBI	54.9	4.71	94.8	0.281
10 DBI	54.2	6.26	95.1	0.268

of the DMTA T_g reference data is based on the $\tan \delta$ peak temperature.

The obtained results for ultimate tensile strength (σ_{ult}), elongation at break (ϵ), Shore D hardness, and Charpy impact strength are presented in Table 3.

The tensile behavior of PMMA denture base materials modified with DMI or DBI is presented in Table 3. The materials modified with itaconates, especially those containing DBI, exhibited a lower ultimate tensile strength compared to the commercial PMMA denture base material, suggesting that the incorporation of aliphatic side chain into methacrylates structure decreased the strength of the material. A plausible explanation for this behavior rests on the fact that mechanical properties depend on the morphology of the material. The introduction of bulky pendant group hinders the ideal lining up of the polymer chains producing microdefects in the polymeric structure that result in

decrease in ultimate tensile strength. The reduction of ultimate tensile strength increased with the increase in itaconate content in the system as well as with the increase in the side chain length which furthermore confirmed the findings stated above. However, bearing in mind the advantages of using of itaconates over methacrylates and the fact that the prescribed tensile strength requirements [46] were attained, it could be concluded that, regarding investigated parameter, denture base materials modified with itaconates are suitable replacements for commercial, pure PMMA material.

Table 3 also shows elongation at break and the mean Shore D hardness of the PMMA denture base materials modified with DMI and DBI.

The increase in itaconate content led to the decrease in elongation at break except for the samples modified with 10 wt% of itaconates in which case elongation increased.

Even though there were some differences between the hardness values of the different samples, these differences were too small to be relevant. Taking into consideration all the factors that affect the hardness of material (surface porosity, residual monomer concentration, etc.) we may presume that the modification of PMMA denture base material had no effect on Shore D hardness. All investigated materials fulfilled the required hardness values for denture base materials [46].

In all impact tests experiments, the specimens broke with a sharp fracture, exhibiting a typical brittle fracture behavior characterized by a lack of distortion of the broken parts. There was a significant difference between the impact resistance of the control and samples modified with 7.5 wt% and 10 wt% of DBI. The lower impact resistance of these modified materials may be attributed to the formation of microdefects in the polymer matrix which were formed as a result of the introduction of linear side chain. These microdefects act as a stress concentrator and crack accelerator. Despite the

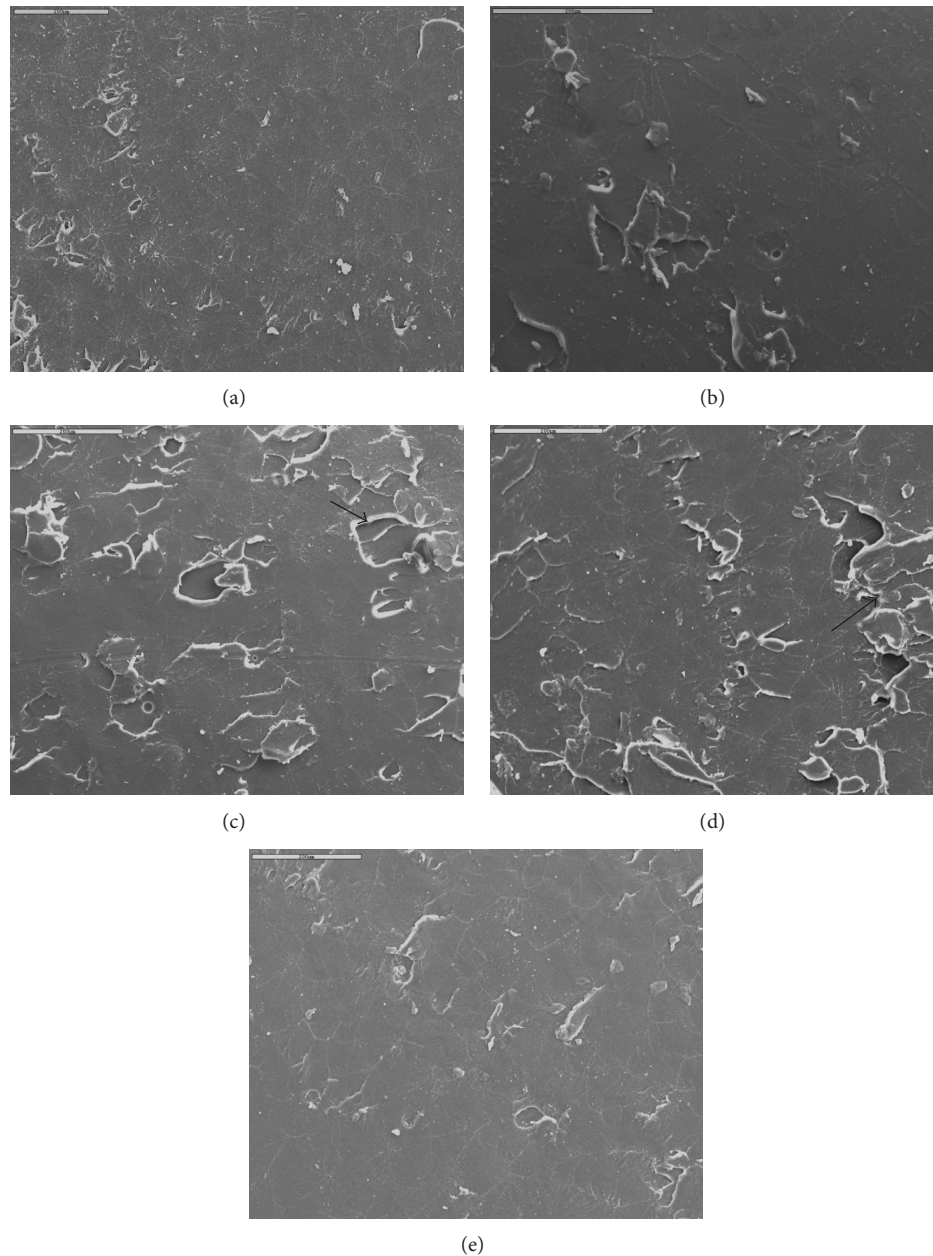


FIGURE 7: SEM images of brittle fractures of PMMA denture base material modified with (a) 2.5 wt% of DMI, (b) 2.5 wt% of DBI, (c) 10 wt% of DMI, (d) 10 wt% of DBI, and (e) commercial PMMA denture base material (original magnification 200 times).

progressive reduction in impact resistance with the increase in itaconates amount (Table 3), the measured impact energy values satisfied the requirements of the American Dental Standards Institute [46].

The SEM images of fractures of PMMA denture base material modified with DMI and DBI are presented in Figure 7. The fractures that exhibited a smooth surface were classified as brittle. Those presenting plastic deformations, exhibiting rough and jagged surfaces, were recorded as intermediate (ductile-to-brittle transition) fractures.

The SEM microscopy showed that most fractures were brittle for the denture base materials modified with low amount of itaconates. The brittle fractures exhibited

well-defined, flat, compact, and organized surface fractures. However, fractures of samples modified with 10 weight percent of itaconates presented higher amount of irregular surface patterns (Figures 7(c) and 7(d)) when compared to the other samples (Figures 7(a), 7(b), and 7(e)), with a high number of grooves in the brittle fractures and presence of crazing in the intermediate fractures. Arrows in Figures 7(c) and 7(d) show the presence of crazing, revealing high plastic deformation. Although irregularities could be seen in each fracture of examined samples, a common finding was that a granular microstructure was clearly distinguishable, demonstrating that acrylic resin fails by transgranular or transcrystalline fracture [47].

4. Conclusions

This study evaluated the possibility of using itaconates, renewable, nonpetrochemical materials, as cocomponents in the synthesis of denture base materials. Two esters of itaconic acid (DMI and DBI) were used in four concentrations (2.5; 5.0; 7.5; and 10 wt%) to modify the commercial denture base materials. The blending of itaconates with other components was homogenous and confirmed by FTIR analysis. Also the DSC analysis showed progressive decrease in T_g values with the increase in itaconate component amount, compared to T_g of the pure PMMA formulation, and, by showing one T_g value for all samples, confirmed their homogeneity. The plasticizing effect of itaconates was observed and confirmed with the decrease in E' and E'' and the ultimate tensile strength values. This effect was more pronounced when the itaconate with longer aliphatic chain, DBI, was used. With the increasing amount of itaconates, except for the 10 DBI sample, the elongation at break was increased. All employed measurements confirmed that required values of mechanical properties for denture base materials were fulfilled. This investigation showed that modification of PMMA denture base materials with alkyl itaconates was a feasible way to obtain materials that were more environmentally and patient friendly than pure PMMA denture base materials.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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