PHYSICAL, MECHANICAL AND ENVIRONMENTAL PROPERTIES
OF DENTURE BASE POLY(METHYL METHACRYLATE)
FILLED WITH HYDROXYAPATITE

BY

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Thesis Submitted in Fulfillment of the
Requirements for the Degree of
Master of Science

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In the name of ALLAH the beneficent and the merciful, all praise and thanks is due to ALLAH for giving me the health and ability to complete my research successfully.

“If I have seen further than others, it is by standing upon the shoulders of Giants”
(Isaac Newton, 1642–1727)

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Lastly and most importantly, I would like to dedicate my sincere appreciation to my parents, who raised me, supported me, taught me, and loved me. They deserve very
special thanks for encouraging me to pursue this project even though they were many miles away. I dedicate this thesis to my father, who taught me that the best kind of knowledge to have is that which is learned for its own sake. I also dedicate this thesis to my mother, who taught me that even the largest task can be accomplished if it is done one step at a time.
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<tr>
<td>ASTM</td>
<td>American Society for Testing and Materials</td>
</tr>
<tr>
<td>BaTiO&lt;sub&gt;3&lt;/sub&gt;</td>
<td>Barium Titanate</td>
</tr>
<tr>
<td>BIS-GMA</td>
<td>Bis-(2-Hydroxypropyl)-Methacrylate</td>
</tr>
<tr>
<td>BPO</td>
<td>Benzoyl Peroxide</td>
</tr>
<tr>
<td>EDX</td>
<td>Energy Dispersive X-Ray Spectroscopy</td>
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<tr>
<td>EGDMA</td>
<td>Ethylene Glycole Dimethacrylate</td>
</tr>
<tr>
<td>FTIR</td>
<td>Fourier Transform Infra Red</td>
</tr>
<tr>
<td>h</td>
<td>Hour</td>
</tr>
<tr>
<td>HA</td>
<td>Hydroxyapatite</td>
</tr>
<tr>
<td>HDPE</td>
<td>High Density Polyethylene</td>
</tr>
<tr>
<td>ISO</td>
<td>International Organization for Standardization</td>
</tr>
<tr>
<td>Min</td>
<td>Minute</td>
</tr>
<tr>
<td>MMA</td>
<td>Methyl Methacrylate</td>
</tr>
<tr>
<td>MW</td>
<td>Molecular Weight</td>
</tr>
<tr>
<td>OD</td>
<td>Optical Density</td>
</tr>
<tr>
<td>ODP</td>
<td>Opaque Dental Porcelain</td>
</tr>
<tr>
<td>PMMA</td>
<td>Poly(methyl methacrylate)</td>
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<tr>
<td>SBF</td>
<td>Simulated Body Fluid</td>
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<td>SEM</td>
<td>Scanning Electron Microscope</td>
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<td>SEN-B</td>
<td>Single Edge Notch Bending</td>
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<tr>
<td>SG</td>
<td>Specific Gravity</td>
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<td>Abbreviation</td>
<td>Full Form</td>
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<tr>
<td>TA</td>
<td>Thermal Analysis</td>
</tr>
<tr>
<td>TGA</td>
<td>Thermogravimetric Analysis</td>
</tr>
<tr>
<td>VHN</td>
<td>Vickers Hardness Number</td>
</tr>
<tr>
<td>XRD</td>
<td>X-Ray Diffraction</td>
</tr>
<tr>
<td>wt</td>
<td>Weight</td>
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<table>
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<tr>
<td>$\gamma$-MPS</td>
<td>3-(methacryloxy) propyl trimethoxysilane</td>
</tr>
<tr>
<td>$\sigma_c$</td>
<td>The composite tensile strength</td>
</tr>
<tr>
<td>$\sigma_m$</td>
<td>The matrix tensile strength</td>
</tr>
<tr>
<td>$\rho$</td>
<td>Density</td>
</tr>
<tr>
<td>$\rho_c$</td>
<td>Density of composite</td>
</tr>
<tr>
<td>$\rho_f$</td>
<td>Density of filler</td>
</tr>
<tr>
<td>$\rho_w$</td>
<td>Density of water</td>
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<tr>
<td>$a$</td>
<td>Notch length</td>
</tr>
<tr>
<td>$b$</td>
<td>Specimen width</td>
</tr>
<tr>
<td>$d$</td>
<td>Specimen thickness</td>
</tr>
<tr>
<td>$E_c$</td>
<td>The composite tensile modulus</td>
</tr>
<tr>
<td>$E_m$</td>
<td>The matrix tensile modulus</td>
</tr>
<tr>
<td>GPa</td>
<td>Gigapascal</td>
</tr>
<tr>
<td>$K_{IC}$</td>
<td>The stress intensity factor</td>
</tr>
<tr>
<td>$S$</td>
<td>Support span</td>
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<tr>
<td>MPa</td>
<td>Megapascal</td>
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<tr>
<td>$P$</td>
<td>Load</td>
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<tr>
<td>$W_1$</td>
<td>Sample weight in air</td>
</tr>
<tr>
<td>$W_2$</td>
<td>Sample weight in water</td>
</tr>
<tr>
<td>$Y$</td>
<td>Geometrical correction factor</td>
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SIFAT FIZIKAL, MEKANIKAL DAN PERSEKITARAN PELANDAS DENTURE POLI(METIL METAKRILAT) TERISI HIDROKSIAPITIT

ABSTRAK

Dalam bidang pergigian, resin poli(metil metakrilat) PMMA mampu bertahan sebagai bahan paling sesuai dan popular untuk fabrikasi asas dentur bagi prostodontik boleh tanggal. Walau bagaimanapun, retak pada asas dentur berlaku secara intra-oral disebabkan intensifikasi tegasan semasa fungsi atau apabila dentur terjatuh secara tidak sengaja pada permukaan yang keras. Dalam kajian ini, pengisian serbuk seramik hidroksiapatit (HA) ke dalam PMMA dilakukan untuk menentukan kesan kandungan pengisian HA terhadap sifat fizikal, mekanikal serta alam sekitar pada asas dentur akrilik. Hal ini sama seperti mengkaji kesan beberapa faktor lain yang dianggap faktor penting dalam menentukan sifat rencam secara keseluruhan, iaitu kesan daripada campuran fizikal sebagai fungsi masa, nisbah agen gandingan dan kepekatan pengisi HA. Serbuk HA diolah dengan dalam nisbah yang berbeza, 3-(trimetoksisili) propil metakrilat (agen gandingan silana) (iaitu 0, 5, 7, dan 10% berdasarkan berat) untuk mengkaji kesan nisbah agen gandingan. Tiga nisbah yang berbeza (iaitu. 5, 10, dan 15% berdasarkan berat) daripada pengisi yang diolah ditambah ke dalam matriks (PMMA dan 0.5% BPO) untuk dibandingkan dengan PMMA tulen dan bahan lazim. Komponen serbuk dicampur berdasarkan teknik pengisar bebola planet (PBM) bagi 10, 20, 30 dan 40 min masing-masing untuk mengkaji kesan masa kisaran. Campuran serbuk pada cecair dilakukan berdasarkan penggunaan makmal gigi yang standard. Dalam kajian campuran, ditemui bahawa saiz partikel rencam PMMA/HA terkesan melalui
penggunaan teknik PBM sebagai suatu fungsi masa. Berasaskan ujian mekanikal, masa kisaran 30 minit dianggap masa optimum untuk mencapai imbangan yang baik bagi sifat mekanikal rencam. Secara amnya, penggunaan agen gandingan silana γ-MPS mampu meningkatkan sifat mekanikal rencam HA/PMMA. Sebagai tambahan, 10% daripada γ-MPS dianggap jumlah optimum untuk mencapai imbangan yang baik bagi sifat mekanikal. Suatu penambahbaikan sifat mekanikal yang munasabah dicapai pada muatan pengisi yang amat rendah, iaitu 5% berat selepas suatu penurunan diperhatikan sebagai suatu fungsi peningkatan muatan pengisi. Pendedahan pada alam sekitar boleh melemahkan sifat rencam PMMA/HA kerana pengambilan air dan SBF secara maksimum akan bertambah dengan peningkatan muatan pengisi. Nilai $K_{IC}$ bagi sampel pengisi dengan 5, 10 dan 15% berat HA berkurangan sebanyak 1.26%, 4.44% dan 6.06%, masing-masing, selepas direndam dalam SBF selama 2 bulan. Penambahbaikan yang signifikan dalam radiopasiti PMMA dicapai melalui penggabungjalinan hidroksiapatit. Sebagai tambahan, ditemui bahawa radiopasiti rencam ditambah baik sebagai suatu fungsi dalam peningkatan muatan HA.
PHYSICAL, MECHANICAL AND ENVIRONMENTAL PROPERTIES OF DENTURE BASE POLY(METHYL METHACRYLATE) FILLED WITH HYDROXYAPATITE

ABSTRACT

In the field of dental materials, poly(methyl methacrylate) (PMMA) resin maintains itself to be the most popular and suitable material for the fabrication of denture bases in removable prosthodontics. However, the fracture of acrylic denture base occurs intra-orally because of the stress intensification which is subjected during the function or when the dentures are accidentally dropped on a hard surface. In this study, the incorporation of hydroxyapatite ceramic (HA) powder into PMMA was carried out in order to find out the effect of HA incorporation on the physical, mechanical and environmental properties of the acrylic denture base. This is as well as investigating the effect of some other factors which are considered key factors in determining the overall properties of the composite, i.e. effect of physical mixing as a function of time, coupling agent ratio and HA filler concentration. The HA powder was treated with different ratios of the silane coupling agent 3-(trimethoxysilyl) propyl methacrylate (i.e. 0, 5, 7, and 10% by weight) with respect to the HA powder in order to evaluate the effect of coupling agent ratio. Three different ratios (i.e. 5, 10, and 15% by weight) of the treated filler were added into the matrix (PMMA and 0.5% BPO) to be compared with pure PMMA and the conventional material. The powder components were mixed using the planetary ball milling technique (PBM) for 10, 20, 30 and 40 min respectively to study the effect of mixing time. The powder to liquid mixing was done according to standard dental laboratory usage. In the mixing study, it was found that the particle size of
PMMA/HA composite was affected by using planetary ball milling technique (PBM) as a function of time. Based on the mechanical testing, the mixing time of 30 minutes was considered to be the optimum time to achieve a good balance of mechanical properties of the composites. Generally, the use of $\gamma$-MPS silane coupling agent led to improved mechanical properties of HA/PMMA composites. In addition, 10% of $\gamma$-MPS was considered to be the optimum amount to achieve a good balance of mechanical properties. A considerable improvement of the mechanical properties was achieved at very low filler loading, i.e. 5 wt% after which a decrease was observed as a function of increasing the filler loading. The environmental exposure has a weakening effect on the properties of the PMMA/HA composites whereby, both maximum water and SBF uptakes increased with increasing the filler loading. In addition, the $K_{IC}$ values for the filled samples with 5, 10 and 15 wt% of HA decreased by 1.26%, 4.44% and 6.06%, respectively, after being immersed in SBF for 2 month. A significant improvement in the radiopacity of the PMMA was achieved by the incorporation of hydroxyapatite. Additionally, it was found that the radiopacity of the composite was improved as a function of the increase of HA loading.
CHAPTER 1
INTRODUCTION

1.1 Polymeric Denture Base Materials

Polymers can be produced either in rigid or flexible state by changing the composition of monomers. Most of clinicians consider that denture base should be rigid. Poly(methyl methacrylate) is a rigid resin that provides sufficient structural integrity to be used as a denture base. The reaction of the methyl methacrylate monomer with other monomers can produce a much softer polymer which can be used to line the underside of a denture for improved fit and comfort for some patients. The rigid acrylic can also be used to produce a custom-made tray for carrying the impression material into the patient’s mouth to maximize accuracy (Ahmed et al., 1990).

There has been a continuous research to fabricate denture base material of natural appearance that would withstand the deterioration during the service life. Before the advent of acrylic, vulcanite rubber had been the most satisfactory material available for almost 75 years. Apart from that, celluloid was also used as it had a more pleasing initial appearance than rubber. But it soon warped and discolored and was otherwise unsatisfactory. Currently, the denture bases available are largely acrylic resins. Since its introduction for the use in dentistry there has been a continual search for modified practices in processing the resins which will lead to improved qualities in finished structure (Kramer, 2004).
PMMA is an acrylic colorless and transparent material used widely as a denture base material (Anusavice, 2003). The adequacy of PMMA’s physical properties proved the material’s feasibility for dental applications and hence gained its popularity in dentistry. Its inherited characteristics are the ones needed for use in oral cavity, and their performance features are related to their biological, physical, aesthetic, and handling characteristics (Schricker et al., 2006). Nowadays, PMMA is the material of choice for denture base fabrication. Most research activity in the field of denture base materials, has concentrated on attempts to improve fracture toughness and impact strength of acrylic materials. Moreover, the favorable working characteristics, processing ease, accurate fit, and superior esthetics of the PMMA make it the most suitable material for the fabrication of denture base (Mohamed et al., 2008). Even though PMMA has these good combinations of properties, there is a need for improvements in the fracture resistance of PMMA. In denture, most of these fractures occur inside the oral cavity during service, primarily because of resin fatigue (Meng and Latta, 2005).

1.2 Characteristics of the Ideal Denture Base Material

A denture base material must make the production of denture effortless and bring them within the reach of the average person. It must also combine easiness and speed of preparation with high resistance of contamination and worsening in oral cavity environment. An ideal denture base material should be capable of matching the physical and the mechanical properties of the natural oral soft tissues (McCabe and Walls, 2008). The appearance is one of the most important aspects of the physical properties that should be taken in consideration when a material is chosen to fabricate the denture base.
However, this depends on whatever the base will be visible when the patient opens his/her mouth. Although, the normal temperature in the mouth varies only from 32°C to 37°C, accounts must be taken of the fact that patients take hot drinks at temperature up to 70°C and cleaning dentures in very hot water or even boiling water. Thus, a polymer which is used to fabricate a denture base should have a high enough value of glass transition temperature ($T_g$) to prevent softening and distortion during the function (Bhola et al., 2010).

The denture base should have a good degree of dimensional stability in order for its shape not to change over a period of time, to avoid the distortions which may occur due to the thermal softening and other mechanisms such as relief of internal stresses also continued polymerization and water absorption may contribute to dimensional instability (Ferracane, 2001). Normally, denture base materials are supposed to have a low value of specific gravity in order for it to be as light as possible. This reduces the gravitational displacing forces which may occur especially on the upper denture. A high value of thermal conductivity would enable the denture user to maintain a healthy oral mucosa and retain a normal reaction to hot and cold stimuli. If the base is a thermal insulator, it is possible that the patient may take a drink which the patient would normally detect as being too hot to bear’, and undergo a painful experience as the drink reaches the throat and gut (Hussain, 2004).

Ideally, the denture base should be radiopaque and capable of detection using normal diagnostic radiographic techniques. Occasionally, patients may swallow their dentures and even inhale fragments of dentures if involved in a violent accident, such as a car crash. Early radiological detection of the denture or fragment of denture is
considered immense help in deciding the best course of treatment (Davy, 1997 & McCabe and Walls, 2008). Although opinion varies slightly, most clinicians are of the view that denture base should be rigid. It is therefore advantageous to have a high value of modulus of elasticity. Thus, to ensure that stresses encountered during biting and mastication does not cause permanent deformation, a high value of elastic limit is required. A combination of a high modulus and high value of elastic limit would have added advantage that it would allow the base to be fabricated in relatively thin section. Fracture of upper denture bases invariably occur through the midline of the denture due to the flexing (Elshereksi et al., 2009). The denture base should have sufficient flexural strength to resist fracture. Fracture of denture base in situ often occurs by a fatigue mechanism in which relatively small flexural stresses, over a period of time, eventually lead to the formation of small crack which propagates through the denture resulting in fracture. Therefore, the base material should have an adequate fatigue life and high value of fatigue limit. When patients remove dentures for cleaning or overnight soaking, there is a danger of fracture if the denture is accidentally dropped onto a hard surface. The ability of the denture to resist such fracture is a function of the impact strength (Ferracane, 2001).

A denture base material should be chemically inert. It should, naturally, be insoluble in oral fluid and should not absorb water or saliva since this may affect negatively on the mechanical properties of the material and cause it to become unhygienic. In the unmixed or uncured states, the denture base material should not be harmful to the person involved in its handling. An ideal denture base material should be relatively inexpensive and have a long shelf life so that material can be purchased in
bulk and stored without deteriorating. The material should be easy to manipulate and fabricate without having to resort to using expensive processing equipment. If fractures do occur they should be easy to repair (McCabe and Walls, 2008).

Currently, it has not yet been achievable to develop a denture base material which fulfils all these requirements. Though, a number of materials were used to conduct testing but they have been discarded because they were considered unsatisfactory from many aspects. It is time that dentists stopped accepting the manufactures’ word that the best possible denture base materials are now available. If the tissues covered by the denture were hard then no dentures would fit. Until we are able to control absolutely the changes in our impression materials and denture bases, we can never produce a really perfect set of dentures (Darvell and Clark, 2000).

1.3 Problem Statement

There is no ideal denture base, although poly(methyl methacrylate) resin is regarded by most practitioners as the best approach to date. Previously, where it fell short of the idealness, it still falls short today, although there have been numerous of attempts and suggestions made to improve it. Its major disadvantages are that of relative softness when compared with tooth enamel, dimensional changes on processing, and susceptibility to attack by organic liquids still applies (Akan et al., 2010). One of the criticisms of methyl methacrylate type of dental resin is a tendency to fracture. This tendency is thought to be coupled with the flexural fatigue resistance of the material, such that the greater the value of flexural fatigue resistance in the material, the greater will be its resistance to mid-line fracture. Even though PMMA has good combinations of
properties, there is a need for improvements in the fracture resistance. Most of fractures of the denture occur inside the oral cavity during service, primarily because of resin fatigue (El-Sheikh and Al-Zahrani, 2006).

The denture base resin is subjected to various stresses during the function, these include tensile and shear stresses. Some of the factors responsible for denture fracture include stress intensification, increased ridge leading to an unsupported denture base, sharp changes at the contours of the denture base (Elshireksi et al., 2009). However, further significant improvements are needed to produce a denture base material with good combinations of mechanical, physical and biological properties in order to reduce the chance of denture base fracture which occurs inside the oral cavity. However, it is clear that the ideal situation of a completely acceptable dental polymer is still far from being reached (Köroğlu et al., 2009). Dentures made from acrylic resin are radiolucent because Carbon, Oxygen and Hydrogen atoms are poor X-ray absorbers. This is a serious disadvantage of these materials. If a patient swallows or inhales a denture or fragment of a denture it is difficult to detect using simple radiological techniques (McCabe and walls, 2008).

1.4 Research Objectives

The aim of this study was to produce a denture base material with a good combination of mechanical, physical and environmental resistance properties. Apart from that, the effect of filler incorporation into poly(methyl methacrylate) PMMA as a denture base material in term of process ability, physical, morphology and mechanical
properties were evaluated. As such, the research project was divided into the following segments:

- To investigate the effect of mixing time on the physical and the mechanical properties of PMMA matrix filled by HA ceramic powder.
- To study the influence of coupling agent ratio on the mechanical properties of PMMA matrix filled by HA ceramic powder.
- To evaluate the effect of HA filler concentration on the physical and the mechanical properties of PMMA denture base material.
- To investigate the influence of the incorporation of HA ceramic powder into PMMA matrix on the absorption of water and simulated body fluid.
- To evaluate the effect of environmental exposure on some mechanical properties of the modified PMMA denture base material.

1.5 Outline of the Thesis

Chapter one provides a brief introduction to polymeric denture base materials along with the historical developments of the polymers which are used in the fabrication of denture bases. Specifically, they are poly(methyl methacrylate), and the ideal properties that are needed in denture base materials. Some of the significant improvements that are needed to produce a denture base material with good combinations of mechanical, physical and biological properties are also included. The objectives behind the present study are listed.
Chapter two gives an overview on the definition of the denture base, a historical review of denture base materials, and some of the materials that are used in the profession of prosthetic dentistry. The factors that affect the mechanical, physical, and environmental resistance properties of the denture base materials with particular reference to acrylic resin. A review of the recent literature on denture base materials is also included in order to link the findings of a large number of experiments were conducted under different environment on large conditions with a view to the correlating results of a large number of experiments that have been conducted.

Chapter three contains the raw materials that were used in the present study and its step by step description of the employed experimental procedures, the details of the laboratory equipment used along with the processing techniques of data that were employed during the present study.

Chapter four includes the outcome of the present study along with the results and discussion. Specifically it reports, the effect of γ-MPS coupling agent ratio, mixing time, and filler concentration on the mechanical properties of poly(methyl methacrylate) filled by HA as denture base material. The influence of filler incorporation on the absorption and solubility of PMMA in water and simulated body fluid are reported along with the influence of environmental exposure on mechanical properties of poly(methyl methacrylate) denture base material. Finally, the result of filler loading on the Radiopacity of PMMA is presented.

Chapter five summarizes the concluding remarks on the outcome of the present study along with some suggestions for future studies.
CHAPTER 2
LITERATURE REVIEW

2.1 Introduction

The loss of teeth by accident or disease plagued mankind throughout the ages. In order to restore a degree of function and appearance, it has been necessary always to adapt contemporary materials to dental applications as they are available in one period of history. As civilization progressed, there has been continued refinement of the materials available for dental practice. As the time passed, the civilization advanced with the development of biological, chemical and physical science. As a result, there occurred a slow but steady increase in both the quantity and quality of useful materials available for dental prostheses. For this reason, the material should be biologically compatible, readily available, reasonably inexpensive and simple to manipulate with a readily controlled technical procedure, to develop prosthesis that is functionally effective and pleasing in appearance (Phoenix, 1996). The means of replacing missing tooth structure by artificial materials continues to account for a large part of the application of material sciences. Denture base materials have always been a matter of research in the field of dental materials (Stanley, 1992). We need a sound foundation for a strong building, similarly for fabricating long lasting, esthetically and biologically acceptable dentures; we need a favorable denture base.
2.2 Definition of Denture Base

According to The Glossary of Prosthodontic Terms (Gpt-8) (2005) a denture base could be defined as the part of the denture that rests on the foundation and to which teeth are attached.

2.3 Historical Review of Denture Base Materials

The review of different denture base materials provides a clear picture about the various developments that have taken place in this field. The polymers, especially acrylic resins, after entering this field more than 70 years ago seem to be undergoing constant change and are the materials of choice. By the 8th century the Japanese were masters of the art of woodcarving and it was possible that the earliest wooden denture was made at that time. Dentures were carved from a single piece of wood, usually sweet smelling species such as box and cherry (Engelmeier, 2003). Natural teeth were fixed with the help of screws. George Washington also had a set of dentures made from wood. The drawbacks were that denture bases warped and cracked in the presence of moisture and posed esthetic and hygienic challenges (Johnson, 1959). Further progress was slow until the 17th century. It has been said that modern dentistry begun with Pierre Fauchard (1678-1761) who developed many prosthetic techniques. He used human teeth or teeth made from hippopotamus or elephant ivory in the denture as shown in Figures (2.1-2.3). He carved dentures from a single piece of ivory or bone. Although bone displayed better dimensional stability than wood, esthetic and hygienic concerns remained. Whereas ivory was stable in the oral environment, and offered significant esthetics but
its drawbacks were that it was not readily available, and was relatively expensive (Engelmeier, 2003).

Fig. 2.1: Ivory upper denture, c.1760, this upper denture set made of hippopotamus ivory (http://www.namibiadent.com/History/HistoricPictures.html).

Fig. 2.2: Two full upper dentures. The plates of these dentures made of hippopotamus ivory; the anterior teeth are human teeth (http://www.namibiadent.com/History/HistoricPictures.html).

Fig. 2.3: Spring retained ivory denture dating to the early 18th century. (http://www.namibiadent.com/History/HistoricPictures.html).
During the later part of the 19\textsuperscript{th} century, polymers entered the field of denture base materials. Charles Goodyear (1839) developed the art of producing rubber and in 1851 his brother Nelson Goodyear invented a process for making hard rubber called vulcanite. In 1854 Thomas Evan introduced vulcanite as a denture base material (Figure 2.4). He made vulcanite-based dentures for Charles Goodyear senior and a year later for Charles Goodyear junior. It was produced by heating natural rubber in the presence of sulfur to produce a hard, reddish-brown rubber with many desirable properties (Ring, 1985 & Rueggeberg, 2002). The introduction of the vulcanite into dentistry is like the discovery of the fire in the history of mankind. Vulcanite was almost the answer to the dentist’s problems in the fabrication of the dentures. Despite its displeasing appearance, vulcanite dentures fitted the ridges of the patient more exactly, so that dentures could be worn with comfort. Other advantages were economy, durability, light weight, and ease of fabrication (Anusavice, 2003). After its introduction, the vulcanite remained the principal denture base material for the next 75 years. Until the introduction of PMMA in 1930, vulcanite and porcelain teeth were standard materials for fabrication of prostheses (McCabe and Walls, 1998).

Fig. 2.4: Vulcanite dentures, also called ‘Rubber Plates’ c. 1851 (http://www.namibiadent.com/History/HistoricPictures.html).
The first known casting of a complete aluminum base (Figure 2.5) was made in 1867 by Dr. Bean who invented the casting machine. In 1888 Carroll presented a method for casting aluminum bases under pressure. Although accuracy of fit and other advantages made aluminum the material of choice, the other difficulty of relining, increased cost of fabrication and a potential relationship between aluminum and Alzheimer’s disease had discouraged the use of aluminum and its alloys (Halperin, 1980).

In 1937, methyl methacrylate was clinically evaluated by Wright and found to fulfill virtually all the requirements of what seemed to be an ideal denture materials by that time (Peyton, 1975). By 1946, the acrylic resin represented such significant improvement in its application. It was estimated that 95% of all dentures were fabricated using methyl methacrylate polymers. Initially, acrylic resins were polymerized by heat. In Germany (1947), acrylic resins were developed using chemical accelerators for polymerization and termed as self or auto polymerization resins (Khindria et al., 2009).

Fig. 2.5: Partial dentures made from aluminum, latter half of the 19th century (http://www.namibiadent.com/History/HistoricPictures.html).
The PMMA and its copolymers continued to be the most popular non-metallic materials. Its advantages were economy, simple processing technique, stable colors, optical properties, adequate strength and other physical properties which make them ideal materials of choice, free from toxicity and easily pigmented (Craig, 2003).

### 2.4 Dental Materials and Their Applications

There are four major classes of materials used in dentistry: metal, ceramics, polymers and composites. Composites are a mixture of two or more of the first three classes in which the different components remain distinct from one another in the final structure. A well-known example is fiberglass, a polymer reinforced with fine glass fibers that remain physically separate and uniformly distributed throughout the polymer matrix (Ferracane, 1995).

#### 2.4.1 Metals

Metals consist of aggregate of atoms regularly arranged in a crystalline structure. It is the oldest of the three main categories of materials used as dental materials. Metal is defined by a particular set of characteristics, including high thermal and electrical conductivity; flexibility (they can be designed without breaking); blackout or opacity (they have a surface that reflects light strongly and looks bright and shiny). Among its features is that elements classified as they tend to like these dissolve in water or other aqueous solutions, and produce atoms with a positive charge. Most of the tools and equipments used in dentistry are usually made of metal steel. It is popular because it is plentiful and economic (Noort, 2002). In addition, they are strong, corrosion resistant,
biocompatible, cleanable, easily sterilized, and can be sharpened to produce cutting edges. Nearly 80% of the elements in the periodic table are classified as metals. All metals are white color with the exception of gold which is yellow and copper which is reddish. Metals harden with their atoms in a very regular or crystalline arrangement, often in the form of a cube. The great strength of the metallic bond that holds the atoms together leads to the high melting temperatures of metals. With the exception of mercury, metals are generally hard and lustrous at ambient temperatures (O'Brien, 2002).

However, mercury metal which is in liquid form at room temperature is used in dentistry to repair decayed teeth. These powdered silver-tin alloys are mixed with drops of mercury for the production of hard material called amalgam restorative. The reaction between mercury and alloy which follows mixing is termed amalgamation reaction. For many years, dental amalgam was the most commonly used of all filling materials with a large measure of success. They can be formed or cast into many different shapes because of the forming ease, high strength and stability (McCabe and Walls, 2008).

Metals are widely used in dentistry as structural components to either fix or restore the tooth structure. Crowns which replace the outer coronal structure of a tooth can be made of metal; the metallic color is not undesirable. Therefore, the metallic restoration is preferred exclusively in the back section of the oral cavity. They can also be used more conventionally to replace portions of a tooth. When a portion of the tooth within the cusp is replaced, this type of restoration is called an inlay. If one cusp or more are included in the restoration and the entire crown is not replaced; it is called an onlay (Ferracane, 1995). Whenever a dentist replaces a lost teeth, the remaining teeth is used
to support the metallic bridges that span the empty spaces to fill in the oral arch. These bridges are permanently fixed on the teeth with dental cement and are often called fixed partials because they replace only a part of the dentition. The missing teeth are replaced by metallic teeth called pontics, which are attached to the bridge either by casting it as one entire unit or by soldering the individual pieces together. In any case, it is the rigidity of the metal that allows it to be used in this way, similar to the manner in which it is used in bridges that span rivers and canyons (Ferracane, 2001).

It is common to veneer the surface of the metal with porcelain, producing a porcelain-fused-to-metal (PFM) restoration, because a dental bridge made entirely of metal is not esthetically satisfactory. The porcelain must be baked onto the metal framework in an oven, just like pottery and dinnerware are produced. In this case, it is high melting temperature of certain metals that allows them to be used for these applications without melting or deforming at the high firing temperatures used to bake the porcelain. When a patient has several missing teeth, or when it is necessary to simulate lost gingival tissue with the dental prosthesis which often called a dental appliance, a removable partial denture is an option for this case (Craig et al., 2000).

### 2.4.2 Ceramics

A ceramic is compound formed by the union of a metallic and a nonmetallic element. Most of these materials are oxides, formed by the union of oxygen with metals such as silicon, aluminum, calcium and magnesium. Glass, concrete, fine crystal and gypsum are all ceramics. Porcelain is a specific type of ceramic used extensively in dentistry and in other industries. Ceramics may be crystalline or noncrystalline. The
atoms that make up a ceramic may be bonded together by ionic or covalent bonds. Ceramics are generally very brittle materials; they cannot be bent or deformed to any extent without actually cracking and breaking. Everyone is aware of what happens when a ceramic dish or cup is dropped onto a hard surface; contrast that behavior to what happens when a metal fork or knife is dropped. It is easy to understand the difference between brittle ceramics and ductile metals (O’Brien, 2002).

Ceramics are characterized by high melting points and low thermal and electrical conductivity. Therefore, they are used as insulators in many industries. Ceramics are manufactured by fusing oxide powders together in ovens at high temperatures. Most pigmenting agents used in dentistry are ceramic oxides. Their inclusion in appropriate ratios enables the ceramist to produce nearly any color imaginable. This quality also provides the dentist with the ability to match almost any tooth color with esthetic results that are unachievable with other materials (Ferracane, 2001).

Finally, the fact that these materials are oxides meant that they are very inert (not very chemically reactive). This distinct quality provides ceramics with an unequaled biologic compatibility. Sometimes, the patient’s body treats them as if they were actually the same as the natural bone or teeth, which in essence are biologically, produced ceramics. Glass ceramics are used extensively as reinforcing agents, or fillers for dental composites. They are also used in several dental cements and temporary restorative materials (Tham et al., 2010).

As mentioned earlier, ceramics have been used routinely as coatings or veneers to improve the esthetics of metallic dental restorations. The use of ceramics in dentistry
is somewhat limited to date, because of their lower fracture resistance compared to that of metals. Recently, materials with improved fracture resistance have been developed for inlays, onlays and full crown restorations because of their good biologic properties. Ceramics are also used extensively as implant materials, either alone or as coatings for metal substructures made from titanium that are placed directly into the mandible or maxilla (AL Amri, 2004).

2.4.3 Polymers

Usually, polymers are expressed in terms of structural unit (or monomer) and have two or more of the available sites and links related to each other by covalent bonds in the polymer molecule. These units can be arranged and connected in a variety of ways. Types in the straightest forward of the polymer and the units are connected to one another in straight chain arrangement (Bower, 2002). A polymer can either be natural or synthetic. Synthetic polymers are formed by addition or condensation polymerization reactions of monomer. If two or more different monomers are involved, a copolymer (such as ethylene vinyl acetate) is obtained (Harper et al., 2003).

2.4.3.1 Natural Polymers

Polymers derived from plants and animals are called natural polymers, have been used for many centuries, this material includes wood, rubber, cotton, wool, leather, and silk. Other natural polymers such as cellulose, starches, enzymes and proteins are important biological and physiological processes in plants and animals. The tools of modern scientific research made a possibility of the determination of the molecular
structures of this group of materials and the development of numerous polymers, which are assembled from small organic molecules (Callister, 2006).

2.4.3.2 Synthetic Polymers

Man-made polymers can be produced inexpensively and their properties can be tailored to a higher degree of the superiority to natural counterparts. Plastics which has the characteristics of a satisfactory and may produce a lower cost replaced other materials such as metal and wood in some applications. The synthetic polymers intricately linked to the properties of the structural elements of the material. We have many useful plastics, rubber, and fibrous materials and synthetic polymers. In fact, since the end of World War II, the area of materials has been virtually revolutionized by the advent of synthetic polymers (Callister, 2006).

In comparison to metals and ceramics, the relatively weak interaction between the polymer chains reduces the structural and thermal stability of the materials (Ferracane, 2001 & Jordan et al., 2005). Due to their good stability and strength, polymers have been used extensively in dentistry as permanent materials. They are used to make both the teeth and base of dentures, appliances that completely replace the teeth and gums of an edentulous person. Polymers are also used extensively as temporary restorative materials for single restorations and bridges to be worn while the permanent metallic or ceramic restoration is being fabricated by laboratory (Ferrance, 2001). They are used as adhesive agents to enhance the bonding between various materials and tooth structure, or as sealants of the pits and fissures present on occlusal surfaces of permanent teeth. When mixed with glass particles, polymers are formed into a dental composite that
used as an anterior or posterior restorative material much in the same way as amalgam. Their limited structural stability still restricts their use, but improvement in fracture and wear resistance have greatly expanded their application (Abudalazez, 2008).

2.5 Factors Influencing the Properties of Polymers

2.5.1 Molecular Weight

The molecular weight of a polymer is equal to the number of repeating units (i.e. the degree of polymerization) multiplied by the molecular weight of the repeating unit. Thus, the degree of polymerization describes the molecular size. High molecular weight polymers have varying degree of polymerization or molecular weight distribution among the chains within individual polymer batch (Yu et al., 2004). In both addition and condensation polymerization, the length of the chain is determined by purely random events, not all of the chains will be of the same length. In general, many different chain lengths will be present; thus, the molecular weight can only be represented by an average value.

There are a number of ways in which the molecular weight can be determined for a polymer. The two main ones are the number average molecular weight and the weight average molecular weight. Number average molecular weight is obtained by counting the number of molecules in a given weight of sample. Weight average molecular weight is obtained by measurement of the weight of the molecules in the total sample weight (Noort, 2002). The molecular weight distribution of polymers plays an important role in the physical properties of the polymer. In general, a narrow molecular weight distribution gives more useful polymers (Bajpai et al., 2008). The softening point and