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# Ceramic Filled Polymer Matrix Composite Used For Bio-Medical Application

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## Abstract

Synthetic polymers such as polyurethane are used widely in the field of biomedical applications such as implants or part of implant systems.

This work focuses on the preparation of base polymer matrix composite materials by (Hand Lay-Up) method ,and studying the effect of selected weight fractions (3 , 6 , 9 , 12 ,15) % wt of Calcium oxide (CaO) , Calcium carbonate ($CaCO_3$), Magnesium oxide (MgO) ,and Magnesium carbonate ($MgCO_3$) particles on some properties of the prepared composite.

Some mechanical tests were used to evaluate the prepared system (Tensile, Compression, Impact, and Hardness) tests, and a physical test of (Water absorption %), and all tests were accomplished at room temperature.
maximum results of tensile strength, compression strength, impact energy, hardness, water absorption were ( (31 MPa),( 51 MPa),( o.265 J), (79.8 Shor (D)), (0.229 %)) at using ( (9% wt ($MgCO_3$)) , ( 9% wt of ($MgCO_3$)) , ( 9% wt of ($CaCO_3$)) , ( 15% wt of (MgO)) , ( 15% wt of (MgCO3)) respectively.

**Keywords:** Bio- composites, Polyurethane- matrix composite bio- material, Polymer- matrix bio- material, Mechanical properties of bio- composites.

# تحضير مادة متراكبة ذات أساس بوليمر- سيراميك والمستخدمة للاغراض الطبية

## الخلاصة

البوليمر الصناعي مثل البولي يوريثان مستعمل على نحو واسع في حقل التطبيقات الطبية الحيوية مثل الزروعات أَو جزء من أنظمة الزرع.

يركز هذا العمل على تحضير مواد متراكبة ذات اساس بوليمري بطريقة (الصب اليدوي)، ودراسة تأثير الكسور الوزنية المختارة (3 , 6 , 9 , 12 ,15) % من أوكسيد الكالسيوم (CaO)، كاربونات الكالسيوم ($CaCO_3$)، أوكسيد المغنيسيوم (MgO)،كاربونات المغنيسيوم ($MgCO_3$) على خواص المواد المتراكبة المحضرة، حيث المادة المتراكبة من راتنج البولي يوريثان (PU).

عدد الاختبارات الميكانيكية التي أجريت لتقييم النظام المتراكب المحضر تتضمن (الشدّ، الانضغاط، الصدمة والصلادة)، بالإضافة الى الاختبار الفيزيائي (النسبة المئوية لامتصاصية الماء)، وكل الاختبارات اجريت في درجة حرارة الغرفة.

النتائج القصوى لمقاومة الشد ومقاومة الانضغاط وطاقة الكسر والنسبة المئوية لامتصاص الماء كانت ((MPa 31)، (MPa51) ، (o.265 J)، (79.8 Shor (D))، (0.229 %)) عند أستخدام ((9% wt ($MgCO_3$)) ، (9% wt ($MgCO_3$))، (9% wt ($CaCO_3$))، (15% wt (MgO))، (15% wt ($MgCO_3$))) على التوالي.

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## Introduction

Biomaterials play a key role in the replacement of body parts and restoration of human anatomical structures [1]. Trauma, degeneration and diseases often make surgical repair or replacement necessary when a person has joint pain. The main concern is the relief of pain and return to a healthy and functional life style [2].

The medical applications currently in position to present significant new opportunities for manufacturers of advanced composites. With the greater need for prosthetics that are lightweight and materials used in surgical implants that are inert and do not react with biological tissue, the composites industry is in a unique position to fill these demands. Some companies have been producing products to fit these niches and some are now developing less expensive methods and materials in anticipation of changes such as limitations on cost reimbursements [3]. Plastics are used in many types of medical devices from catheters to tissue adhesives. The metals and ceramics are generally so much more rigid than hard tissues that their use for most tissue replacement or augmentation is counter-intuitive, it is far more logical to use polymers for these purposes because of their greater similarity to natural structures [4].

Polymers are long-chain molecules that are formed by connecting large numbers of repeating units (monomers) by covalent bonds [5]. Polymers form the largest category of diverse biomaterials. Based on their source of origin, they can be categorized as synthetic (e.g. polyethylene) or natural type (e.g. collagen). Synthetic polymers can be further sub-divided into biodegradable And non- degradable types [3]. In the degradable type, the polymer is broken down in vivo due to hydrolytic and enzymatic degradation [6]. The resultant nontoxic compounds include lactic and glycolic acid, respectively [3, 6]. One of the key issues while considering polymers for bio applications is their biocompatibility with the host tissue and their degradation characteristics over extended periods of time. Biopolymer applications range from drug release carriers, implants, tissue regeneration scaffolds to sutures [3, 4].

However, metals and ceramics are not biodegradable and their processing ability is very limited. Polymeric materials have received increasing attention and been widely used for tissue engineering because of easy control over biodegradability and processability, Resorbable polymers belong to the group of biodegradable polymers [3]. The special feature of biodegradable polymers is their comparatively fast degradation to biologically harmless substances. Polymers degrading to substances that are not only non - toxic but also can be metabolized by the human organism are called resorbable or bioresorbable. This characteristic makes resorbable polymers particularly interesting for medical applications [6].

One of the primary restrictions on clinical use of bioceramics is the uncertain lifetime under the complex stress states, slow crack growth, and cyclic fatigue that arise in many clinical applications. Two creative approaches to these mechanical limitations are use of bioactive ceramics as coatings or in composites [3]. Composite materials are solids which contain two or more distinct constituent materials or phases, on a scale larger than the atomic, and in which properties such as the elastic modulus are significantly altered in comparison with those of a homogeneous material [3, 6]. Accordingly, reinforced plastics such as fiberglass as well as natural materials such as bone are viewed as composite materials, but alloys such as brass are not natural biological materials that tend to be composites. Natural composites include bone, wood, dentin, cartilage, and skin in biomaterials, it is important that each constituent of the composite be biocompatible [3].

## Experimental Work:

**a-** The masses of the reinforcement materials (Calcium oxide (CaO), Calcium carbonates ($CaCO_3$), Magnesium oxide (MgO) and Magnesium carbonates ($MgCO_3$). were calculated according to the required weight fractions.

**b-** The masses of the resin (polyurethane) were calculated, according to the required volume of cast, the hardener, and added as a weight % with an amount of (3:1).

**c-** The reinforcement filler of (CaO, $CaCO_3$, MgO and $MgCO_3$) and the matrix were mixed at room temperature continuously and slowly to avoid bubbling during mixing, and then the hardener was added to the mixture with mixing gently.

**d-** The mixture was poured from one corner into the mould (to avoid bubble formation which causes cast damage) and the uniform pouring is continued until the mould is filled to the required level.

**e-** The mould was placed on an electrical vibrator to remove any residual bubbles (i.e. captured gas that evaporates during mixing and temperature rise) and to guarantee the distribution of the cast inside the mould.

**f-** The mixture was left in the mould for (24) hrs at room temperature to solidify. Then the cast was aged for one week to dry off. This step was important to reveal complete polymerization, best coherency, and to relieve residual stresses.

**g-** The specimens were cut according to the standard dimensions for each test, using different cutting tools.

**h-** The weight fractions of the filler materials were (3, 6, 9, 12, 15) Wt%.of polymer resin, except $TiO_2$ that was 0.5%wt fixed with all fillers.

**i-** Water absorption test was performed according to (ASTM D 570- 98) at room temperature [7]. Tensile test was performed according to (ASTM D638M- 87b) at room temperature [7, 8]. Compression test was performed according to (ASTM D695- 85) at room temperature [9]. Impact test was performed according to (ISO- 179) at room temperature [9, 8]. Hardness test was performed by using shore hardness (D) and according to (ASTM D 2240) at room temperature [8].

## Results and Discussion

### Water Absorption

Some studies had revealed that mechanical properties such as toughness and resistance will fall after exposure the specimen for moisture, or after absorption of specific quantity of water and the reason for this, is that the moisture has a role in breaking the interface between the matrix material and the reinforced material and by reducing the adhesion between the matrix material and the reinforced material, or the reinforced material may absorb greater amounts of the water, causing swelling of the matrix material and therefore separation in the matrix material will be expected and the detoration of the interface in the composite material will happen and this leads to decreasing in transfer of stress to the filler material and thus lead to decrease in toughness and resistance will be decreased **.**

Figure (1) shows the relationship between the weight fraction of particulate filler and the percentage of water absorption, where we note an increase in the value of absorption slightly when fillers of (CaO, $CaCO_3$, MgO, $MgCO_3$) were added to the polyurethane. The reason for this is that these materials have low absorption coefficient. No enhancement was observed in decreasing the water absorption % when fillers within scope of our work were added i.e.(CaO, $CaCO_3$, MgO, $MgCO_3$) respectivly , compared with water absorption of the base polymer matrix of polyurethane i.e.(0.137%) , but at least there will be a chance in decreasing the acidity of the by-product when the polyurethane starts degradation because of our choice, that is alkaline fillers will tend to normalize the acidic by- product, avoiding or decreasing by that any expected inflammatory that may initiate at the site of bio-implantation.

**Mechanical PropertiesTensile Test.**

Figure (2) shows the relationship between tensile strength with weight fraction for material composite of polyurethane reinforced by (CaO, $CaCO_3$, MgO and $MgCO_3$). Figure (2) reveals that tensile strength of the prepared composite material, will increase with increase in the weight fraction up to 9%wt for all types of mentioned fillers, due to the power of high linkage between the matrix material and reinforcement material, which leads to reducing the slip during the tension.

Further explanation is that there are particulates easily penetrate the matrix and especially when volume of particulate is small as well as ease of penetration of these particulate in the glades located within matrix material. This will increase the ability of the media on wettability and especially when be the media of liquid material before the completion of the process of hardening of the composite material, the increase in property of wettability between the media and reinforced materials will increase from surface area for interface between the reinforced materials and matrix material and among the reinforced material itself.

Composite material supported by particulate does not depend only on the properties of components, but also depends on the nature of the interface between the components and the weight fraction and sometimes on the geometry of the particulate [7, 9, 10].

The value of the tensile strength for the pure polyurethane is (15.5 MPa) and this value had improved when filler of (CaO, $CaCO_3$, MgO and $MgCO_3$) were added which reached its maximum amount (31, 26.5 29.5 and 27) MPa respectively at weight fraction of (9%wt). Results had revealed that the addition of carbonates gives values of tensile strength higher than their oxides.

While the decrease in the tensile strength when exceding the weight fraction of 9%wt , may be due to the bond weakness, as well as the increase in the weight fraction of particulates had made it difficult to penetrate the matrix material when the weight fraction was more than (9%wt). Additions of (CaO, $CaCO_3$, MgO and $MgCO_3$) resulted in further increase in filler amount that will reduce the convergence between the matrix material surface and particulate filler of each other adequately. the process of wetting for surface of particulates by liquid matrix material is incomplete, leading to weakening of the bond between matrix material and the filler material which reduces the efficiency of transfer of load from composite material by the matrix material to particulate and therefore composite material will break at less stresses. In addition to the difficulty of penetration which may weaken the forces of adhesion as well as will create a number of defects within the composite material and this will generate many concentration zones of the stresses that will accelerate from the process of the failure of the sample and making the material behave as brittle material [11].

## Compression Test

Figure (3) illustrates the compression strength.of polyurethane base matrix composite filled with different fillers i.e.(CaO, $CaCO_3$, MgO and $MgCO_3$)

Results had revealed that compression strength had increased when adding (CaO, $CaCO_3$, MgO, $MgCO_3$) filler to polyurethane up to weight fraction of 9%wt, and the maximum values were (49, 51, 50, 53) MPa respectively compared to the maximum value of compression strength of pure polyurethane had reached to (47 MPa). Increase in the values of compression strength when adding particulate filler to the composite material may be due to that particulates will impede the cracks movement, while a decrease in compression strength happens when the weight fraction exceeds 9%wt for (CaO, $CaCO_3$, MgO and $MgCO_3$). This may be related to an increase in the viscosity of the liquid at high rates from these particulates, causing difficulty in fluidity for the matrix and penetrate between fillers which reduces wetting for fillers prior to hardening of the matrix causing adhesion decrease between the matrix and the fillers and the emergence of a lot of flaws and gaps within the prepared composite material and eventually lead to lower compression strength at high rates of fillers. As is known, adhesion force will decrease significantly with increase in weight fraction of fillers [12, 13].

## Impact Test

Impact test is used to determine the tolerance range of composite material for impact force through the absorbed energy by these materials to resist this kind of power .

The impact test is different from the rest of the mechanical tests because it is very fast, where the specimen will be subjected to the rapid stress leading to change in the behavior of material. Figure (4) shows the behavior of the prepared composite materials depanding on the change of impact energy with the change of the fillers type and their weight fraction .

Results in figure (4) revealed that the fracture energy (Uc) increases with increasing the weight fraction of (CaO, $CaCO_3$, MgO and $MgCO_3$) up to 9%wt ,and reach its maximum amount, i.e. (0.195, 0.265, 0.22, 0.25) J respectively compared to the fracture energy of pure polyurethane (0.15) J.

The increase in fracture energy may be related to the particulate fillers that may act as obstacles that will retard the crack growth in the prepared composite system and this will cause the crack deflection in shape and direction i.e. blunting of crack tip will be expected, then toughness being increased.

While decrease in the fracture energy at weight fraction higher than 9%wt for carbonates and their oxides, may be related to the bond weakness between the matrix and the particulate filler [14].

## Hardness test

Hardness test types Shore (D) were carried out for polyurethane (PU) before and after particulate filler addition. Figure (5) shows the effect of weight fraction for the particulate addition on hardness.

As shown in fig (5) hardness for the polyurethane will be increases by adding all types of fillers, i.e. (CaO, $CaCO_3$, MgO and $MgCO_3$) and the hardness will keep increasing with increasing weight fraction, and the concept of hardness can be adopted as a measure of plastic deformation, where material will suffer under the influence of external stress and the addition of particulate fillers will contribute in raising the hardness as a consequence of increased resistance to plastic deformation**.**

Hardness had reached its maximum value at weight fraction of 15% for (CaO, $CaCO_3$, MgO and $MgCO_3$) i.e. (77.4, 74.5, 79.8, .75.8) Shore (D), compared with the hardness of the pure polyurethane i.e. (70.2) Shore (D) .Generally oxides had revealed higher hardness compared with their carbonates selected within the scope of this work.

## Conclusions

Tensile test results had shown that tensile strength has increased with increasing weight fraction of (CaO, $CaCO_3$, MgO and $MgCO_3$) filler particles and reached their maximum value at (9 % wt). Furthermore, the increasing weight fraction revealed decreasing properties of the prepared system.

Also results had shown that compression strength and impact energy increased with increasing weight fraction of (CaO, $CaCO_3$, MgO and $MgCO_3$) up to 9%wt, while further increase in weight fraction revealed a decrease in the properties of the prepared system.

Hardness test results had shown that hardness increase with increasing weight fraction of (CaO, $CaCO_3$, MgO and $MgCO_3$) filler particles and reached their maximum value at (15 wt%) of addition, and the hardness of the samples filled with (oxides) particles was higher than that filled with (carbonates) particles .

The water absorption percentage as a physical property for the prepared composite material had shown an increase with increasing weight fraction of (CaO, $CaCO_3$, MgO and $MgCO_3$) filler particles.

The improvement of polyurethane resin with carbonates filler particles had shown greater values for the fore-mentioned properties than the improvement of polyurethane resin with oxides filler particles, except for hardness value.

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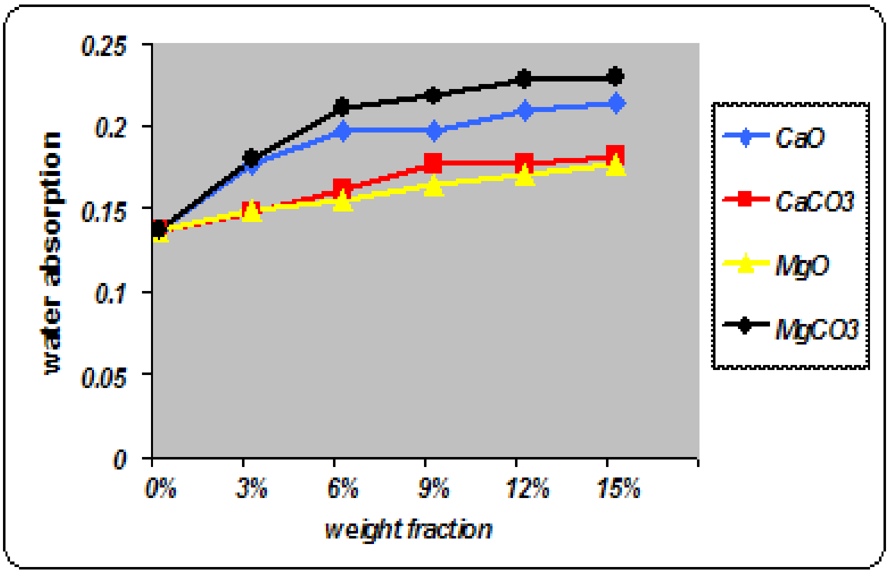

Figure (1) the relationship between the water absorption percentage and weight fraction for polyurethane resin filled with (CaO, CaCO3, MgO and MgCO3) powder, at a rang of (3-15) %wt.

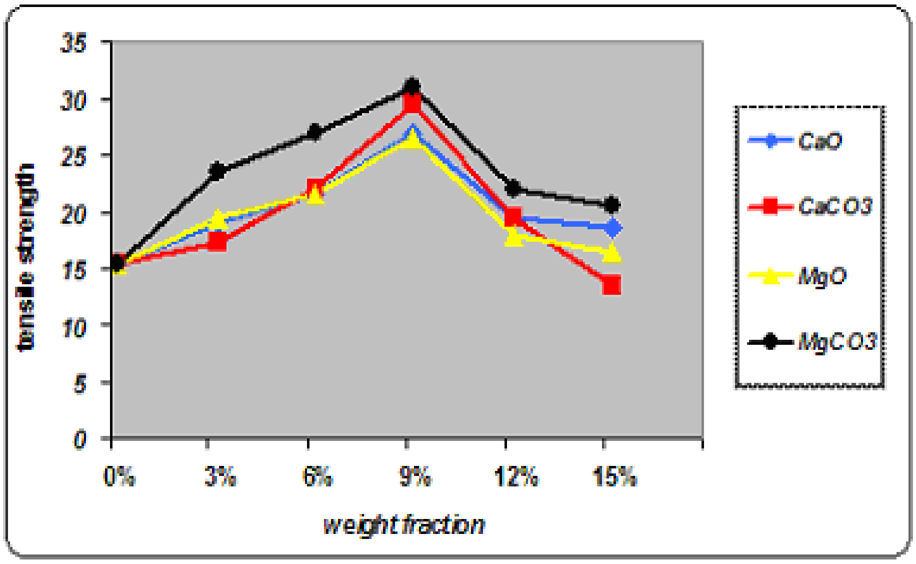

Figure (2) the relationship between the tensile strength and weight fraction for polyurethane resin filled with (CaO, CaCO3, MgO and MgCO3) powders, at a rang of (3-15) %wt.

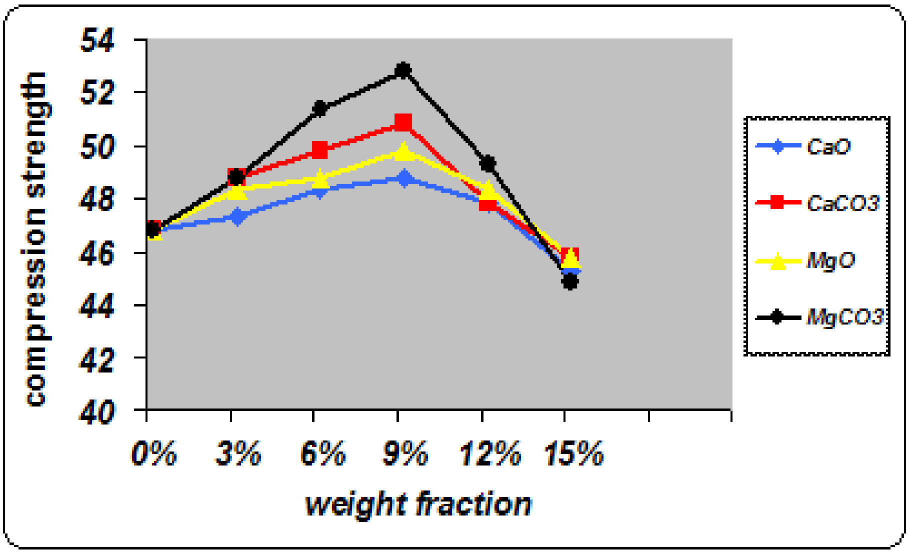

Figure (3) the relationship between the compression strength and weight fraction for polyurethane resin filled with (CaO, CaCO3, MgO and MgCO3) powders, at a rang of (3-15) %wt.

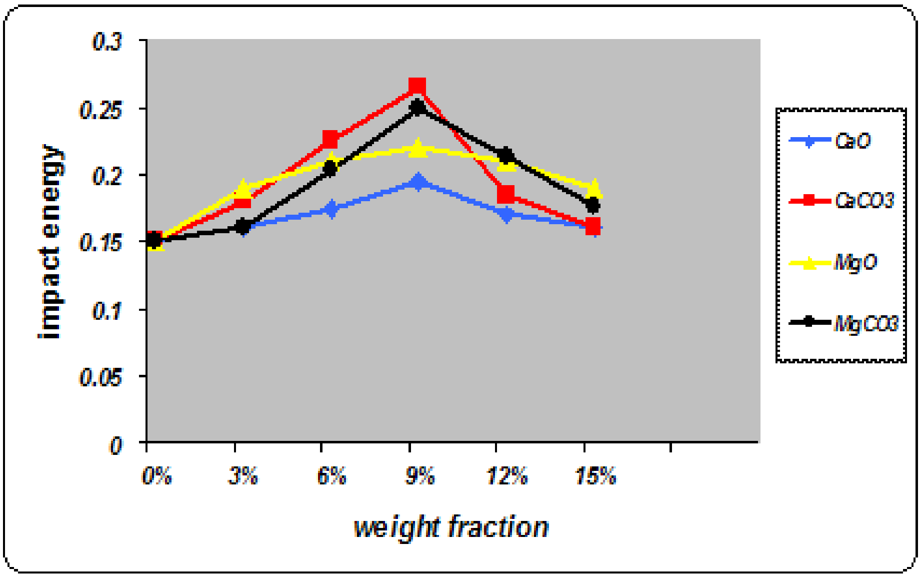

Figure (4) the relationship between the impact energy and weight fraction of polyurethane resin filled with (CaO, CaCO3, MgO and MgCO3) powders, at a rang of (3-15) %wt.

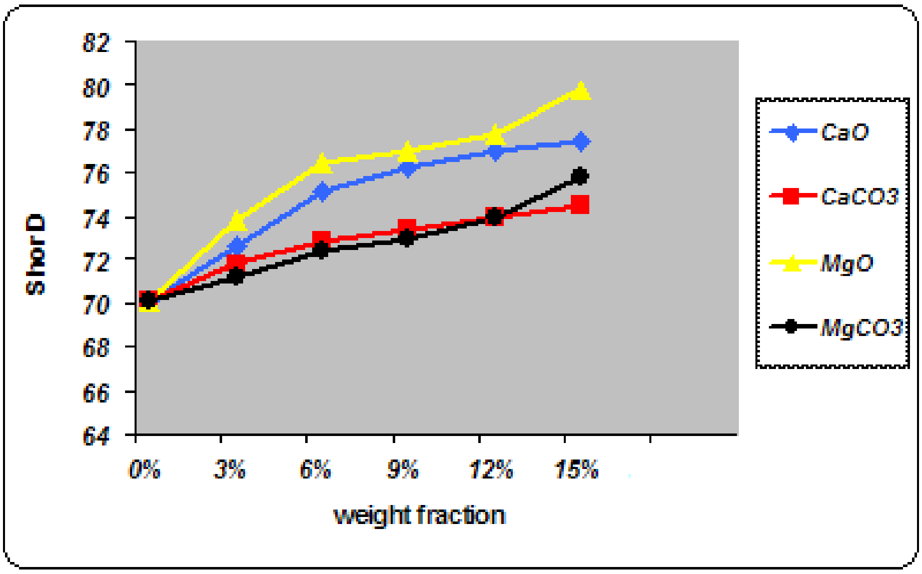

Figure (5) the relationship between the hardness and weight fraction for polyurethane resin filled with (CaO, CaCO3, MgO and MgCO3) powders, at a rang of (3-15) %wt.