

LAMPIRAN

FABRIKASI TRICALCIUM PHOSPHATE BERPORI MENGGUNAKAN PARTIKEL WHEAT SEBAGAI AGEN PEMBENTUK PORI

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Abstract

The present work reports a simple process for fabrication of tricalcium phosphate (TCP) ceramics from aqueous powder slurries using wheat particles as pore forming agent. Wheat particles incorporated in aqueous TCP slurries rapidly absorb water on heating and transform it into a strong gel. The dried green bodies showed shrinkage in the range 53,22-55,87%. Removal of the pore former followed by sintering at 1000 and 1100°C produced TCP bodies with porosity 59,48–78,40% and compressive strength 0,30-2,53 MPa.

Keywords: Compressive strength; Porosity; Tricalcium phosphate; Wheat particles

1. Pendahuluan

Tulang merupakan jaringan yang berfungsi sebagai rangka, penyokong dan pelindung organ tubuh serta sebagai penghubung antar otot sehingga memungkinkan terjadinya gerakan [Rivera-Munoz, 2011]. Kerusakan/cacat pada tulang mengakibatkan terganggunya fungsi tersebut sehingga tulang perlu diperbaiki. Dewasa ini, penggunaan *biomaterials* sebagai tulang implan merupakan salah satu alternatif yang telah banyak dikembangkan. *Biomaterials* merupakan material yang berfungsi mengembalikan dan meregenerasi jaringan hidup yang rusak [Park *et al.*, 2000]. *Autograft*, *allograft* dan *xenograft* adalah *biomaterials* yang umumnya digunakan untuk perbaikan dan penggantian jaringan tulang [Dumitrescu, 2011]. Ketersediaan dalam jumlah yang terbatas, rasa sakit yang ditimbulkan dan risiko penularan penyakit merupakan kelemahan dari material ini. Oleh karena itu, perlu adanya alternatif yang mampu mengatasi keterbatasan material sebelumnya, seperti penggunaan *biomaterials* sintetik.

Tri kalsium fosfat (TCP) adalah *biomaterials* sintetik yang memiliki kemampuan untuk berinteraksi dengan jaringan tubuh manusia. β -TCP merupakan *bioceramics* dengan sifat *biocompatibility* yang baik dan dapat berperan dalam pertumbuhan dan regenerasi tulang [Uchida *et al.*, 1984]. Aplikasi TCP dalam bidang medis terutama berfokus pada TCP berpori. TCP berpori telah digunakan sebagai *drug-releasing agent* seperti antibiotik, anti tumor dan anti inflamasi serta dipakai dalam implantasi jaringan [Kalita *et al.*, 2007]. Morfologi pori keramik dapat dibentuk melalui beberapa metode, salah satunya adalah penggunaan *wheat particles* pada *starch consolidation*. Penggunaan *wheat particles* memiliki beberapa

keunggulan, yaitu sifatnya yang mudah terlepas (*easy to burn out*), harganya murah, ramah lingkungan dan mampu menghasilkan keramik dengan distribusi pori yang tersebar merata [Abdurrahim & Sopyan, 2008].

2. Metodologi

Bahan baku penelitian meliputi bubuk TCP (Merck, Jerman), *wheat particles* (PT Indofood Sukses Makmur Tbk, Indonesia) dan HNO₃ (Merck, Jerman). Akuades berperan sebagai pelarut, *wheat particles* berfungsi sebagai pembentuk pori sedangkan HNO₃ digunakan sebagai zat untuk mengatur pH campuran menjadi 3,5.

Penelitian ini dimulai dengan persiapan *slurry*. TCP bubuk dicampur dengan akuades kemudian ditambahkan *wheat particles*. *Slurry* yang terbentuk lalu ditambahkan HNO₃ dan diaduk dengan kecepatan 150 rpm. Campuran tersebut kemudian dicetak ke *mould* yang sebelumnya diolesi minyak sawit (PT Multimas Nabati Asahan, Indonesia) sebagai pelumas. Selanjutnya campuran dalam *mould* dipanaskan pada 100°C selama 30 menit. Setelah itu *green bodies* dilepas dari *mould* dan dikeringkan dalam oven pada 80°C selama 24 jam dan 120°C selama 8 jam. Sampel yang telah kering tersebut kemudian dimasukkan ke dalam *furnace*. Pembakaran dilakukan pada temperatur 350°C, diikuti dengan temperatur 600°C dan diakhiri dengan *sintering* masing-masing selama 1 jam.

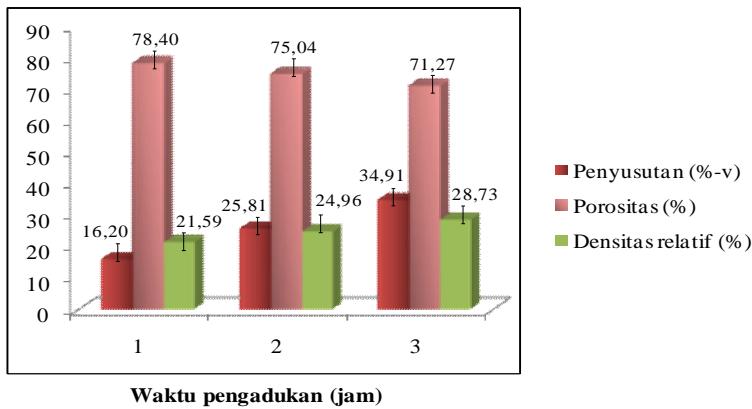
3. Hasil dan pembahasan

3.1 Porous Body Properties

Wheat particles dan air akan membentuk gel melalui pemanasan. Partikel terdispersi sempurna dalam air tanpa adanya aglomerasi. Tiga puluh lima gr *wheat particles* akan menyerap 100 ml air pada 100°C [Prabhakaran *et al.*, 2007]. Pengadukan dilakukan selama 1, 2 dan 3 jam. Selama pengadukan berlangsung, campuran akan mengental membentuk pasta. Pasta tersebut selanjutnya dimasukkan ke dalam oven untuk menghasilkan *green bodies* yang mengalami penyusutan 53,22-55,87%. Persentase penyusutan setelah *drying* dapat dilihat pada Tabel 1.1 untuk setiap variasi waktu pengadukan. Dari Tabel 1.1 dapat dilihat bahwa semakin lama waktu pengadukan maka persentase penyusutan akan semakin kecil. Gambar 1.3 menunjukkan foto *green bodies* dengan waktu pengadukan 1-3 jam.

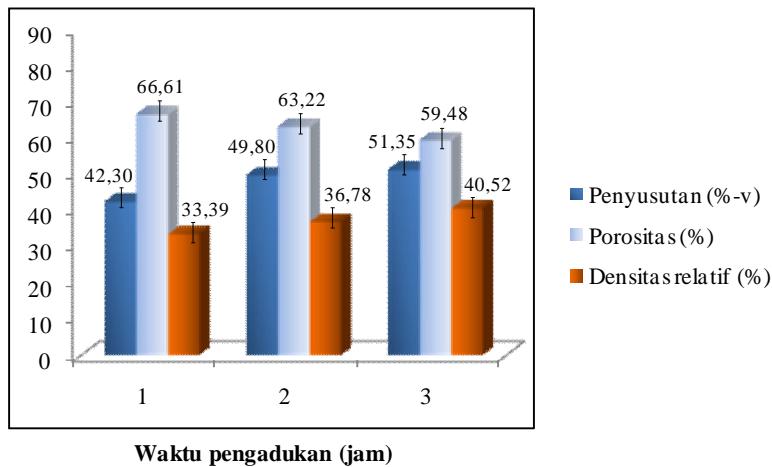
Tabel 1.1 Persentase penyusutan setelah *drying*
(TCP = 12 gr, Wheat = 6 gr, Aquadest = 35 mL)

Pengadukan (jam)	Penyusutan <i>drying</i> (%-v)
1	55,87
2	54,83
3	53,22



Gambar 1.3 Penyusutan, porositas dan densitas relatif setelah *sintering* 1000°C

Sintering dilakukan pada temperatur 1000 dan 1100°C. Hasil menunjukkan tidak terjadi deformasi atau kerusakan pada badan sampel. Gambar 1.3 dan 1.4 memperlihatkan persentase penyusutan setelah *sintering*, porositas dan densitas relatif dari TCP berpori yang diperoleh. Penyusutan sampel setelah *sintering* pada 1000°C terjadi pada rentang 16,20-34,91% sedangkan porositas yang dihasilkan 71,27-78,40%.



Gambar 1.4 Penyusutan, porositas dan densitas relatif setelah *sintering* 1100°C

Penyusutan sampel setelah *sintering* pada 1100°C terjadi pada rentang 42,30-51,35% sedangkan porositas yang dihasilkan 59,48-66,61%. Dari kedua gambar dapat dikatakan bahwa waktu pengadukan yang semakin lama mengakibatkan berkurangnya porositas. Porositas yang semakin kecil menyatakan sampel berstruktur lebih padat sehingga memiliki densitas yang lebih besar. Fenomena ini sesuai dengan HA berpori

yang mengalami kenaikan porositas dari 42,7 menjadi 49,4% dan penurunan densitas dari 1,81 menjadi 1,60 gr/cm³ setelah diaduk selama 4 dan 20 jam [Sopyan & Kaur, 2009]. Gambar 1.3 dan 1.4 menunjukkan adanya kenaikan densitas ketika temperatur *sintering* semakin besar. Kenaikan ini disebabkan partikel semakin kompak dan memadat (densifikasi) pada temperatur yang lebih tinggi. Densitas yang lebih besar akan mereduksi pori dan menghasilkan porositas yang lebih kecil. Hal ini telah dibuktikan oleh Ploeg *et al.*, [2010] dalam fabrikasi TCP pada temperatur *sintering* 950, 1050 dan 1150°C yang menghasilkan densitas 2,27; 2,84 dan 3,22 gr/cm³ dengan porositas 24, 6 dan 0%. Hal serupa juga terjadi pada fabrikasi TCP pada temperatur *sintering* 1450 dan 1550°C, TCP yang diperoleh memiliki porositas 95 dan 90% [Udoh *et al.*, 2010].

3.2 Mikrostruktur

Sintering green bodies pada 1000°C dan 1100°C menghasilkan TCP berpori dengan morfologi seperti pada Gambar 1.5. Gambar 1.5a-c dan 1.5d-f menunjukkan bahwa waktu pengadukan yang semakin lama mengakibatkan kurangnya aglomerasi dan menghasilkan partikel yang lebih homogen. Hal ini sesuai dengan Ramay & Zhang [2003] yang melaporkan bahwa waktu pengadukan yang lama akan mengurangi aglomerasi dan porositas sehingga meningkatkan *compressive strength* dan densitas. Dari Gambar 1.5a-c dapat dilihat bahwa jarak antar partikel sangat rapat sehingga pori yang dihasilkan tidak begitu kelihatan sedangkan Gambar 1.5d-f menunjukkan ukuran pori yang lebih besar dan mengindikasikan bahwa TCP tersebut memiliki pori terbuka dengan interkoneksi antar pori yang baik. Pori terbuka dengan interkoneksi yang baik merupakan karakteristik implan untuk penetrasi tulang dan *osteointegration* [Ravaglioli & Krajewski, 1997].

3.3 Mechanical Properties

Gibson & Asby [1988] melaporkan bahwa kuat tekan (*compressive strength*) keramik berpori akan berkurang seiring dengan kenaikan porositas. Gambar 1.3 dan 1.4 menunjukkan bahwa porositas semakin berkurang ketika waktu pengadukan semakin lama. Dari Gambar 1.6 terlihat ketika waktu pengadukan semakin lama (porositas semakin kecil) maka kuat tekan TCP semakin besar. Pada 1000°C, kuat tekan TCP adalah 0,30-2,32 MPa sedangkan kuat tekan pada 1100°C berkisar 0,98-2,53 MPa. Secara umum, terdapat beberapa sampel yang termasuk pada rentang kuat tekan *cancellous bone*, yaitu berkisar 1-100 MPa [Lanza *et al.*, 2000].

4. Kesimpulan

Fabrikasi tri kalsium fosfat (TCP) menggunakan *wheat particles* sebagai agen pembentuk pori telah berhasil dilakukan. Ukuran makropori TCP yang diperoleh adalah 300-310 µm dengan porositas berkisar 59,48–78,40% dan kuat tekan 0,30-2,53 MPa. Pada 1000°C, mikrostruktur TCP menunjukkan jarak antar partikel yang rapat dengan pori yang sangat kecil sedangkan pada 1100°C pori yang dihasilkan lebih besar dengan interkoneksi antar pori yang baik.



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Fabrication of Porous Tricalcium Phosphate Scaffolds by Starch Consolidation Method

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Abstract

The present work reports a simple process for fabrication of tricalcium phosphate (TCP) ceramics from aqueous powder slurries using wheat particles as pore forming agent. Wheat particles incorporated in aqueous TCP slurries rapidly absorb water on heating and transform it into a strong gel. The dried green bodies showed shrinkage in the range 53,22–55,87%. Removal of the pore former followed by sintering at 1000 and 1100°C produced TCP bodies with porosity 59,48–78,40% and compressive strength 0,30–2,53 MPa.

Keywords: Compressive strength; Porosity; Tricalcium phosphate; Wheat particles

1. Introduction

Porous TCP ceramic is essentially a bioresorbable calcium phosphate. Its tissue biocompatibility is excellent and it may act as a scaffold allowing bone regeneration and ingrowth. Because of its resorbable characteristic, the porous TCP is regarded as an ideal biomaterial for bone substitutes that should degrade by advancing bone growth [1]. In this regard, most important aspects in fabrication of scaffolds are porosity, pore size, pore interconnectivity and mechanical strength. In the terms of microstructure of the mineral phase, the bone can be classified as cortical and cancellous. Cortical bone has porosity in the range 0-30%. Porosity of cancellous bone is 30-90% [2]. Hulbert et al. [3] suggested that the optimum pore size for osteoconduction is 150 µm. Pore interconnectivity allows circulation and exchange of body fluids, ion diffusion, nutritional supply and osteoblast cell penetration [4]. Compressive strength of porous human bones vary between 2-12 MPa for cancellous bone and between 100-230 MPa for cortical bone [5].

A number of technique have been developed to fabricate porous TCP scaffolds, typically including gel casting of foams and replication of a polymer sponge. Gel casting or ceramic foaming technique can be applied to produce ceramic scaffolds with high mechanical strength. The disadvantage of this technique is that it typically results in a structure of poorly interconnected pores and non-uniform pore size distribution. Fabricating porous ceramic via the replication of a polymeric sponge produced open-celled porous ceramics. Porous ceramic prepared via this technique have shown well interconnected pores but has poor mechanical strength for load bearing applications [6].

Lyckfeldt and Ferreira [7] reported a starch consolidation process for

fabrication of porous ceramics. In this process, native and chemically modified potato starch granules were incorporated into aqueous ceramic powder suspensions. Characteristics such as higher water absorption and swelling of starch at 60-80°C is used for the gellation of the slurry. The process produces porous ceramics with 30-70% porosities. Wheat is a low cost starchy material which can be used as gelling and pore forming agent in fabrication of porous TCP ceramics.

2. Experimental

Tricalcium phosphate (Merck, Germany) and wheat particles (food grade and procured from local market) were used. Distilled water was used for preparation of slurries. Nitric acid (Merck, Germany) was used for pH adjustments. Twelve gr of TCP powder were mixed by 35 ml of distilled water. Then the mixture was added by 6 gr of wheat particles. Adjusting the pH to 3.5 using dilute nitric acid. The slurry was stirred for 1, 2 and 3 hours. Then the slurry was cast in cylindrical open stainless steel mould and heated in oven at 100°C for 30 min. Vegetable oil was used as lubricant for easy mould removal. The gelled body removed from the mould was dried in oven at 80°C for 24 h and 120°C for 8 h. The dried samples were burned in a furnace at a rate of 2°C/min up to 350°C, 2°C/min up to 600°C and at a rate of 5°C/min up to sintering temperature (1000 and 1100°C). A holding time of 1 h was given at 350, 600 and sintering temperature. Density of the sintered samples was determined from dimensional measurement. Mechanical strength of the sintered samples was measured in a universal testing machine (Llyold LR10K Plus). Microstructure of sintered samples was observed in a scanning electron microscope (Phenom Pro-X, Netherlands).

3. Results and Discussion

3.1 Porous Body Properties

Wheat particles when mixed with water slowly absorb water at room temperature. The particles in wheat particle-water mixture at room temperature are found non sticky. However, on heating, particles absorb water rapidly and transform the mixture into a strong gel. It has been observed that 35 gr of the wheat particles absorb 100 ml water at 100°C and form a strong gel [8]. The gelled bodies undergo shrinkage on drying in the oven. Fig. 1 shows green bodies of the samples. We can see that shrinkage decreases with increasing stirring time.

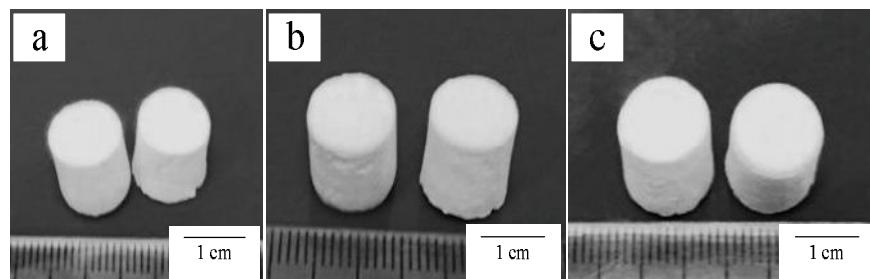


Fig. 1. Green bodies from various stirring time (a) 1 (b) 2 and (c) 3 hours

The drying shrinkage values for gelled bodies prepared from various stirring time is 55.87, 54.83 and 53.22% for 1, 2 and 3 hours, respectively.

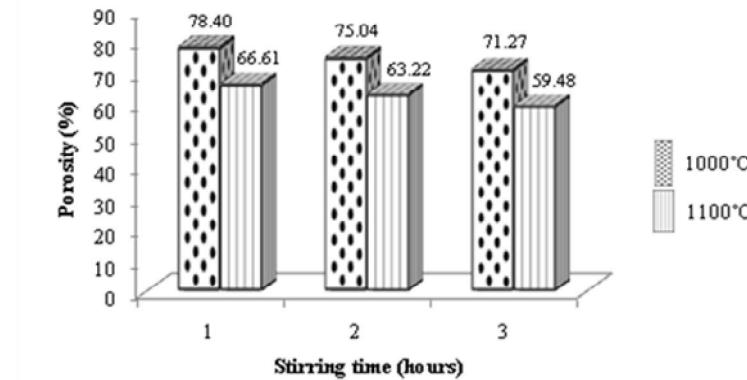


Fig. 2. Porosity of TCP sintered bodies

The bodies did not show any deformation or cracks during burnout of pore former and sintering. Porosity of the samples is in the range 59.48-78.40%. Fig. 2 shows porosity values of sintered samples from various variable. The porosity decreases with increasing stirring time. Conversely, a decreasing in porosity resulted in increasing density. The findings obtained in this research are in agreement with a study by Sopyan & Kaur [9] who reported a similar correlation between porosity and density. As the sintering temperature was raised, the porosity decreases but not significantly. These result are in agreement with a study by Udo et al. [10] who reported that TCP porosity decreases from 95 to 90% with increasing sintering temperature from 1450 to 1550°C. A tendency of decrease in porosity with increase in sintering temperature might arise from the fact that when sintering temperature increases, densification rate of particles escalates to form a denser structure with fewer pores.

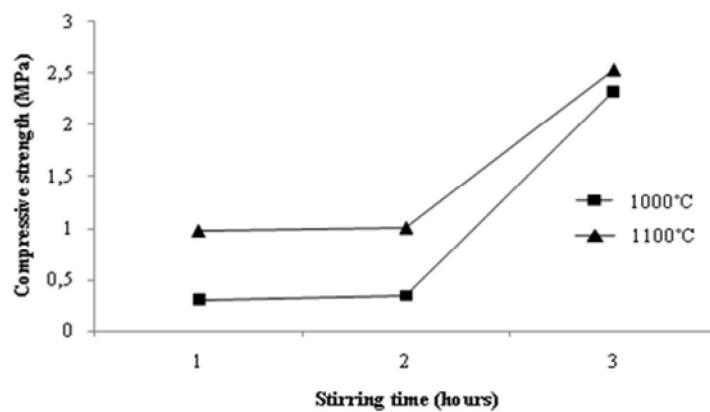


Fig. 3. Mechanical strength of porous TCP

Relationship between the compressive strength with stirring time and sintering temperature is shown in Fig. 3. It could be seen that the strength of samples at stirring time 3 hours was rocketed raise than two before. The strength of sintered samples is in the range 0.3-2.53 MPa. The strength increases with increasing stirring time and sintering temperature. Increasing the stirring time results in the breakdown of agglomerates and the slurry obtained tends to be homogenous. This in turn reduces the porosity and increases compressive strength. In essence, lower porosity was accompanied with higher compressive strength

3.2 Microstructure of Ceramics

The micrographs given in Fig. 4 illustrate the change in morphology of pore structure as the sintering temperature was increased. It could be seen that pore interconnectivity in the porous TCP fired at 1100°C was better than fired at 1000°C. Clinically, this kind of interconnective microporosity is a desirable feature since it facilitates the diffusion of calcium and phosphorus ions for mineralization [11].

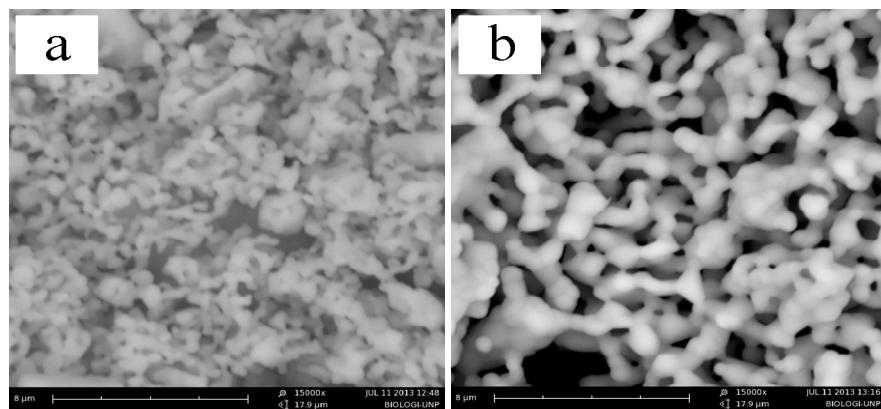


Fig. 4. Microstructure of porous TCP stirred 3 hours (a) 1000°C (b) 1100°C

CONCLUSION

Our findings demonstrated that the high porosity TCP exhibited interconnected pore structure and reasonable compressive strength. While the TCP fired at 1100°C and stirred for 3 hours showed excellent porosity and compressive strength much as human cancellous bone, which suggested that it may be an ideal bioceramic for bone implant.

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