



Biologic potential of calcium phosphate biopowders produced via decomposition combustion synthesis

N. Vollmer^{a,*}, K.B. King^{b,1}, R. Ayers^{b,2}

^aGeorge S. Ansell Department of Metallurgy and Materials Engineering, Colorado School of Mines, 1500 Illinois Street, Golden, CO 80401, USA

^bUniversity of Colorado School of Medicine, Department of Orthopaedics, Aurora, CO 80045, USA

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Abstract

The aim of this research was to evaluate the biologic potential of calcium phosphate (CaP) biopowders produced with a novel reaction synthesis system. Decomposition combustion synthesis (DCS) is a modified combustion synthesis method capable of producing CaP powders for use in bone tissue engineering applications. During DCS, the stoichiometric ratio of reactant salt to fuel was adjusted to alter product chemistry and morphology. In vitro testing methods were utilized to determine the effects of controlling product composition on cytotoxicity, proliferation, biocompatibility and biomineralization. In vitro, human fetal osteoblasts (ATCC, CRL-11372) cultured with CaP powder displayed a flattened morphology, and uniformly encompassed the CaP particulates. Matrix vesicles containing calcium and phosphorous budded from the osteoblast cells. CaP powders produced via DCS are a source of biologically active, synthetic, bone graft substitute materials.

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1. Introduction

Combustion synthesis is an overarching term coined for self-sustaining exothermic reactions that utilize internally generated energy [1] to rapidly produce a desired product. Traditional combustion synthesis methods utilize solid-state reactions of reactant powders [2] whereas decomposition combustion synthesis (DCS) requires the aqueous decomposition and subsequent reconstitution of the reactants into a gelatinous foam prior to ignition [3,4]. Calcium phosphate DCS is a self-propagating high-temperature synthesis (SHS) process that utilizes the heat

generated by the exothermic reaction of urea, calcium nitrate and ammonium phosphate to generate and sustain the propagation of a combustion wave [5–7] through the reactants.

DCS is a combustion synthesis technique that has been used to produce many advanced materials, including calcium phosphate (CaP) [3,8]. An advantage of CaP DCS reactions is that reactant salt and fuel stoichiometries can be modified to produce CaP powders with diverse chemistries and morphologies [9] allowing the final product to be customized to specific manufacturing requirements. In addition, product synthesis time is significantly lower than traditional synthesis techniques such as wet chemistry precipitation [10–14] or solid-state reactions [15–17].

Synthetic CaP materials currently used in bone tissue engineering applications are comprised of hydroxyapatite (HA) [18,19], tricalcium phosphate (TCP) [20] or mixtures with varying percentages of HA and TCP [21–25]. Biphasic, triphasic and multiphasic CaPs are commonly utilized in reconstruction of bone defects [25] and it has been demonstrated that the bioactivity of biphasic CaPs can be directly controlled via manipulation of the CaP composition [21,23,26]. In addition to calcium and phosphate,

*Corresponding author. Present address: 608 Boxwood Road, Woodland, CA 95695. Tel.: +1 303 902 6934.

E-mail addresses: nvollmer@ucdavis.edu (N. Vollmer),

karen.king@UCDenver.edu (K.B. King),

reed.ayers@UCDenver.edu (R. Ayers).

¹Present address: 12800 E. 19th Avenue, Room 2103, Mail Stop 8343, Aurora, CO 80045. Tel.: +1 303 724 1596; fax: +1 303 724 0394.

²Present address: 13001 E. 17th Place, Campus Box B202, Aurora, CO 80045.

Tel.: +1 303 724 6695; fax: +1 303 724 0919.

bone mineral can contain carbonate ions [12,27–29] and trace elements [28,29].

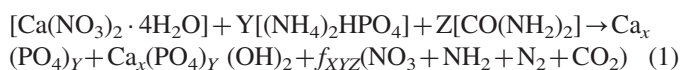
An advantage of the CaP DCS reactions is that product composition can be controlled. Specifically, reactant stoichiometry can be controlled to produce products with specific bi-, tri- or multiphase compositions. Additionally, the CaP DCS reaction system could be modified to include constituents that enhance the mechanical, chemical and biological properties of the synthesized biopowders e.g. AgNO₃ [30] or ionic Ag [31] for antimicrobial effects, Mg²⁺ and F⁻ [32] for increased microhardness and cellular response. Whereas post synthesis heat treatment [13,14] could be applied to the final products to produce pure (single) phase CaP powders. Subsequent to interaction with the human body, a CaP implant with biomimetic properties and compositional similarity to natural bone may reduce healing time, improve tissue integration and improve long term in vivo function [33,34].

DCS represents a synthesis technique for the production of biologically active CaP biopowders that can be used as components in medical devices, bone cements, coatings and bone tissue engineering applications. The intent of this research is to examine the effects of altering DCS reaction stoichiometry on the biologic potential of CaP biopowders synthesized via DCS and to quantify the effects of controlling reaction parameters (e.g. reactant stoichiometry) on biologic activity.

2. Materials and methods

2.1. Decomposition combustion synthesis procedure

A detailed description of the DCS procedure has been given elsewhere [8]. Briefly, salts of calcium nitrate tetrahydrate [Ca(NO₃)₂·4H₂O], ammonium phosphate [(NH₄)₂·HPO₄] and urea [CO(NH₂)₂] were dissolved in 10 ml of de-ionized water in a beaker and heated on a hot plate for 20 min. During heating, the hydrolyzed reactants were vaporized until a white foam formed. Then the beaker with foam was placed into a Blue M™ box-type muffle furnace at 1000 °C (+/-)10 °C until the foam ignited (~2–8 s) [8]. The decomposition of calcium nitrate tetrahydrate into NO₃⁻ and CaO and ammonium phosphate into NH₄⁺ and PO₄³⁻ drives the combustion reaction while the oxidation and complete decomposition of urea energetically sustains the reaction. Reaction 1:



2.2. Powder stoichiometry

The ratio of calcium nitrate tetrahydrate (CaN) to ammonium phosphate (AmP) directly determines the amount of calcium and phosphate in the system [3]. This ratio will be referred to as the calcium to phosphate (c:p) ratio. For example, to obtain a stoichiometric ratio (3:2) of calcium to phosphate in the products, a c:p ratio of 1.5 is needed. Therefore the CaN term (X) in reaction 1 needs to be multiplied by 3 and the AmP term (Y)

needs to be multiplied by 2. c:p Ratios and the coefficients used to obtain these ratios are shown in Table 1.

A stoichiometric fuel ratio (amount of urea to the amount of CaN and AmP) was determined by calculating an elemental equivalence ratio (ϕ_e = Sum of oxidizing and reducing elements in the mixture) for the reaction [9,35]. Calculations for ϕ_e for this system are given by the following equation:

$$3[\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}] + 2[(\text{NH}_4)_2\text{HPO}_4] + Z[\text{CO}(\text{NH}_2)_2] \quad (2)$$

$$3[1]0 + 2[16] + Z[6] = 3$$

Therefore $Z=3=\phi_e$.

Table 2 lists the valences for the reactants in this system. These valences were used to calculate ϕ_e .

Fuel ratios were obtained by multiplying (ϕ_e) by the elemental stoichiometric coefficient multipliers shown in Table 2. For example, to obtain a stoichiometric fuel ratio, the urea term (Z) in reaction 1 is multiplied by 3.

Table 3a and b presents matrices of sample stoichiometries studied. Samples in Table 3a have varying c:p ratios, and a constant fuel ratio (urea:CaN+AmP). Samples in Table 3b have a constant c:p ratio, but varying fuel ratios.

2.3. Product characterization

Scanning electron microscopy (SEM) was performed with a FEI Quanta 600 SEM. High vacuum with a 20 kv beam and spot size of 4.0 created optimal measurement conditions and minimized charging [36]. The CaP powders were sputter-coated with gold (Hummer V) and analyzed for composition, morphology, grain growth and specific structural formations (i.e. Necking).

X-ray diffraction (XRD) patterns were obtained with a Phillips Analytical PW3240 X'PERT data collector using 2θ scans ranging from 10° to 80°. An internal silicon powder (99.9% pure, Alfa Aesar) with a 1:4 ratio of silicon to CaP was used to

Table 1
C:p ratios with their corresponding coefficients.

c:p Ratio	CaN (X) coefficients	AmP (Y) coefficients
1.3	3	2.3
1.5	3	2
1.7	3	1.765

Table 2
Elemental equivalence ratio, ϕ_e , stoichiometric coefficient multipliers and the calculated urea coefficients that were studied, $\phi_e=3$. Valences of elements used in this system.

Elemental stoichiometric coefficient multiplier (ϕ_e)	Urea (Z) Coefficients	Element	Valence
1	3	P	+5
		Ca	+2
1.5	4.5	C	+4
		H	+1
2.5	7.5	O	-2
		N	0

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