SYNTHESIS AND CHARACTERIZATION OF CERAMIC COMPOSITES DERIVED FROM EPPAWALA APATITE FOR AUTOMOTIVE AND INDUSTRIAL APPLICATIONS

By

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Thesis submitted to the University of Sri Jayewardenepura for the award of the degree of Doctor of Philosophy

Declaration of the candidate

The work described in this thesis was carried out by me under the supervision of Dr. R. D. Gunaratne and Prof. H.M.J.C. Pitawala and a report on this has not been submitted in whole or in part to any university or any other institution for another Degree / Diploma.

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Certification of the supervisors

"We certify that the candidate has incorporated all corrections, additions and amendments recommended by the examiners to this version of the Ph.D. thesis."

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DEDICATION

To Dr. Rajitha Gunaratne, Prof. Jagath Pitawala

&

To my family

Appachchi, Amma, Isuru malli, Thanula malli

Without you all, this would still be a dream…….

Thank you so much!!!

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ABBREVIATIONS

ASTM	American society for testing and materials
ATR	Attenuated total reflection
Avg.	Average
BNE	Boron nitride embedded E-glass fiber reinforced epoxy matrix composites
BNNT	Boron nitride nanotube
CMC	Ceramic matrix composites
(CR)-glass	Corrosion resistant glass
CTE	Coefficient of thermal expansion
CVD	Chemical vapor deposition
DC	Deficit counter
DMA	Dynamic mechanical analysis
DMTA	Dynamic-mechanic thermal analysis
DMPT	N, N-dimethyl p-toluidine
DSC	Differential scanning calorimetry
EDS	Energy dispersive x-ray spectroscopy
ERP	Eppawala rock phosphate
FE-SEM	Field emission scanning electron microscopy
FOS	Fiber optic sensors
FRP	Fiber reinforced polyester
FTIR	Fourier-transform infrared spectroscopy
GF	Glass fibers
GIC	Glass ionomer cement
HAp	Hydroxyapatite
HAp-BN	Boron nitride embedded hydroxyapatite composites
HApE	Hydroxyapatite embedded E-glass fiber reinforced epoxy matrix composites
HApP	Eppawala nano hydroxyapatite filler added polyester matrix composites

HApPF	Eppawala nano hydroxyapatite filler added E- glass fiber reinforced polyester matrix composites
HAp-SiC	Silicon carbide embedded hydroxyapatite composites
HEMA	2-hydroxyethyl methacrylate
HERP	Eppawala high-grade rock phosphate
HDPE	High-density polyethylene
hrs.	Hours
(HS)-glass	High strength glass
LDPE	Low-density polyethylene
Lkr.	Sri Lankan rupees
MEKP	Methyl ethyl ketone peroxide
MEMS	Microelectromechanical systems
MESFETs	Metal semiconductor field effect transistors
MFC	Macro fiber composites
MFCs	Magnetic framework composites
MMA	Methyl methacrylate
MMABS	Methyl methacrylate-butadiene-styrene
MMC	Metal matrix composites
MOFs	Metal-organic frameworks
MOSFETs	Metal oxide semiconductor field effect transistors
MRHERP	Moisture removed Eppawala high-grade rock phosphate
MWCNTs	Multi-wall carbon nanotubes
ND	Not in the detection level
NE	Pure cross-linked epoxy material
NP	Cross-linked pure polyester material
PDI	Poly dispersity index
PHBV	Poly hydroxybutyrate co vel
PHEMA	Poly(2-hydroxyethyl methacrylate)
PMC	Polymer matrix composites
PMMA	Poly (methyl methacrylate)
PSA	Particle size analyzer
PVC	Polyvinyl chloride

RD	Reactive distillation
Rf	Reflective index
SBF	Stimulated body fluids
SCWS	Silicon carbide whiskers
SD	Standard deviation
SEM	Scanning electron microscopy
SGHAp1	Sol gel alcoholic hydroxyapatite powder
SGHAp2	Sol gel acidified hydroxyapatite powder
SiCE	Silicon carbide embedded E-glass fiber reinforced epoxy matrix composites
SSHAp	Solid state sintered hydroxyapatite powder
TGA	Thermo-gravimetric analysis
UTM	Universal testing machine
US	United States
UV	Ultra violet
VOC	Volatile organic compounds
wt	Weight
XRD	X-ray diffraction spectroscopy
XRF	X-ray fluorescence spectroscopy

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ABSTRACT

The Sri Lankan Eppawala rock phosphate deposit, found in the North Central province, is a non-renewable natural resource that does not replenish over humanly meaningful time scales. It contains nearly 34% to 40% phosphorous pentoxide (P_2O_5). Even though there are numerous viable and value-added phosphorous (P) industries, Sri Lanka exports rock phosphate as a raw material at lower prices to other developed nations. Considering its resource-related wealth, it is a timely requirement to obtain the proper compensation for this non-renewable resource. Therefore, it is crucial to consider developing and commercializing fresh P-related finds from the Eppawala rock phosphate deposit. In order to value add Eppawala high-grade rock phosphate (HERP), this dissertation thoroughly and critically discusses three patented innovative processes (sol-gel alcoholic route, sol-gel acidified route, and solid-state sintering technique) for synthesizing three patented nano-hydroxyapatite (HAp) ceramic varieties. It provides significant insights into processcomposites incorporating those synthesized HAp ceramic varieties withdifferent polymers [methyl methacrylate (MMA), 2-hydroxyethyl methacrylate (HEMA)], polyester, epoxy, glass fibers, silicon carbide (SiC) and boron nitride (BN) which can be used for several biomedical, automotive and industrial applications. Synthesized nano-HAp ceramic varieties and processed HAp incorporated composites were characterized using X-ray fluorescence spectroscopy (XRF), particle size analysis (PSA), X-ray diffraction spectroscopy (XRD), fourier-transform infrared spectroscopy (FTIR), scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM with

EDS), differential scanning calorimetry (DSC), thermogravimetric analysis (TGA), dynamic mechanical analysis (DMA), tensile tests, and impact tests in order to find out composition, the size distribution of particles, crystallographic structure, identification of functional groups, the morphology of micro/nanostructures, glass transition temperatures, thermal stability, thermomechanical properties, tensile properties, and impact strength. American society for testing and materials (ASTM) standards were applied for all mechanical analyses. This dissertation compared those results with human hard tissues, commercial products, and literature according to their nature of applications. It has been concluded that HAp can be synthesized using the three routes to add value, HERP. These methods are highly cost-effective. Synthesized hexagonal, nano-HAp products perform similarly in chemical composition and structure to mammalian hard tissues via exhibiting different characteristics as bioceramic. The resulting composite materials obtained after reinforcing hydroxyapatite ceramic products with MMA and HEMA orderly for orthopedics and dentistry applications have proven that their performances are better than commercial products. It shows the highly profitable possibility of introducing Eppawala HAp products into the market as value-added bioceramic and its composites. Processed HAp incorporated E-glass fiber reinforced polyester matrix composites and ceramic (HAp/SiC/BN) incorporated epoxy matrix composites in this study demonstrate better properties than pure polyester and epoxy materials. Resulted composites can be used for automotive and aircraft applications. Also, it has confirmed the possibility of processing SiC and BN incorporated HAp composites under high temperatures (1600°C-1650°C), for other relevant industrial applications.

Keywords: Eppawala high-grade rock phosphate; Hydroxyapatite; Hydroxyapatite composites; Biomedical applications; Automotive applications; Industrial applications