



UNIVERSITI PUTRA MALAYSIA

**DEVELOPMENT OF CAPACITIVE COMPOSITE MATERIALS USING
ALKALINE TITANATES AND KAOLINITE CLAY**

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**DEVELOPMENT OF CAPACITIVE COMPOSITE MATERIALS USING
ALKALINE TITANATES AND KAOLINITE CLAY**

By

ALEX SEE

**Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia,
in Fulfilment of the Requirements for the Degree of Master of Science**

August 2009



This thesis is dedicated to God and my family.



Abstract of thesis presented to the Senate of University Putra Malaysia in fulfilment of requirement for the degree of Master of Science In Material Science.

**DEVELOPMENT OF CAPACITIVE COMPOSITE MATERIALS USING
ALKALINE TITANATES AND KAOLINITE CLAY**

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August 2009

Chairman: Associate Professor Dr. Jumiah Hassan, PhD

Faculty: Faculty Science

This research was designed to form a better dielectric composite material using one stable state dielectric matrix material and a good dielectric material as filler powder. The hypothesis is that the encapsulation of the filler powder by the matrix material, thus providing a bridge between the net polarizations of the composites. There are some limitations to the hypothesis, namely in the reactions and the limited control of the encapsulation process.

Distinct dielectric composites were successfully produced using locally sourced kaolinite clay and chemically produced alkaline titanates for example, Barium Titanate (BT) and Strontium Titanate (ST). The samples were made using kaolinite as the base matrix and alkaline titanate added in varying ratios. The alkaline titanate were synthesized via solid-state reaction using a carbonate derivative of the alkaline

cation and rutile titanium (IV) oxide sintered at 1200°C to 1300°C. White kaolinite was used to fuse the alkaline titanate material in varying weight ratios. The powders were dry-mixed and made into pellets for calcination at 1000°C.

The XRD data of BT-kaolinite type composites revealed a chemical reaction between the matrix and the filler powder at various combinations. Thus, BT additives reacted with the mullite and silica components and produced celsian feldspar in the composite system. This suggests that the system polarization contains the sum of four interfacial polarization processes at different temperatures. ST-mullite composites showed distinct varying interfacial cladding and dipolar relaxation for all composites.

SEM micrographs revealed fused and flaky parts formed in the BT composite samples whereby the powders were melted together. ST composite samples only showed inter-grain formation of the filler and matrix material. EDX, however, showed that there was some contamination in the matrix powder with potassium and carbon elements in the system.

Dielectric properties for both BT and ST composites exhibited low dielectric constant values ranging from 10 to 20 in the frequency range 10 Hz to 1 MHz. Dielectric modelling showed that the BT composite samples displayed single relaxation processes for the sample series whereas the ST composite samples exhibited multiple thermally activated dielectric relaxations. The modelling of the

dielectric data was done using the Havriliak-Negami equation to show two distinct dielectric processes occurring within the same framework.

Impedance plots showed significant varied impedance based on the measured temperatures with both composite series exhibiting high resistance. DC conductivity measurements were carried out on the samples, yielding very high activation exothermic and endothermic activation of energy of reactions in the medium. Dielectric modulus plots, however showed varied dispersion due to deformations in dielectric stresses in the composite series. Microwave frequency measurements showed dielectric properties of the medium without the interfacial polarization.

**Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia
sebagai memenuhi keperluan untuk ijazah Master Sains dalam Sains Bahan**

**KAJIAN TENTANG BAHAN KOMPOSIT JENIS KAPASITOR
MENGUNAKAN TITANATE BERALKALI DAN KAOLINITE**

Oleh

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Ogos 2009

Pengerusi: Profesor Madya Dr. Jumiah Hassan, PhD

Faculti: Faculti Sains

Kajian ini telah direka untuk membentuk bahan komposit dielektrik yang lebih baik dengan menggunakan bahan matriks bersifat dielektrik stabil dan bahan dielektrik yang tinggi sebagai serbuk pengisi. Hipotesis adalah bahawa penglitupan serbuk pengisi oleh bahan matriks membentuk jambatan di antara jumlah pengutuban dalam komposit tersebut. Terdapat beberapa had dari kajian ini, antaranya dalam tindakbalas-tindakbalas dan kekurangan kawalan dalam proses pelitupan.

Komposit dielektrik tertentu telah berjaya dihasilkan daripada kaolinit tempatan and titanat beralkali yang dibuat secara kimia, iaitu, barium titanat dan strontium titanat. Sampel telah dibentuk menggunakan kaolinite sebagai matriks asas and titanat beralkali dicampur dalam nisbah tertentu. Titanat beralkali telah disintesis menggunakan bahan karbonat berkation alkali dan titanium (IV) oksida yang dipanaskan pada suhu 1200°C hingga 1300°C. Kaolinit tempatan yang putih telah

digunakan untuk menggabungkan bahan titanat beralkali yang nisbah berat tertentu. Bahan serbuk telah dicampur secara kering dan dibentukkan kepada cakera kecil untuk dipanaskan pada suhu 1000°C.

Data XRD bagi komposit jenis BT-kaolinit telah menunjukkan tindakbalas kimia antara bahan matriks dan bahan pencampur dalam beberapa sampel yang digabungkan. Ini menunjukkan bahawa bahan pencampur BT telah bertindakbalas dengan mullit dan seterusnya membentuk ‘celsian felspar’ dalam sistem komposit tersebut. Oleh yang demikian, pengutuban sistem ini mungkin 4 jenis pengutuban sifat antaramukaan pada suhu berlainan. Komposit ST-mullit menunjukkan sifat pelitupan antaramukaan yang berlainan dan santaian dielektrik jenis dwikutub untuk semua sampel kompositnya.

Mikrograf SEM menunjukkan bahagian lakur dan leper terbentuk dalam sampel komposit BT di mana serbuk tersebut telah tergabung. Manakala sampel komposit ST hanya menunjukkan perbentukan butiran antara bahan pencampur dan matriks. EDX telah menunjukkan bahawa terdapat sedikit pencemaran unsur kalium dan karbon dalam sistem bahan matriks.

Sifat dielektrik untuk kedua-dua komposit BT dan ST menunjukkan nilai pemalar dielektrik yang rendah berukuran 10 hingga 20 pada frekuensi 10 Hz hingga 1 MHz. Pemodelan dielektrik telah menunjukkan bahawa sampel komposit BT mempunyai hanya sejenis proses santaian dielektrik manakala sampel ST menunjukkan variasi

dalam pengenduran dielektrik berganda teraktif secara terma. Pemodelan data dielektrik telah dibuat berpadukan model Havriliak-Negami untuk memperlihatkan dua proses dielektrik yang ketara berlaku serentak dalam sistem yang sama.

Plot impedans menunjukkan nilai impedans berubah dengan ketara berlandaskan suhu yang diambil dengan kedua-dua kumpulan komposit tersebut menunjukkan sifat kerintangan yang tinggi. Kajian sampel dalam sifat konduktiviti arus terus menunjukkan sifat tenaga pengaktifan eksotermik dan endotermik yang tinggi dalam bahan tersebut. Plot modulus dielektrik telah menunjukkan bahawa terdapat sebaran data yang tidak konsisten disebabkan oleh kewujudan kecacatan disebabkan oleh tekanan dielektrik di dalam siri komposit tersebut. Kajian dalam frekuensi mikro-gelombang hanya menunjukkan sifat dielektrik bahan medium tanpa pengutuban antara-mukaan di antara butiran.

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I certify that a Thesis Examination Committee has met on **24th August 2009** to conduct the final examination of **Alex See** on his master thesis entitled **Development of Capacitive Composite Materials Using Alkaline Titanates and Kaolinite Clay** in accordance with Universities and University Colleges Act 1971 and the Constitution of Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Masters of Science in Material Science.

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DECLARATION

I declare that the thesis is my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously, and is not concurrently, submitted for any other degree at Universiti Putra Malaysia or at any other institution.

(Signature)

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