

**DIELECTRIC PROPERTIES OF TOUGHENED POLYBENZOXAZINE  
BASED COMPOSITES IN THE MICROWAVE FREQUENCY REGION**

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Prof. Hatsuo Ishida

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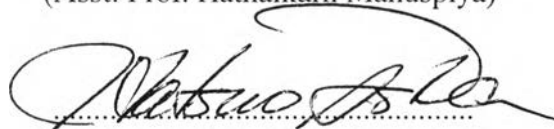


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## ABSTRACT

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Polybenzoxazine based/BST composites were proposed as new dielectric materials operated at microwave frequency. To alleviate the brittleness of usual monomeric benzoxazines, the toughness of polybenzoxazine was improved via two approaches. The first method is by alloying with flexible urethane and another strategy is by synthesizing high molecular weight benzoxazine precursors. It was found that for alloying system, at 90/10 vol.% of poly(benzoxazine/urethane), (PBA-a/PU) yielded good dielectric and thermal properties. For the synthesis route, those properties of methylenedianiline-based (BA-mdm) polybenzoxazine were superior to hexamethylenediamine (BA-hda). Thus at 90/10 vol.% of PBA-a/PU and poly(BA-mdm) were used as matrices for composite fabrication. The dielectric constant of all the polymer based composites was increased in relation to the amount of BST. The better filler distribution was obtained in PBA-a/PU than BA-mdm matrix when using 3-aminopropyl-trimethoxysilane and polymer based as surface modifiers on BST. The loss tangent and especially the dielectric constant of prepared composites had a weak dependence on frequency (300 MHz to 1 GHz) and temperature (-50 °C to 150 °C), indicating low relaxation behaviors when compared with general polymers. According to the study, at 90/10 vol.% of PBA-a/PU composed of silane coupling agent modified BST exhibited the prominent dielectric characteristics at various BST loadings. This type of composite, at 60 wt.% of BST content provided the highest dielectric constant (13.9) with low dissipation factor (0.0095), demonstrating high-performance as a candidate material for microwave frequency applications.

## บทคัดย่อ

สิรินภา วงศ์วิลาวัณย์ : การศึกษาสมบัติไดอิเล็กตริกของวัสดุคอมพอสิตพอลิเบนซอกซาซีนที่ถูกปรับปรุงสมบัติทางความเหนียวในคลื่นความถี่ไมโครเวฟ (Dielectric Properties of Toughened Polybenzoxazine Based Composite in the Microwave Frequency Region)  
 อ. ที่ปรึกษา : ผศ.ดร. หทัยกานต์ มนต์สปียะ และ ศ.ดร. ฮัทซีโอะ อิซิดะ 154 หน้า

วัสดุคอมพอสิตพอลิเบนซอกซาซีนและผงแบเรียมสตรอนเทียมไดดแทนด์ (BST) ได้ถูกนำเสนอขึ้นเพื่อเป็นวัสดุไดอิเล็กตริกชนิดใหม่ที่สามารถใช้งานได้ในพื้นที่ความถี่ไมโครเวฟ เพื่อลดความเปราะของพอลิเมอร์เบนซอกซาซีนทั่วไป ในการศึกษาครั้งนี้คุณสมบัติทางความเหนียวของพอลิเบนซอกซาซีนได้ถูกปรับปรุงผ่านสองวิธีการ โดยวิธีแรกคือการทำพอลิเมอร์อัลลอยกับยูรีเทนชนิดยืดหยุ่น ส่วนอีกวิธีหนึ่งเป็นการสังเคราะห์ด้วยสารตั้งต้นเบนซอกซาซีนมวลโมเลกุลสูง ผลการวิจัยในระบบของพอลิเมอร์อัลลอย พบว่าที่อัตราส่วนผสม 90/10 โดยปริมาตรของพอลิเบนซอกซาซีนชนิดเอ-อะนิลีน/ยูรีเทนให้คุณสมบัติทางไดอิเล็กตริกและสมบัติทางความร้อนที่ดี ในส่วนของการสังเคราะห์พบว่าพอลิเบนซอกซาซีนชนิดเมทิลีนไดอะนิลีน มีคุณสมบัติทางไดอิเล็กตริกและคุณสมบัติความร้อนที่เหนือกว่าชนิดเฮกซะเมทิลีนไดเอมีน ดังนั้นที่อัตราส่วน 90/10 โดยปริมาตรของสารผสมเบนซอกซาซีนชนิดเอ-อะนิลีน/ยูรีเทนและพอลิเบนซอกซาซีนชนิดเมทิลีนไดอะนิลีน ถูกนำมาใช้เป็นเมทริกซ์สำหรับการขึ้นรูปสารคอมพอสิต ค่าไดอิเล็กตริกของสารคอมพอสิตมีการเพิ่มขึ้นเป็นสัดส่วนกับปริมาณของผงแบเรียมสตรอนเทียมไดดแทนด์ โดยพบว่าสารตัวเติมมีการกระจายตัวที่ดีในเนื้อของพอลิเบนซอกซาซีนชนิดเอ-อะนิลีน/ยูรีเทนมากกว่าพอลิเบนซอกซาซีนชนิดเมทิลีนไดอะนิลีนเมื่อใช้สาร 3-อะมิโน โพรพิล-ไตรเมททอกซีไซเลน และพอลิเมอร์เมทริกซ์ในการปรับปรุงพื้นผิวของผงแบเรียมสตรอนเทียมไดดแทนด์ ค่าลอสเทนเจนต์และโดยเฉพาะอย่างยิ่งค่าไดอิเล็กตริกของวัสดุคอมพอสิตทั้งหมดที่เตรียมได้นี้ขึ้นต่อความถี่ในช่วง 300 เมกะเฮิร์ต ถึง 1 จิกะเฮิร์ตและอุณหภูมิในช่วง -50 ถึง 150 องศาเซลเซียสในระดับที่ต่ำแสดงถึงการพฤติกรรมที่คงที่เมื่อเปรียบเทียบกับพอลิเมอร์ทั่วไป และจากการศึกษาที่อัตราส่วนผสม 90/10 โดยปริมาตรของพอลิเบนซอกซาซีนชนิดเอ-อะนิลีน/ยูรีเทนประกอบด้วยผงแบเรียมสตรอนเทียมไดดแทนด์ที่ถูกปรับปรุงพื้นผิวโดยสารคู่ควบไซเลนให้คุณสมบัติทางไดอิเล็กตริกที่โดดเด่นในทุกปริมาณการเติมผงแบเรียมสตรอนเทียมไดดแทนด์ การเติมผงแบเรียมสตรอนเทียมไดดแทนด์ที่ 60% โดยน้ำหนักให้ค่าไดอิเล็กตริกที่สูงสุด (13.9) และมีค่าการสูญเสียในระดับต่ำ (0.0095) แสดงถึงการเป็นวัสดุที่มีประสิทธิภาพสูงในการใช้งานในคลื่นความถี่ไมโครเวฟ

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**ABBREVIATIONS**

Ba	2,2-bis(3,4-dihydro-3-phenyl- 2H-1,3-benzoxazine) propane
BA-a	Aniline-based benzoxazine monomer
BA-hda	Hexamethylenediamine-based benzoxazine precursor
BA-mda	Methylenedianiline-based benzoxazine precursor
BaTiO <sub>3</sub>	Barium titanate
BPU <sub>s</sub>	P-nitrophenol blocked polyurethanes
BST	Barium strontium titanate
eda	ethylenediamine
EPO732	Epoxy
hda	hexamethylenediamine
IPDI	Isophorone diisocyanate
J-S model	Jayasundere-Smith model
MCBP	Main-chain type benzoxazine polymers
mda	methylenedianiline
MDI	Diphenylmethane diisocyanate
NCO	Diisocyanate group
PBZ	Polybenzoxazine
PMC	Polymer-matrix composites
PT	Poly(tetramethyleneether) glycol
PU	Urethane prepolymer
TDI	Toluene diisocyanate

## SYMBOLS

A	Area
a	Weight of the sample in air
b	Weight of the sample immersed in deionized water
c	Weight of the damp sample after being wiped off excess water
C	Capacitance
D	Electric displacement
$D$	Distance between the plate
$D_{\text{water}}$	Density of deionized water
E	Electric intensity applied
$E_0$	Amplitude
$\tan \delta$	Loss tangent
Q	Charge
$Q$	Quality factor
$V$	Potential difference
$\alpha$	Total polarizability
$\alpha_e$	Electronic polarization
$\alpha_a$	Atomic polarization
$\alpha_o$	Dipole orientation polarization
$\epsilon_0$	Permittivity of free space ( $8.854 \times 10^{-12} \text{ C}^2/\text{m}^2$ or F/m)
$\epsilon'$	Dielectric constant
$\epsilon''$	Dielectric loss
$\epsilon$	Dielectric constant of the composites
$\epsilon_p$	Dielectric constants of the polymer matrix
$\epsilon_c$	Dielectric constants of the BST ceramic
$n$	Refractive index
$\emptyset$	Volume fraction
$\phi_c$	Volume fraction of the ceramic
$\phi_p$	Volume fraction of the polymer
$\rho_i$	Density of polyurethane

$\rho_2$	Density of polybenzoxazine
$\rho_c$	Density of ceramic filler
$\rho_p$	Density of polymer matrix
$\omega$	Angular frequency