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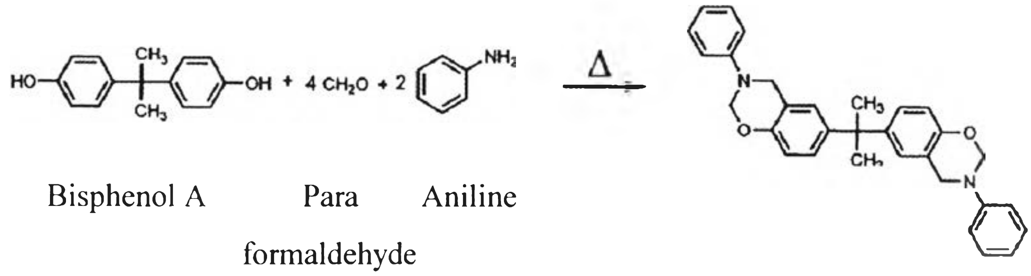
# APPENDICES

## Appendix A  Synthesis of Aniline Based Benzoxazine Monomer



Bisphenol A    Para formaldehyde    Aniline

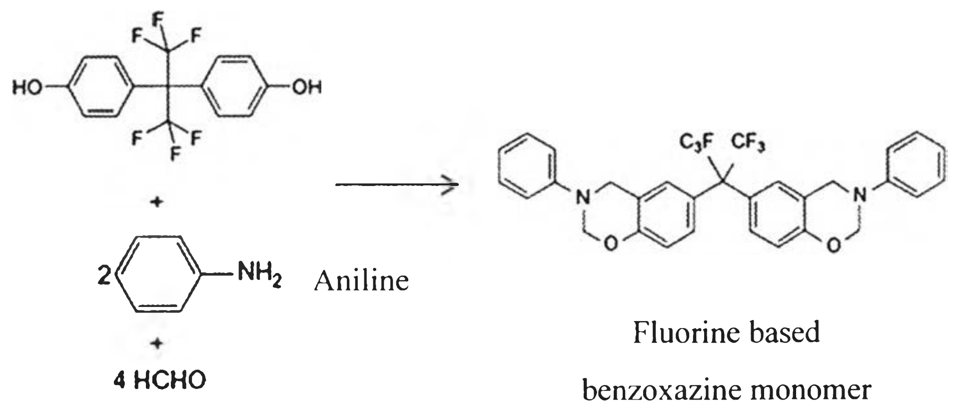
Aniline based benzoxazine monomer

**Table A1** Experimental data precursor for aniline based benzoxazine monomer

| | Precursors | | | Reaction time |
|---|---|---|---|---|
| | Bisphenol A | Paraformaldehyde | Aniline | |
| Mole | 0.06 | 0.24 | 0.12 | 30 min |
| Molecular weight | 228.29 | 30.03 | 93.13 | |
| Weight (g) | 13.70 | 7.21 | 11.18 | |

## Appendix B Synthesis of Fluorine Based Benzoxazine Monomer

Hexafluoro-bisphenal A




Aniline

Fluorine based benzoxazine monomer

Para formaldehyde

**Table B1** Experimental data precursor for fluorine based benzoxazine monomer

| | Precursors | | | Reaction time |
|---|---|---|---|---|
| | Hexafluoro-Bisphenol A | Paraformaldehyde | Aniline | |
| Mole | 0.06 | 0.24 | 0.12 | 30 min |
| Molecular weight | 336.24 | 30.03 | 93.13 | |
| Weight (g) | 20.17 | 7.21 | 11.18 | |

## Appendix C Preparation of Barium strontium titanate (BST) by Sol-Gel method

Precursor materials

1. Barium acetate ( $Ba(CH_3COO)_2$ ), d = 2.47 g/cm$^3$
2. Strontium acetate ( $Sr(CH_3COO)_2$ ), d = 2.099 g/cm$^3$
3. Titanium tetra-n-butoxide ( $Ti(CH^3(CH2)^3O^4)$ ), d = 0.998 g/cm$^3$
4. Glacial acetic acid
5. Methanol

**Table C1** Experimental data precursor for $Ba_{0.3}Sr_{0.7}TiO_3$

| | Precursors | | |
|---|---|---|---|
| | Barium acetate | Strontium acetate | Titanium tetra-n-butoxide |
| Mole | 0.00882 | 0.02058 | 0.0294 |
| Molecular weight | 225.42 | 205.71 | 340.36 |
| Weight (g) | 1.988 | 4.23 | - |
| Volume (ml) | - | - | 10 |

**Appendix D Calculation of BST Volume Fraction in PBA-a/BST Composite**

The volume fraction of BST was calculated by using the following formula:

$$f = \frac{(\frac{M_c}{\rho_c})}{(M_c/\rho_c) + (M_p/\rho_p)}$$

Where $M_c$ and $\rho_c$ are the mass and density (5.19 g/cm$^3$) of BST powder

$M_p$ and $\rho_p$ are the mass and density (1.27 g/cm$^3$) of aniline based benzoxazine monomer

**Table D1** Volume fraction of BST powder at various BST wt% in the composites

| PBA-a/BST Composites | BST volume fraction |
|---|---|
| 30 wt% BST | 0.00875 |
| 40 wt% BST | 0.1297 |
| 50 wt% BST | 0.1828 |
| 60 wt% BST | 0.2512 |
| 70 wt% BST | 0.3429 |
| 80 wt% BST | 0.4876 |

## Appendix E The Dielectric Constant and Loss Tangent of The Composites at Low Frequencies (1 kHz – 1 MHz)

**Table E1** The dielectric constant of polybenzoxazine based and BST ceramic

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^3$ | $10^4$ | $10^5$ | $10^6$ |
| PBA-a | 2.3500 | 2.3179 | 2.2797 | 2.2470 |
| PBA-f | 2.0930 | 2.0613 | 2.0148 | 1.9669 |
| $Ba_{0.3}Sr_{0.7}TiO_3$ | 395 | 347 | 296 | 283 |

**Table E2** The dielectric constant of aniline based polybenzoxazine/BST composites

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^3$ | $10^4$ | $10^5$ | $10^6$ |
| 30 wt% BST | 7.5152 | 7.3974 | 7.2438 | 7.0784 |
| 40 wt% BST | 11.0884 | 10.9002 | 10.6929 | 10.5025 |
| 50 wt% BST | 11.9500 | 11.7236 | 11.4968 | 11.2894 |
| 60 wt% BST | 19.1015 | 18.7507 | 18.3963 | 18.0717 |
| 70 wt% BST | 24.1590 | 21.8824 | 21.2127 | 20.8965 |
| 80 wt% BST | 26.3915 | 24.5220 | 23.7046 | 23.4501 |

**Table E3** The dielectric constant of silane treated aniline based polybenzoxazine/BST composites

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^3$ | $10^4$ | $10^5$ | $10^6$ |
| 30 wt% BST | 17.7594 | 17.5464 | 17.2813 | 16.9944 |
| 40 wt% BST | 19.4997 | 19.2057 | 18.8487 | 18.4804 |
| 50 wt% BST | 21.9234 | 21.5140 | 21.1212 | 20.7348 |
| 60 wt% BST | 23.9457 | 23.5727 | 23.1947 | 22.8259 |
| 70 wt% BST | 32.1474 | 31.5017 | 30.8887 | 30.2515 |
| 80 wt% BST | 34.0921 | 30.5723 | 28.1889 | 27.7551 |

**Table E4** The dielectric constant of phthalocyanine treated aniline based polybenzoxazine/BST composites

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^3$ | $10^4$ | $10^5$ | $10^6$ |
| 30 wt% BST | 17.3441 | 17.0093 | 16.6017 | 16.1661 |
| 40 wt% BST | 17.7149 | 17.4928 | 17.2010 | 16.8958 |
| 50 wt% BST | 20.0150 | 19.4587 | 19.0938 | 18.7891 |
| 60 wt% BST | 23.3220 | 22.3581 | 21.5584 | 20.8628 |
| 70 wt% BST | 29.0910 | 26.7362 | 25.0179 | 23.9113 |
| 80 wt% BST | 32.8792 | 28.1421 | 27.6642 | 24.1221 |

**Table E5** The dielectric constant of benzoxazine monomer treated aniline based polybenzoxazine/BST composites

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^3$ | $10^4$ | $10^5$ | $10^6$ |
| 30 wt% BST | 17.2564 | 17.0459 | 16.7662 | 16.4557 |
| 40 wt% BST | 17.2294 | 16.8718 | 16.5976 | 16.3091 |
| 50 wt% BST | 19.2935 | 18.8964 | 18.5097 | 18.0793 |
| 60 wt% BST | 20.5940 | 20.4031 | 20.2969 | 20.2733 |
| 70 wt% BST | 24.8521 | 23.4791 | 22.7909 | 22.3624 |
| 80 wt% BST | 33.1004 | 29.0102 | 27.8834 | 25.1635 |

**Table E6** The loss tangent of polybenzoxazine based and BST ceramic

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^3$ | $10^4$ | $10^5$ | $10^6$ |
| PBA-a | 8.3581e-3 | 0.0108 | 0.0107 | 9.0395e-3 |
| PBA-f | 0.0102 | 0.0132 | 0.0162 | 0.0143 |
| $Ba_{0.3}Sr_{0.7}TiO_3$ | 0.8816 | 0.8204 | 0.7926 | 0.7715 |

**Table E7** The loss tangent of aniline based polybenzoxazine/BST composites

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^3$ | $10^4$ | $10^5$ | $10^6$ |
| 30 wt% BST | 0.0110 | 0.0126 | 0.0116 | 0.0107 |
| 40 wt% BST | 0.0141 | 0.0127 | 0.0121 | 0.0116 |
| 50 wt% BST | 0.0180 | 0.0135 | 0.0132 | 0.0115 |
| 60 wt% BST | 0.0177 | 0.0134 | 0.0133 | 0.0125 |
| 70 wt% BST | 0.0965 | 0.0446 | 0.0162 | 0.0107 |
| 80 wt% BST | 0.0992 | 0.0772 | 0.0413 | 0.0248 |

**Table E8** The loss tangent of silane treated aniline based polybenzoxazine/BST composites

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^3$ | $10^4$ | $10^5$ | $10^6$ |
| 30 wt% BST | 0.0088 | 0.0093 | 0.0112 | 0.0121 |
| 40 wt% BST | 0.0118 | 0.0119 | 0.0145 | 0.0235 |
| 50 wt% BST | 0.0183 | 0.0130 | 0.0134 | 0.0162 |
| 60 wt% BST | 0.0163 | 0.0112 | 0.0112 | 0.0098 |
| 70 wt% BST | 0.0302 | 0.0152 | 0.0143 | 0.0140 |
| 80 wt% BST | 0.3512 | 0.0187 | 0.0174 | 0.0151 |

**Table E9** The loss tangent of phthalocyanine treated aniline based polybenzoxazine/BST composites

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^3$ | $10^4$ | $10^5$ | $10^6$ |
| 30 wt% BST | 0.0141 | 0.0152 | 0.0177 | 0.0177 |
| 40 wt% BST | 0.0099 | 0.0102 | 0.0121 | 0.0124 |
| 50 wt% BST | 0.0283 | 0.0163 | 0.0122 | 0.0110 |
| 60 wt% BST | 0.0410 | 0.0283 | 0.0241 | 0.0237 |
| 70 wt% BST | 0.0765 | 0.0543 | 0.0393 | 0.0330 |
| 80 wt% BST | 0.0802 | 0.0649 | 0.0481 | 0.0385 |

**Table E10** The loss tangent of benzoxazine monomer treated aniline based polybenzoxazine/BST composites

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^3$ | $10^4$ | $10^5$ | $10^6$ |
| 30 wt% BST | 0.0094 | 0.0100 | 0.0130 | 0.0207 |
| 40 wt% BST | 0.0088 | 0.0099 | 0.0117 | 0.0104 |
| 50 wt% BST | 0.0195 | 0.0142 | 0.0154 | 0.0151 |
| 60 wt% BST | 0.0112 | 0.0064 | 0.0044 | 0.0035 |
| 70 wt% BST | 0.0499 | 0.0302 | 0.0169 | 0.0111 |
| 80 wt% BST | 0.0514 | 0.0478 | 0.0217 | 0.0173 |

**Appendix F The Dielectric Constant and Loss Tangent of The Composites at High Frequencies (1 MHz – 1 GHz)**

**Table F1** The dielectric constant of polybenzoxazine based and BST ceramic

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^6$ | $10^7$ | $10^8$ | $10^9$ |
| PBA-a | 6.6621 | 6.7478 | 6.6395 | 6.1249 |
| PBA-f | 5.4766 | 5.3283 | 5.1629 | 4.4878 |
| $Ba_{0.3}Sr_{0.7}TiO_3$ | 219.63 | 90.11 | 78.30 | 46.10 |

**Table F2** The dielectric constant of aniline based polybenzoxazine/BST composites

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^6$ | $10^7$ | $10^8$ | $10^9$ |
| 30 wt% BST | 29.1190 | 30.2405 | 28.2851 | 14.8426 |
| 40 wt% BST | 30.9765 | 31.8539 | 30.1346 | 19.0570 |
| 50 wt% BST | 43.3231 | 49.3218 | 35.4077 | 21.1490 |
| 60 wt% BST | 56.9236 | 56.6486 | 51.9371 | 21.9388 |
| 70 wt% BST | 84.2451 | 87.9619 | 57.1181 | 20.7278 |
| 80 wt% BST | 90.1301 | 90.2207 | 76.2935 | 20.7672 |

**Table F3** The dielectric constant of silane treated aniline based polybenzoxazine/BST composites

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^6$ | $10^7$ | $10^8$ | $10^9$ |
| 30 wt% BST | 40.4242 | 40.3967 | 37.5946 | 19.1756 |
| 40 wt% BST | 54.2722 | 58.7691 | 40.1185 | 28.0693 |
| 50 wt% BST | 54.7593 | 60.8451 | 49.7788 | 35.7389 |
| 60 wt% BST | 64.7624 | 66.4320 | 62.3506 | 38.0252 |
| 70 wt% BST | 92.0381 | 98.0881 | 70.1652 | 34.2999 |
| 80 wt% BST | 95.1997 | 94.3594 | 81.3330 | 29.2645 |

**Table F4** The dielectric constant of phthalocyanine treated aniline based polybenzoxazine/BST composites

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^6$ | $10^7$ | $10^8$ | $10^9$ |
| 30 wt% BST | 33.2806 | 34.3710 | 30.9931 | 20.0548 |
| 40 wt% BST | 41.4009 | 52.4105 | 37.3312 | 25.1346 |
| 50 wt% BST | 51.5200 | 55.7152 | 43.5140 | 27.7392 |
| 60 wt% BST | 64.3073 | 62.2621 | 53.8462 | 33.8734 |
| 70 wt% BST | 85.5951 | 89.9069 | 64.4364 | 21.6805 |
| 80 wt% BST | 94.7847 | 90.9499 | 79.9012 | 25.7060 |

**Table F5** The dielectric constant of benzoxazine monomer treated aniline based polybenzoxazine/BST composites

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^6$ | $10^7$ | $10^8$ | $10^9$ |
| 30 wt% BST | 32.2062 | 32.0612 | 30.4764 | 18.5863 |
| 40 wt% BST | 40.8296 | 46.7985 | 35.7307 | 25.5159 |
| 50 wt% BST | 49.3196 | 50.2799 | 37.3045 | 21.4295 |
| 60 wt% BST | 61.7538 | 61.3733 | 59.0900 | 27.4911 |
| 70 wt% BST | 87.4405 | 88.8731 | 67.5809 | 24.3993 |
| 80 wt% BST | 92.8239 | 90.9931 | 77.4614 | 24.8860 |

**Table F6** The loss tangent of polybenzoxazine based and BST ceramic

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^6$ | $10^7$ | $10^8$ | $10^9$ |
| PBA-a | 7.5902e-4 | 9.9373e-5 | 2.5219e-4 | 6.5495e-4 |
| PBA-f | 1.6886e-4 | 1.4119e-4 | 2.9306e-4 | 4.9635e-4 |
| $Ba_{0.3}Sr_{0.7}TiO_3$ | 0.6703 | 0.4089 | 0.2381 | 0.0356 |

**Table F7** The loss tangent of aniline based polybenzoxazine/BST composites

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^6$ | $10^7$ | $10^8$ | $10^9$ |
| 30 wt% BST | 1.1848e-3 | 1.1673e-3 | 1.1738e-3 | 1.3879e-3 |
| 40 wt% BST | 1.3300e-3 | 1.1710e-3 | 1.2179e-3 | 1.5795e-3 |
| 50 wt% BST | 1.3988e-3 | 1.1678e-3 | 1.4601e-3 | 2.3925e-3 |
| 60 wt% BST | 1.4476e-3 | 1.4422e-3 | 1.5057e-3 | 3.2545e-3 |
| 70 wt% BST | 1.6072e-3 | 1.4950e-3 | 2.1919e-3 | 5.4034e-3 |
| 80 wt% BST | 1.9505e-3 | 1.8455e-3 | 2.1299e-3 | 5.4124e-3 |

**Table F8** The loss tangent of silane treated aniline based polybenzoxazine/BST composites

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^6$ | $10^7$ | $10^8$ | $10^9$ |
| 30 wt% BST | 9.9460e-4 | 9.7345e-4 | 9.2420e-4 | 1.1714e-3 |
| 40 wt% BST | 9.4527e-4 | 8.3658e-4 | 9.7800e-4 | 1.0578e-3 |
| 50 wt% BST | 1.1543e-3 | 9.6000e-4 | 1.2559e-3 | 2.2891e-3 |
| 60 wt% BST | 9.1416e-4 | 9.1993e-4 | 9.2311e-4 | 2.4068e-3 |
| 70 wt% BST | 1.1446e-3 | 1.1741e-3 | 2.0072e-3 | 2.6225e-3 |
| 80 wt% BST | 1.8072e-3 | 1.7791e-3 | 1.9548e-3 | 4.8057e-3 |

**Table F9** The loss tangent of phthalocyanine treated aniline based polybenzoxazine/BST composites

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^6$ | $10^7$ | $10^8$ | $10^9$ |
| 30 wt% BST | 6.6219e-4 | 5.8383e-4 | 5.4150e-4 | 7.1367e-4 |
| 40 wt% BST | 5.0907e-4 | 6.1514e-4 | 6.8300e-4 | 7.8353e-4 |
| 50 wt% BST | 1.0431e-3 | 8.9519e-4 | 9.1294e-4 | 1.9481e-3 |
| 60 wt% BST | 8.2022e-4 | 8.2896e-4 | 8.4427e-4 | 2.0447e-3 |
| 70 wt% BST | 9.8720e-4 | 1.1482e-3 | 1.8408e-3 | 2.4677e-3 |
| 80 wt% BST | 1.6410e-3 | 1.6402e-3 | 1.6819e-3 | 4.6334e-3 |

**Table F10** The loss tangent of benzoxazine monomer treated aniline based polybenzoxazine/BST composites

| Materials | Frequency (Hz) | | | |
|---|---|---|---|---|
| | $10^6$ | $10^7$ | $10^8$ | $10^9$ |
| 30 wt% BST | 3.2620e-4 | 3.4328e-4 | 3.5427e-4 | 5.2529e-4 |
| 40 wt% BST | 7.8583e-4 | 6.4815e-4 | 7.1953e-4 | 9.7344e-4 |
| 50 wt% BST | 8.1459e-4 | 7.3481e-4 | 8.4531e-4 | 1.7604e-3 |
| 60 wt% BST | 9.0708e-4 | 9.2405e-4 | 9.2452e-4 | 2.1464e-3 |
| 70 wt% BST | 9.5510e-4 | 1.0748e-3 | 1.9402e-3 | 2.4678e-3 |
| 80 wt% BST | 1.6428e-3 | 1.5522e-3 | 1.5701e-3 | 4.5879e-3 |

# CURRICULUM VITAE

**Name:** Ms. Wasinee Sapmaneenukul

**Date of Birth:** January 27, 1988

**Nationality:** Thai

**University Education:**

2006–2009 Bachelor Degree of Science, Faculty of Science, Chulalongkorn University, Bangkok, Thailand

**Proceedings:**

1. Sapmaneenukul, W.; Ishida, H.; and Manuspiya, H. (2012, January 11-13) Microwave dielectric properties of polybenzoxazine based composite for microwave substrate application. Proceedings of the Pure and Applied Chemical Conference (PACCON) 2012, Chiang Mai, Thailand.
2. Sapmaneenukul, W.; Ishida, H.; and Manuspiya, H. (2012, April 24) Significant improvement of dielectric properties in polybenzoxazine/BST composites. Proceedings of the 3rd Research Symposium on Petrochemical and Materials Technology and The 18th PPC Symposium on Petroleum, Petrochemicals and Polymers, Bangkok, Thailand.

**Presentations:**

1. Sapmaneenukul, W.; Ishida, H.; and Manuspiya, H. (2012, January 11-13) Microwave dielectric constant of polybenzoxazine based composite for microwave substrate application. Poster presented at the Pure and Applied Chemical Conference (PACCON) 2012, Chiang Mai, Thailand.
2. Sapmaneenukul, W.; Ishida, H.; and Manuspiya, H. (2012, April 24) Significant improvement of dielectric properties in polybenzoxazine/BST composites. Poster presented at the 3rd Research Symposium on Petrochemical and Materials Technology and The 18th PPC Symposium on Petroleum, Petrochemicals and Polymers, Bangkok, Thailand.