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### ACCEPTED MANUSCRIPT

# Ultra-low temperature sintering and microwave dielectric properties of a novel temperature stable $Na_2Mo_2O_7$ - $Na_{0.5}Bi_{0.5}MoO_4$ ceramic

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#### **Abstract**

The novel ultra-low temperature sintering  $(1-x)Na_2Mo_2O_7$ - $xNa_{0.5}Bi_{0.5}MoO_4$  ceramics have been obtained via solid-state reaction method for passive integration use. The  $Na_2Mo_2O_7$  and  $Na_{0.5}Bi_{0.5}MoO_4$  crystal phases are found to be compatible with each other from the results of XRD and SEM-EDS. With the x value changing from 0.36 to 1.00, the  $\varepsilon_r$  increases from 16.0 to 32.0 and the  $\tau_f$  value varies from -58 to 47 ppm/°C. At x=0.75, the 0.25 $Na_2Mo_2O_7$ -0.75 $Na_{0.5}Bi_{0.5}MoO_4$  ceramic sintered at an ultra-low sintering temperature of 580 °C can be densified (>96%) and possesses good microwave dielectric properties of an  $\varepsilon_r$  of 24.0, a Q×f value of 13,000 GHz (at 6.2 GHz), and a  $\tau_f$  value of 3 ppm/°C. The theoretical  $\varepsilon_r$  and  $\tau_f$  of the  $(1-x)Na_2Mo_2O_7$ - $xNa_{0.5}Bi_{0.5}MoO_4$  composites were calculated using the mixing law and in accordance with the measured values.

*Keywords*: Ceramics; Dielectrics; Microwave dielectric properties; Temperature stable material; Ultra-low temperature sintering.

#### 1. Introduction

In recent years, with the development of wearable wireless communication, it requires that microwave dielectric ceramics have a suitable permittivity, low loss, a near zero temperature coefficient of the resonant frequency ( $\tau_f$ ) and an ultra-low sintering temperature to co-fire with semiconductor, polymer and electrode materials, such as the Al paste (550 °C) or the nano particle silver ink [1-4]. However, most of the ultra-low firing microwave ceramics have the negative temperature coefficient of the resonant frequency. Many kinds of ultra-low temperature co-fired ceramic (ULTCC) materials (about 100) have been reported in the literature [4]. However, among the materials with sintering temperatures below 600 °C, there are only two ceramics with temperature stability, namely BaTe<sub>4</sub>O<sub>9</sub>-40wt%TiTe<sub>3</sub>O<sub>8</sub> ( $\varepsilon_r$  = 25.0, Q×f = 19,300 GHz,  $\tau_f$  = -3 ppm/°C) and 0.47BaTe<sub>4</sub>O<sub>9</sub>-0.53TiTe<sub>3</sub>O<sub>8</sub> ( $\varepsilon_r$  = 28, Q×f = 12,000 GHz,  $\tau_f$  = 4 ppm/°C) [5,6]. However, tellurium oxide is highly toxic. Therefore, the temperature-stable ultra-low firing ceramics especially sintered below 600 °C still need to be studied.

The previous study demonstrates that the ceramics in  $A_2O\text{-}MoO_3$  (A = Na, K, Ag) systems have ultra-low sintering temperatures of 460-660 °C and good microwave dielectric properties ( $\varepsilon_r$  = 4.1-14.0, Q×f = 8,500-62,400 GHz and  $\tau_f$  = -142~-57 ppm/°C) [7,8]. Among them, the Na<sub>2</sub>Mo<sub>2</sub>O<sub>7</sub> ceramic sintered at 575 °C has the best Q×f value of 62,400 GHz, an  $\varepsilon_r$  of 12.9 and a  $\tau_f$  of -72 ppm/°C [8]. Among the microwave dielectric materials with ultra-low sintering temperatures and positive  $\tau_f$  values, the Na<sub>0.5</sub>Bi<sub>0.5</sub>MoO<sub>4</sub> ceramic sintered at 690 °C has a high Q×f

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