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# A new additive-free microwave dielectric ceramic system for LTCC applications: $(1-x)CaWO_4-x(Li_{0.5}Sm_{0.5})WO_4$

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**Abstract**
In this work, a novel additive-free microwave dielectric ceramic (MWDC) system of $(1-x)CaWO_4-x(Li_{0.5}Sm_{0.5})WO_4$ (CW-LSW, $0.12 \le x \le 0.20$) for low temperature co-fired ceramic (LTCC) applications was successfully fabricated by a solid-state reaction route. The sintering temperature and composition effects on the microwave dielectric (MWD) properties, microstructures, and crystalline phases were investigated. The optimal sintering temperatures of the CW-LSW ceramics are ranging from 875 to 950 °C. When sintered at 900 °C for 3 h, the sample of 0.86CW-0.14LSW exhibited excellent MWD properties: $\varepsilon_r = 10.76$, $Q \times f = 28{,}754$ GHz, and $\tau_f = -0.54$ ppm/°C. Furthermore, it is found that the CW-LSW ceramics are well compatible with Ag electrode during the sintering process. Therefore, the findings suggest that CW-LSW ceramic materials are promising for LTCC applications.

## 1 Introduction

Low temperature co-fired ceramic (LTCC) materials have aroused increasing international interest because of their promising applications in electronic industry [1]. As known to all, the LTCC materials are widely applied to many microwave devices, including antennas, filters, oscillators, dielectric waveguides, and substrates [2–4]. To meet the requirements of the applications mentioned above, it is necessary for the LTCC materials to have a combination of a near-zero temperature coefficient of resonant frequency ($\tau_f$), a high quality factor ($Q \times f$), and a low dielectric constant ($\varepsilon_r$) [5, 6]. Moreover, in order to be co-fired with electrode metals (e.g., Au, Ag, and Cu), the sintering temperatures of the LTCC materials have to be lower than the melting points of metal electrodes (e.g., < 950 °C) [7, 8]. Therefore, effective sintering additives, such as $H_3BO_3$, $Li_2CO_3$, $BaCu(B_2O_5)$, and low-melting glass were usually employed to decrease the sintering temperature. However, as a result of the formation of secondary phases, glassy phases, and structural defects caused by the use of the sintering additives, the $Q \times f$ value will be inevitably decreased, which significantly hinders the applications of the LTCC materials [2–4, 9, 10]. Consequently, it is highly necessary to study the fabrication of additive-free LTCC materials with high $Q \times f$ values.

However, it is quite challenging to develop additive-free LTCC materials, as relatively few ceramic materials can be fully sintered at temperatures lower than 950 °C without any additive [5, 11–18]. Recently, tungstate ceramic materials (e.g., $CaWO_4$, $SrWO_4$, and $BaWO_4$) were found to be excellent candidates for LTCC applications and therefore be intensively studied [19]. Among these tungstate ceramic materials, $CaWO_4$ with a scheelite structure presented outstanding microwave dielectric (MWD) properties (i.e., $\varepsilon_r = 10$, $Q \times f = 75{,}000$ GHz, and $\tau_f = -25$ ppm/°C) [19, 20]. Although the sintering temperature of $CaWO_4$ is at about 1100 °C, it is easy to be effectively decreased by combining $CaWO_4$ with some other tungstate materials, which have lower sintering temperatures. As reported by Zhang et al., a high $Q \times f$ value (i.e., 117,600 GHz), a low dielectric constant (i.e., 9), and a relatively large negative $\tau_f$ (i.e., − 55 ppm/°C) were achieved in the binary ceramic system of $CaWO_4$-$Li_2WO_4$, when sintered at 900 °C [12]. Furthermore, as shown by Bian et al. with an addition of $(Li_{0.5}Nd_{0.5})WO_4$, low sintering temperatures

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