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Citation: Li, Chunchun, Yin, Changzhi, Deng, Ming, Shu, Longlong and Khaliq, Jibran (2020) Tunable microwave dielectric properties in SrO-V2O5 system through compositional modulation. Journal of the American Ceramic Society, 103 (4). pp. 2315-2321. ISSN 0002-7820

Published by: Wiley-Blackwell

URL: https://doi.org/10.1111/jace.16955 < https://doi.org/10.1111/jace.16955 >

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# Tunable microwave dielectric properties in SrO-V2O5 system through compositional modulation

Journal:	Journal of the American Ceramic Society				
Manuscript ID	JACERS-44698.R2				
Manuscript Type:	Rapid Communication				
Date Submitted by the Author:	n/a				
Complete List of Authors:	Li, Chunchun; Guilin University of Technology, College of Information Science and Engineering; Guilin University of Technology, Key laboratory of New Processing Technology for Nonferrous Metal and MaterialsMinistry of Education Yin, Changzhi; State Key Laboratory Breeding Base of Nonferrous metals and specific Materials Processing, Guangxi universities key laboratory of non-ferrous metal oxide electronic functional materials and devices, College of Material Science and Engineering, Guilin University of Technology Deng, Ming; Guilin University of Technology, College of Information Science and Engineering Shu, Longlong; Nanchang University, School of Materials Science and Engineering Khaliq, Jibran; University of Northumbria at Newcastle				
Keywords:	ceramic matrix composites, dielectric materials/properties, LTCC, microwaves				
Author-supplied Keyword: If there is one additional keyword you would like to include that was not on the list, please add it below::					



# Tunable microwave dielectric properties in SrO-V<sub>2</sub>O<sub>5</sub> system

# through compositional modulation

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#### Abstract

Adjustment on resonance frequency stability against the sintering temperature of  $Sr_3V_2O_8$  was realized through adjusting the Sr:V mole ratio. Effects of Sr:V ratio on sintering behavior and dielectric properties of  $Sr_3V_2O_8$  were studied. The sintering temperature was successfully reduced to 950 °C from 1150 °C. With increasing vanadium content, both relative permittivity and quality factor decreased, while the temperature coefficient of resonance frequency shifted from positive to negative values. Especially, a near-zero  $\tau_f$  of -1.1 ppm/°C along with a low permittivity ( $\epsilon_r$ ) of 9.8 and a quality factor  $Q \times f$  of 24,120 GHz was successfully achieved in  $Sr_{3-y}V_2O_{8-y}$  ceramic (y = 0.6, sintered at 950 °C). The wide compositional and processing adjustment window, favorable dielectric performances, and good chemical compatibility with silver render  $Sr_{3-y}V_2O_{8-y}$  ceramics potential candidates in multilayer electronic devices.

Keywords: Ceramics; Dielectric properties; Microwave resonance; Composite

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ceramics

#### 1. Introduction

The developments of commercial wireless technologies, especially the fifthgeneration (5G) telecommunication, Internet of Things (IoTs) and military radar systems, have expanded the operating frequency to the millimeter-wave range. This new technological paradigm brings out the increasing demands of high-speed signal propagation at high-frequency regions [1-4]. For ceramics used as substrates, low permittivity ( $\varepsilon_r < 15$ ) is required for fast signal transmission and minimizing the crosscoupling between the substrates and the conductors [5-7]. Other properties such as high quality factors ( $Q \times f$ ) and near-zero temperature coefficient of resonant frequency ( $\tau_{7}$ ) are also essential for practical applications [8-10]. To date, a large number of dielectric materials have been reported, however, only a few numbers of available options could meet the combination requirements simultaneously. Thus, developing new materials with desired microwave dielectric properties is still a challenge.

To reduce the permittivity, two possible approaches have been proposed in the literatures [11, 12], one of which is to decrease the number of dipoles while the second is to lower the dipole strength. The former method involves lowering the density through introduction of porosity. This however sacrifices mechanical strength and thermal conductivity while increasing dielectric loss. Lowering the dipole strength is more desirable through introducing covalent bond. To date, a number of promising low-permittivity materials ( $\varepsilon_r < 15$ ) have been reported. All of those materials have

tetrahedral unit cell, such as borates, silicates, phosphates, and vanadates [3, 12-14]. Amongest them, vanadates have attracted considerable attention due to cheap raw materials, simple synthetic process, and good microwave dielectric properites [15-18]. For example, alkaline earth orthovanadates,  $M_3(VO_4)_2$  (M = Mg, Ba, Sr) are promising candidates with low dielectric loss and low- $\varepsilon_r$  for high-frequency application [17-19]. Mg<sub>3</sub>(VO<sub>4</sub>)<sub>2</sub> sintered at 950 °C possesses  $\varepsilon_r = 9.3$ ,  $Q \times f = 65,540$  GHz and  $\tau_f = -89.5$ ppm/°C [19], and Ba<sub>3</sub>(VO<sub>4</sub>)<sub>2</sub> exhibited good dielectric performances with  $\varepsilon_r \sim 11.3$ ,  $Q \times f$ ~ 62,347 GHz and  $\tau_f \sim 28.8$  ppm/°C when sintered at 1400 °C [20]. In our previous work,  $Ba_{3-x}Sr_x(VO_4)_2$  solid solution series were reported to have promising microwave dielectric properties with  $\varepsilon_r = 11-16$ ,  $Q \times f = 40,000-66,000$  GHz, and  $\tau_f = 20-70$  ppm/°C [21]. Particularly, Sr<sub>3</sub>(VO<sub>4</sub>)<sub>2</sub> has the lowest densification temperature (~1150 °C) along with a combination of promising dielectric performances with a high quality factor of 44,340 GHz and a low dielectric permittivity of 12.2. The high sintering temperature (>1000 °C) and relatively large positive  $\tau_f$  value (~+63.5 ppm/°C), however, still limits its potential utilization in low temperature cofired ceramics (LTCC) application in which the ceramic layers should be cofired with the inner electrodes (generally silver) [22]. Thus, reduction in sintering temperature to below the melting temperature of silver (961 °C) and tailoring  $\tau_f$  to near-zero is necessary for M<sub>3</sub>(VO<sub>4</sub>)<sub>2</sub> [23-25].

According to the binary phase diagram of SrO-V<sub>2</sub>O<sub>5</sub>, there are two stable phases  $Sr_2V_2O_7$  with triclinic structure and  $Sr_3(VO_4)_2$  and both of the phases can coexist [26]. Importantly, one of the advantages of  $Sr_2V_2O_7$  is to have a negative  $\tau_f$  value ~ -34.8 ppm/°C which can behave as a  $\tau_f$  compensator for  $Sr_3(VO_4)_2$ . Both phases can coexist by adjusting Sr:V mole ratio in the binary SrO-V<sub>2</sub>O<sub>5</sub> system to complement each other [27]. Based on this rationale, this paper proposed the formation of the second phase to compensate  $\tau_f$  value in-situ by compositional modification, a reliable and simple method, which was validated and verified in Sr<sub>3-y</sub>V<sub>2</sub>O<sub>8-y</sub> ( $0 \le y \le 1$ ) ceramics system. A series of different Sr:V ratio compounds in the Sr<sub>3-y</sub>V<sub>2</sub>O<sub>8-y</sub> (y = 0.2-0.8) system were prepared and characterized for the relationships between the phase composition and microwave dielectric properties.

#### 2. Experimental Procedure

#### 2.1 Sample preparation

 $Sr_{3-y}V_2O_{8-y}$  (y = 0.2, 0.4, 0.6, 0.8) ceramics were prepared via conventional solidstate method from individual reagent-grade oxide powders:  $SrCO_3$ , (> 99.95%, Guo-Yao Co., Ltd Shanghai, China) and NH<sub>4</sub>VO<sub>3</sub> (> 99.9%, Guo-Yao Co., Ltd Shanghai, China). The powders were weighted according to the stoichiometric composition of  $Sr_{3-y}V_2O_{8-y}$ , and ball milled in alcohol medium for 6 h in nylon battle with zirconia balls. After drying the slurry at 120 °C for 1 h, the obtained powders were calcined at 850 °C for 6 h in air. The calcined powders were ball-milled again for 6 h followed by coldpressing into cylinders (10 mm in diameter and 6 mm in thickness) in a steel die under a pressure of 200 MPa with polyvinyl alcohol (PVA, 10 vol.%) as s binder. The  $Sr_{3-y}V_2O_{8-y}$  pellets were sintering at in the range of 900-1150 °C for 6 h in the air.

#### 2.2 Characteristics

The crystal structure and phase(s) of the specimens were analyzed using X-ray diffraction (Cu $K\alpha I$ , 1.54059 Å, Model X'Pert PRO, PANalytical, Almelo, Holland).

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The diffraction patterns were taken at room temperature in the range of  $10^{\circ}-80^{\circ}$  by step scans. The bulk density was determined by the Archimedes' method and the theoretical density was obtained by the following equation:

$$\rho_{th} = \frac{\omega_1 + \omega_2}{\omega_1 / \rho_1 + \omega_2 / \rho_2} \tag{1}$$

where  $\omega_1$ ,  $\omega_2$  and  $\rho_1$ ,  $\rho_2$  are the mass fractions and theoretical density of Sr<sub>2</sub>V<sub>2</sub>O<sub>7</sub> and Sr<sub>3</sub>(VO<sub>4</sub>)<sub>2</sub>, respectively.

The surface morphologies of the sintered samples were observed by scanning electron microscope (FE-SEM, Model S4800, Hitachi, Japan). The relative permittivity  $(\varepsilon_r)$  and quality factor  $(Q \times f)$  of the samples were measured using a network analyzer (Model N5230A, Agilent Co., Palo Alto, America). The temperature coefficient of resonant frequency  $(\tau_f)$  was measured by noting the temperature shift of the resonance scope in the temperature range of 25-85 °C using a temperature chamber (Delta 9039, Delta Design, San Diego, CA) and were calculated as follows:

$$\tau_f(\text{ppm/°C}) = \frac{f_2 - f_1}{f_1(T_2 - T_1)} \times 10^6$$
(2)

where,  $f_1$  and  $f_2$  represent resonant frequencies at temperatures  $T_1$  and  $T_2$ , respectively.

#### 3. Results and discussion

In order to get a clear understanding of the chemical reaction happening within  $Sr_{3-y}V_2O_{8-y}$  system, thermal analysis was carried out. Fig. 1(a) shows the TG/DSC thermograph of the mixed precursor of the y = 0.2 sample. Three endothermic peaks between 150 °C to 450 °C were observed in the DSC curve, accompanied by a total mass loss of nearly 8% on the TGA curve. These three peaks, located at 200 °C, 225 °C and 370 °C correspond to the gradual decomposition of NH<sub>4</sub>VO<sub>3</sub> into V<sub>2</sub>O<sub>5</sub> and NH<sub>3</sub>

with the first as the primary decomposition process. A broad exothermic peak around 527 °C can be related to the chemical reaction of  $V_2O_5$  with SrCO<sub>3</sub> to form Sr<sub>2</sub>V<sub>2</sub>O<sub>7</sub> and/or Sr<sub>3</sub>V<sub>2</sub>O<sub>8</sub> with mass loss of nearly 8% due to the release of carbon dioxide. The endothermic peak at 880 °C is associated with the complete decomposition of SrCO<sub>3</sub>. Fig. 1(b) shows the XRD pattern of the y = 0.2 sample sintered at 860 and 960 °C. A peak belonging to SrCO<sub>3</sub> is visible at 860 °C, which disappeared at 960 °C, hence confirming the thermal analysis.

Fig. 2(a) illustrates the XRD patterns of  $Sr_{3-\nu}V_2O_{8-\nu}$  (y = 0.2-0.8) ceramics sintered at 950 °C. Within the range of  $0.2 \le y \le 0.8$ , only two main crystalline phases,  $Sr_2V_2O_7$ with a space group P-1 (No. 2) and  $Sr_3(VO_4)_2$  were observed and the volume fraction of  $Sr_2V_2O_7$  increased with increasing y value, as listed in Table 1, which was verified based on the bi-phase Rietveld refinement. As representatives, Figs. 2(b, c) show the refined XRD patterns with y = 0.2 and y = 0.8. The reasonable reliability factors indicate that a mixed-phase of  $Sr_3(VO_4)_2$  and  $Sr_2V_2O_7$  were obtained in all samples. Additionally, Fig. 2(d) shows the change of relative density as a function of y value and phase fraction of Sr<sub>2</sub>V<sub>2</sub>O<sub>7</sub> calculated from refinement. All the sintered Sr<sub>3-v</sub>V<sub>2</sub>O<sub>8-v</sub> samples exhibited a relative density of over 95% and an upward trend with an increase in y value, indicating that the increment of V2O5 content facilitates densification of Sr3-yV2O8-y. The phase content of  $Sr_2V_2O_7$  increased from 20.47% to 81.31%, which is very close to the theoretical values (17.88% to 77.7%) obtained from the nominal formula  $Sr_{3-}$  $_{y}V_{2}O_{8-y}$ .

Fig. 3(a-d) present the SEM images of  $Sr_{3-\nu}V_2O_{8-\nu}$  (y = 0.2-0.8) sintered at their

optimum temperatures. Following the high relative density (over 95%, Table 1), all the samples show well-densified microstructures. It can be seen that two different shapes of grains (round and columnar grains) coexisted in the samples in the composition range studied. Fig. 3(e, f) presented the elemental content in different grains (spot 1 and 2) captured using EDS. The Sr/V ratio of the columnar phases is approximately 1.52, which is close to the composition of  $Sr_3V_2O_8$ , while Sr/V ratio for the round phase is around 1.01, corresponding to  $Sr_2V_2O_7$  phase and the columnar-like grains were  $Sr_3V_2O_8$  phase.

The microwave dielectric properties ( $\varepsilon_r$ ,  $Q \times f$ , and  $\tau_f$ ) of Sr<sub>3-y</sub>V<sub>2</sub>O<sub>8-y</sub> (y = 0.2-0.8) ceramics exhibited a downward trend with increasing y value as shown in Fig. 4. Particularly, the  $\tau_f$  value of the Sr<sub>3-y</sub>V<sub>2</sub>O<sub>8-y</sub> ceramics decreased from +48.7 ppm/°C to -20.1 ppm/°C. Sr<sub>3-y</sub>V<sub>2</sub>O<sub>8-y</sub> composite having y = 0.6 and sintered at 950 °C demonstrated a near-zero  $\tau_f$  of -1.1 ppm/°C, along with  $\varepsilon_r$  of 9.8 and quality factor  $Q \times f$  of 24,120 GHz. According to the empirical Lichtenecker mixing rule for a two-phase composite, the effective  $\varepsilon_r$ ,  $Q \times f$ , and  $\tau_f$  values can be theoretically estimated by the following equations [28]:

$$\varepsilon^n = V_1 \varepsilon_1^n + V_2 \varepsilon_2^n \quad (-1 \le n \le 1) \tag{3}$$

$$\tau_f = v_1 \tau_{f1} + v_2 \tau_{f2}$$
(4)  
$$\frac{1}{Q} = \frac{v_1}{Q_1} + \frac{v_2}{Q_2}$$
(5)

where  $\varepsilon_1$  and  $\varepsilon_2$  are the respective permittivities of Sr<sub>3</sub>(VO<sub>4</sub>)<sub>2</sub> and Sr<sub>2</sub>V<sub>2</sub>O<sub>7</sub> phase;  $\tau_{f1}$ and  $\tau_{f2}$  are the  $\tau_f$  values of the pure Sr<sub>3</sub>(VO<sub>4</sub>)<sub>2</sub> and Sr<sub>2</sub>V<sub>2</sub>O<sub>7</sub> phase,  $V_1$  and  $V_2$  ( $V_1 + V_2 =$ 1) are the volume fractions of the corresponding phases. And n = 1 or -1 correspond to the parallel and series mixing law, respectively. When n approaches 0, Eq. (3) becomes logarithmic, usually used for randomly distributed composites:

$$\ln \varepsilon = V_1 \ln \varepsilon_1 + V_2 \ln \varepsilon_2 \tag{6}$$

As shown in Fig. 4(a), the measured values of  $\varepsilon_r$  for Sr<sub>3-v</sub>V<sub>2</sub>O<sub>8-v</sub> (v = 0.0-1.0) composites are matched with values calculated using equations of parallel or series mixing law, and Eq. (6). This indicates that the measured values of  $\varepsilon_r$  follow the logarithmic mixing law with the respective volume fraction. The theoretical  $Q \times f$  and  $\tau_f$  values of Sr<sub>3-v</sub>V<sub>2</sub>O<sub>8-v</sub> (y = 0.2-0.8) are calculated using the Eq. (4) and (5) and shown in Figs. 4(b, c). The measured  $Q \times f$  and  $\tau_f$  values were in agreement with the theoretical values, confirming that the dielectric properties can be conveniently and precisely tailored by this method.

The low sintering temperature (950 °C) of sample with y = 0.6 enables its potential application in LTCC technology. To evaluate the chemical compatibility with silver, cofiring was carried out between the v = 0.6 and 20 wt.% Ag powders at 950 °C for 2 h. As shown, XRD pattern recorded on the cofired sample only exhibits peaks of silver (ICDD No. 87-0717),  $Sr_3V_2O_8$ , and  $Sr_2V_2O_7$ . In addition, backscattered electron image (BEI) shows distinguish grains with different element contrasts and the bright grains were verified as silver by EDS analysis. These results suggest that the present ceramics have a good chemical compatibility with Ag electrode, rendering their potential use in LTCC technology.

#### 4. Conclusions

To tailor the temperature coefficient of resonance frequency and lower the sintering temperature of Sr<sub>3</sub>V<sub>2</sub>O<sub>8</sub>, in-situ composite formation through modified Sr:V

 ratio was proposed and a series of Sr<sub>3-y</sub>V<sub>2</sub>O<sub>8-y</sub> were prepared. Successful reduction in sintering temperature was achieved from 1150 °C to 950 °C, rendering their future possible application in LTCC technology. Microwave dielectric properties can also be tuned by varying the Sr:V ratio. In particular, a composition with y = 0.6 sintered at 950 °C possessed a near-zero  $\tau_f$  of -1.1 ppm/°C, along with  $\varepsilon_r$  of 9.8 and quality factor  $Q \times f$ of 24,120 GHz. This work paves the way for Sr<sub>3</sub>(VO<sub>4</sub>)<sub>2</sub> ceramics to utilize in multilayer electronic devices. for per period

### Acknowledgments

This work was supported by the Natural Science Foundation of Guangxi Zhuang

Autonomous Region (No 2018GXNSFAA281253).

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**Table 1** Comparison of calculated and theoretical values, sintering temperature, andmicrowave dielectric properties of  $Sr_{3-y}V_2O_{8-y}$  ceramics

	The volume fraction of $Sr_2V_2O_7$		- Theoretical	Relative	Optimum	Dielectric properties		
у	Calculation from Fullprof (%)	Theoretical Values (%)	density (g/cm <sup>3</sup> )	density ( %)	sintering temperatur e (°C)	E <sub>r</sub>	Q×f (GHz)	τ <sub>f</sub> (ppm/°C)
0.2	20.47	17.88	4.39	96.36	1000	11.8	36,400	48.7
0.4	37.19	36.70	4.31	97.21	1000	10.6	34,960	20.6
0.6	57.48	56.51	4.24	98.11	950	9.8	27,550	-1.1
0.8	81.31	77.67	4.14	98.55	950	9.4	24,100	-20.1

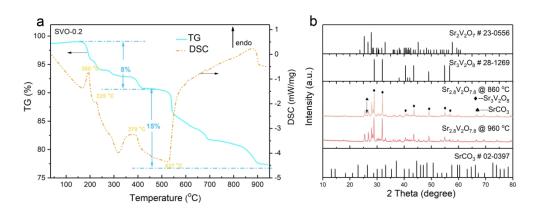


Figure 1 (a) TG/DSC analysis of the y = 0.2 sample, (b) XRD patterns of Sr3-yV2O8-y (y = 0.2) sintered at 860 oC and 960 oC.

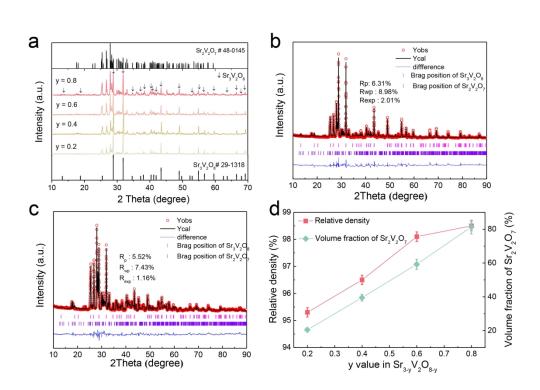
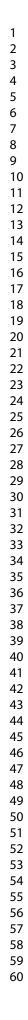


Figure 2 (a) XRD patterns of the Sr3-yV208-y (y = 0.2, 0.4, 0.6, and 0.8) sintered at optimum temperature, (b) Rietveld refinement profiles for y = 0.2, and (c) for y = 0.8 and (d) change of phase fraction of Sr2V207 calculated from refinement.



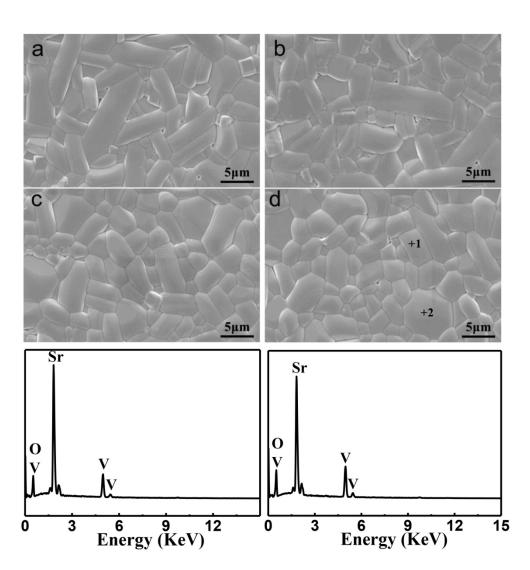


Figure 3 SEM images recorded on the polished and thermally etched surfaces of Sr3-yV2O8-y ceramics: (a) y = 0.2, (b) y = 0.4, (c) y = 0.6, (d) y = 0.8; (e) and (f) EDS for spot 1 and 2, respectively.

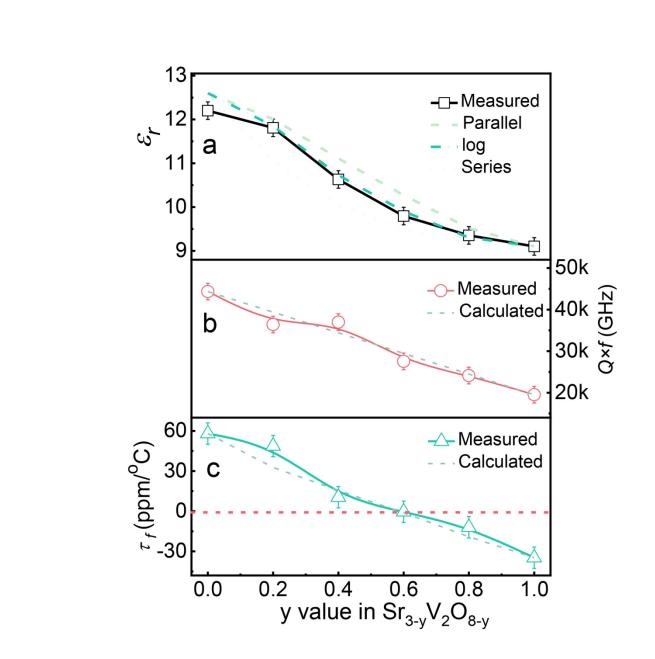


Fig. 4 Variations in microwave dielectric properties as a function of y in the Sr3-yV2O8-y ceramics; the corresponding calculated values are also given for comparison.

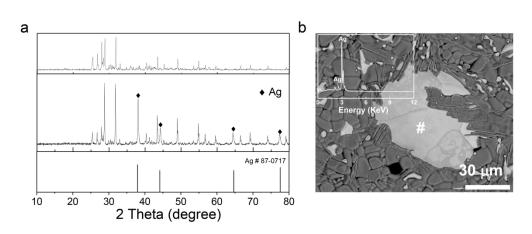


Figure 5 (a) XRD patterns and (b) SEM micrograph of Sr2.4V2O7.4 + 20 wt.% Ag powders at 950 °C for 2 h (EDS analysis of Ag is shown in the inset of Fig. 5(b)).