# The Effect of Applied Frequencies and Multiple Firing on the Resistance of Thick Film Resistors

Bi-Shiou Chiou, Wei-Yung Hsu, and Jenq-Gong Duh

Abstract — The resistance and impedance as a function of frequency for Dupont 1600 series thick film resistors are investigated. There exists an abrupt decrease in the resistance around 1 MHz. An equivalent circuit model is proposed to explain the high frequency behavior in the resistor. After multiple times of firing, the resistance is altered and the relative change is related to the intrinsic resistance in the resistor. It is argued that the resistance variation is associated with the microstructure evolution of the resistor, in which the rearrangement of conducting grains in the glass matrix plays a major role.

### I. INTRODUCTION

HICK film resistor is a very complicated, nonequilibrium system [1]. Most thick film resistor pastes consist of three major ingredients, including conductive particles, glass frits, and organic vehicle. After firing the resistor paste pattern, the organic vehicle is burned out and a microstructure with conductive particles sintered into continuous conductive chains in an insulating glass matrix is formed. The electrical conduction of the resistor is via these conductive chains. The more continuous conductive chains and/or the wider the chains, the lower the resistance. According to theoretical calculation, a conductive-to-frit volume ratio of 0.01 is sufficient to form continuous conductive chains. However, as the thick film resistor is a nonequilibrium system, rearrangement and/or ripening of the conductives as well as reactions between the conductive and glass matrix occur when resistors are sintered for a long period of time.

Thick film resistor, with its thermal stability, mass productivity, and cost effectiveness, is widely applied in various electronic products. As high frequency communication instrumentation is prevailing today, the high frequency behavior of the electronic device is becoming an important aspect to probe. The purpose of this paper is to investigate the effect of the firing condition on the high frequency characteristics of commercial thick film RuO<sub>2</sub>-based resistors. An equivalent circuit model is proposed to explain the electrical behavior of the resistors.

Manuscript received April 1, 1991; revised December 12, 1991.

B.-S. Chiou is with the Department of Electronic Engineering and the Institute of Electronics, National Chiao Tung University, Hsinchu, 300 Taiwan, Republic of China.

W-Y Hsu was with the Department of Materials Science and Engineering, National Tsing Hua University, Hsinchu, Taiwan, Republic of China. He is now with the Department of Materials Science and Engineering, Cornell University, Ithaca, NY 14853.

J.-G Duh is with the Department of Materials Science and Engineering, National Tsing Hua University, Hsinchu, 300 Taiwan, Republic of China. IEEE Log Number 9106981. II. EXPERIMENTAL PROCEDURE

Dupont 6120 Pd/Ag conductive was employed as the electrode material, which was then printed and fired on 96% Al<sub>2</sub>O<sub>3</sub> substrates. Resistor pastes used in this paper were the Dupont 1600 series, including 1621, 1631, 1641, 1651, and 1661 with sheet resistances of 10<sup>2</sup>, 10<sup>3</sup>, 10<sup>4</sup>, 10<sup>5</sup>, and 10<sup>6</sup>  $\Omega$ /square, respectively. The resistor pastes were printed through a 325 mesh stainless steel screen onto Al<sub>2</sub>O<sub>3</sub> substrates. The printed samples were allowed to remain level at room temperature for 5 min, and dried at 150°C for 5–10 min. The dried thickness is 25  $\mu$ m. The resistor was fired with a 60-min cycle to a peak firing temperature of 850°C for 15 min.

The electrical properties of the samples were measured with two LCR meters (HP 4274 and HP 4191) interfaced with an HP 9836 computer for automatic data acquisition and analysis. Impedance spectroscopy was employed to analyze the electrical characteristics of the resistors.

## **III. RESULTS AND DISCUSSION**

Fig. 1 shows the relative resistance as a function of frequency for 1600 series resistors. A constant resistance is obtained for all resistors between  $10^2$  and  $10^5$  Hz. However, there are abrupt decreases in resistance at high frequencies, for all the five resistors tested. These are also indicated in Fig. 1; the larger the sheet resistance of the resistor, the smaller the frequency at which the drop of resistance occurs. A decrease in resistance takes place at  $\sim 10^5$  Hz for resistors 1661 (sheet resistors 1621 ( $R_s:10^2 \Omega/\text{square}$ ). There exist small peaks at the high frequency ends for the curves in Fig. 1, which are attributed to the resonance phenomena caused by the inductance and capacitance of the test fixture leads and the specimens.

According to Pike and Seager [2], the electrical transport between two neighboring conductive particles of a thick film resistor is via the so-called tunneling mechanism. There are tunneling barriers in the interfacial region of conductive particles as shown schematically in Fig. 2(a). The resistance of the tunneling barrier is temperature dependent [2]:

$$R(T) \propto (\sin aT/aT)[1 + \exp(E_c/2kT)]$$
(1)

where T is absolute temperature,  $E_c$  is the barrier height, k is the Boltzmann constant, and a is a parameter which depends on the barrier height.

0148-6411/92\$03.00 © 1992 IEEE



Fig. 1. Relative resistance as a function of frequency for Dupont 1600 series resistor.  $R_o$ : resistance at 1 kHz.



Fig. 2. (a) Schematic diagram of tunneling barrier model for the conductive chain of thick film resistors. (b) Equivalent circuit model of the tunneling barriers.

When two materials of different conductivities are brought into contact, interfacial polarization occurs. Blocking of charge carriers happens across the interface, and this will manifest itself as an increase in the capacitance to the outside observer. The interfacial tunneling barrier between two neighboring conductive particles has a resistance described by (1). However, the conductive particles RuO<sub>2</sub>, with a room temperature resistivity of the order of  $10^{-5}$   $\Omega$ -cm, has a very small resistance. Hence, interfacial polarization should occur in the interface between two neighboring conductive particles. Consequently, a parallel *RC* circuit, as shown in Fig. 2(b), can describe the electrical behavior in the interface region. In the equivalent circuit model, *R* represents the resistance of the tunneling barriers and *C* describes the interfacial polarization phenomenon in the interface region.

The thick film resistor consists of conductive chains distributed in an insulating glass matrix as shown schematically in Fig. 3(a). An equivalent circuit model based on the microstructure of the resistor is proposed as indicated in Fig. 3(b). The resistance  $R_q$  and capacitance  $C_q$  in Fig. 3(b)



Fig. 3. (a) Schematic diagram of thick film resistors. (b) Equivalent circuit model for thick film resistors. (c) Simplified equivalent circuit for thick film resistors. (d) Schematic complex impedance diagram of the equivalent circuit.

are associated with the impedance of the insulating glass matrix.  $R_c$  represents the resistance of the conductive particles, while R and C are related to the interface region described previously. In general, thick film glasses are insulators with a large resistance and small dielectric constant, and the electric transport is via the conductive chains, so the impedance of the  $R_gC_g$  circuit is very large. As compared to that of the conductive chains, we have  $R_g \gg R$ ,  $R_g \gg R_c$ ,  $C_g \ll C$ , and  $1/\omega C_g \gg 1/\omega C$ . Thus the equivalent circuit can be simplified as shown in Fig. 3(c). Also shown in Fig. 3(d) is the schematic complex impedance diagram of the simplified equivalent circuit.



Fig. 4. Complex impedance diagram of Dupont 1621 resistor fired for various times x.



Fig. 5. Relative resistance as a function of frequency for Dupont 1631 resistor,  $R_o$ : resistance at 1 kHz.

Fig. 4 is the impedance plot of 1621 resistor fired for multiple times. A semi-circle is obtained for resistors fired once, twice, four times, and six times as exhibited in Fig. 5. This suggests that the proposed equivalent circuit suffices to describe the electrical behavior of the thick film resistors.

The resistance of the simplified equivalent circuit R can be derived:

$$\overline{R} = R_c + \frac{R}{1 + (\omega\tau)^2} \tag{2}$$

where  $\tau = RC$  and  $R_c \ll R$ . According to (2), one finds that the resistance  $\overline{R}$  is inversely proportional to the square of the frequency. Fig. 5 gives the resistance as a function of frequency for the 1631 resistor and theoretical prediction from the equivalent circuit model. There is good agreement between these two data. The resistance at the low frequency range appears to be constant, which is attributed to the resistance of the interfacial tunneling barrier as well as that of the conductive particles, i.e.,  $(R_c + R)$ . At high frequencies, the reactance of the capacitor,  $1/\omega C$ , decreases, and the resultant resistance decreases due to the parallel transport of both RC components, as shown in Figs. 1 and 5. The frequency corresponding to the resistance drop is smaller for resistors with larger resistance. The electrical transport through the capacitor becomes non-negligible only when  $1/\omega C$  decreases to a magnitude comparable to the parallel resistance R. The larger the resistance, the larger the  $1/\omega C$ , and hence, the smaller the applied frequency, as indicated in Fig. 1.



Fig. 6. Relative change of resistance as a function of firing times.  $R_o$ : resistance for single firing.

Firing of the resistor alters the microstructure and results in a change in the resistance for thick film resistors [1], [5]-[7]. Vest's group [1], [6], [7] classified the kinetics of resistor microstructure development into six physical processes: sintering of the glass; spreading of the glass; rearrangement of the conductive particles in the presence of glass; densification of the glass; sintering of the conductive particles in the presence of glass; and Ostwald ripening of the conductive in the glass. Recently, Yamaguchi and Iizuka [5] argued that the coalescence of glass decreased the resistivity, while infiltration of glass into RuO<sub>2</sub> particles aggregates and agglomeration of RuO<sub>2</sub> particles increased the resistivity of the thick film resistor. Fig. 6 is the relative change of resistance as a function of firing times for various resistors. For resistors with large resistance, i.e., 1661, 1651, and 1641, a decrease in resistance is observed for multiple firing. However, for those with small resistance, i.e., 1631 and 1621, the resistance increases with firing time. As mentioned previously, the electrical transport of thick film resistors is via the conduction chain. The resistance should decrease when more chains and/or wider chains are formed. It is argued that for specimens with large resistance, repeated firing enhances the coalescence of glass as well as the rearrangement of the conductive particles, and more conduction chains are formed. As a result, the resistance decreases. For the low resistance specimen, some chains may be broken due to the agglomeration of conductive particles during repeated firing, and the resistance thus increases.

The frequency dispersion of the resistance mostly results from the interfacial polarization effect, which is associated with blocking of charge carriers across the interface when two materials of different conductivities are brought into contact. For the resistor system studied, the polarization originated from the interface of the tunneling barrier and the conductive particle. It is believed that repeated firing acts to change the arrangement of the conductive particles itself. Thus repeated firing should not have appreciable effect on the frequency dependence of the resistance. The frequency dependence of resistance, shown in Fig. 7, is similar for resistors fired for various times as is expected. Also indicated in Fig. 4 is the impedance of 1621 resistor after multiple firing. 1621 resistor paste has a small sheet resistance of  $10^2 \Omega$ /square after single firing. There is no appreciable change in the resistance of the conductive particle  $R_c$  for resistors after repeated firing.



Fig. 7. Relative resistance as a function of frequency for Dupont 1621 resistors fired for various times,  $R_o$ : resistance for single firing x = 0.

However, the resistance associated with the tunneling barrier R increases when samples are fired multiple times. This is caused by the agglomeration of the conductive particles after multiple firing. As discussed previously, the breaking of some conductive chains after multiple firing results in the increase of resistance of the low resistance resistor.

## IV. SUMMARY AND CONCLUSIONS

1) An equivalent circuit model for a thick film resistor is proposed, which includes the interfacial polarization effect. This model can be employed to explain the frequency response of the impedance in thick film resistors.

2) There is an abrupt decrease of resistance at frequencies larger than 1 MHz for Dupont 1600 resistor series. The decrease in resistance is attributed to the small reactance  $1/\omega C$  at high frequencies.

3) Resistance variation for thick film resistors exists after repeated firing. Resistance increases for the resistor with low intrinsic resistance, while it decreases for the high resistance specimens. The change in resistance is attributed to the rearrangement of the conduction particles. Firing times has, in fact, no appreciable influence on the frequency dependence of the specimens.

#### REFERENCES

- R. W. Vest, "Conduction mechanisms in thick film microcircuits," Final Tech. Rep. on Grant DAHC-15-70-G7 and DAHC-15-73-G8, DARPA Order No. 1642, Dec. 1975.
- [2] G. E. Pike and C. H. Seager, "Electrical properties and conduction mechanisms of Ru-based thick-film (cermet) resistors," J. Appl. Phys., vol. 48, no. 12, pp. 5152-5169, 1977.

- [3] G. Blatter and F. Greuter, "Carrier transport through grain boundaries in semiconductor," *Phys. Rev. B*, vol. 33, no. 6, pp. 3952-3966, 1986.
- [4] B. S. Chiou and M. C. Chung, "Admittance spectroscopy and trapping phenomena of ZnO based varistors," to be published.
- [5] T. Yamaguchi and K. Iizuka, "Microstructure development in RuO<sub>2</sub>-Glass thick-film resistors and its effect on the electrical resistivity," J. Amer. Ceram. Soc. vol. 73, no. 7, np. 1953–1957, 1990.
- Ceram. Soc., vol. 73, no. 7, pp. 1953-1957, 1990.
  [6] D. H. R. Sarma and R. W. Vest, "Kinetics of spreading and with application to RuO<sub>2</sub>-Glass thick film resistors," J. Amer. Ceram. Soc., vol. 68, no. 8, C-215-C-216, 1986.
- [7] S. M. Chitale and R. W. Vest, "Critical relationships between particle size, composition, and microstructure in thick-film resistors," *IEEE Trans. Comp., Hybrids, Manuf. Technol.*, vol. 11, pp. 604–610, 1988.



Wei-Yung Hsu received the B.S. and M.S. degrees from the Department of Materials Science and Engineering, National Tsing Hua University, Hsinchu, Taiwan in 1983 and 1985, respectively. He is currently working toward the Ph.D. degree in the Department of Materials Science and Engineering, Cornell University, Ithaca, NY.

His major work is the growth of ferroelectric thin film on semiconductors, with special focus on the structure-property relation which is characterized by nonlinear optical properties, electro-optics, and

second harmonic generation.



Jenq-Gong Duh received the B.S. degree in nuclear engineering from the National Tsing Hua University, Taiwan, and the Ph.D. degree in materials engineering from Purdue University, West Lafayette, IN, in 1974 and 1983, respectively.

During 1987–1988 he was a Visiting Scientist at the Department of Material Science and Engineering, Cornell University. Currently, he is a Professor with the Department of Materials Science and Engineering, National Tsing Hua University. His interests include electron microscopy, interfacial

phenomena of materials, ceramic-metal bonding, high temperature alloy, and surface modification of ceramics.



**Bi-Shiou Chiou** received the B.S. degree in nuclear engineering and the M.S. degree in health physics from the National Tsing Hua University, Taiwan, and the Ph.D. degree in materials engineering from Purdue University, West Lafayette, IN, in 1975, 1977, and 1981, respectively.

She worked as a Visiting Professor at Purdue University and a Visiting Scientist at Cornell University during 1987–1988. Currently, she is a Professor of the Institute of Electronics, National Chiao Tung University, Hsinchu, Taiwan. Her interests include

electronic ceramics, thick-film technology, and thin-film deposition.

II I