An equivalent network for resistance and temperature coefficient of resistance versus temperature and composition of thick resistive films

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Two types of elementary resistances in thick resistive films have been considered: (i) constriction resistance R_C determined by the bulk properties of conducting material and by the geometry of constriction, and (ii) barrier resistance R_B determined by the parameters of a thermally activated type of tunneling process and by the geometry of the metal-insulator-metal unit. On this basis a resistance network composed of a large number of the two types of resistances has been defined. The network has been considered as being equivalent to thick resistive film (TRF) from the point of view of the resistance and temperature coefficient of resistance (TCR). The parameters of this network have been evaluated by the computer-aided approximation of the experimental data found for RuO_2-based TRFs. On the basis of the equations derived for the network as well as the results of the approximation process, it can be concluded that the small values of the network TCR result from the superposition of the TCR of the conducting component β_C and of the temperature coefficient of barrier resistance α_B. In this superposition β_C is attenuated (by 1–2 orders of magnitude), while α_B is attenuated by only few percentages. The network has been found to be strongly barrier dominated.

I. INTRODUCTION

The problem of the explanation of the conduction mechanism in thick resistive films (TRFs) still needs further investigation. One of the common conclusions of the research in this field, until now, is that the electric charge transport in TRFs takes place via chains of conducting particles.[1–9] The chains form a percolation network, part of which is called giant cluster and spans the body of the film and terminates many times on both film electrodes. Some of the adjacent conducting particles in the giant cluster can be sintered and some can be separated by thin layers of glass. If two adjacent conducting particles are sintered, the conduction mechanism is characteristic for the conducting component and is often of the metallic type. On the other hand, the electric charge transport through the barrier in the metal-insulator-metal (MIM) unit formed by two separated conducting particles can occur by thermally activated tunneling and/or by hopping of electrons via localized states in the glass layer.

Let us characterize the mechanism of the charge transport through the barrier by the temperature coefficient of resistance (TCR) denoted α_B. Let us also denote β_C as the TCR of the conducting component. The sintered contact (constriction) will, in general, display a different TCR, α_C. The problem which can be stated at this point is how the TCR of the film, $\alpha(T,v)$, is related to α_C (T,v) [or to $\beta_C(T)$] and $\alpha_B(T,v)$, where T is the absolute temperature and v is the volume fraction of the conducting component.

This problem is, in general, very complex. One should generally consider the stochastic structure of the giant cluster in the film. Furthermore, one should also take into account the action of the substrate which, e.g., via piezoresistive effect also influences the TCR of the film. There are other effects or facts which should be taken into account; for instance, the existence of many types of thick resistive film structures such as ruthenate-based and RuO_2-based ones as well as the fact that these structures can be, in general, three or even multicomponent heterogeneous systems.

In this paper, instead of considering the giant cluster as well as the other effects mentioned above, we consider as a first step of the approach to the stated problem a resistance network which, from the point of view of resistance and TCR, is equivalent to a thick resistive film. Although this is deterministic and not a random type of network, this simplification enables us to solve the stated problem exactly. We think that the results calculated for the equivalent network are useful in research of the TRFs conduction mechanism for at least two reasons: (i) they can be used as a certain reference and starting point for further investigation of random resistance networks[7,8]; and (ii) the answer to the stated problem of how $\alpha_C(T,v)$ [or $\beta_C(T)$] and $\alpha_B(T,v)$ participate in the resultant $\alpha_N(T,v)$ of the network helps to explain the experimental fact of how very small values of TCR of thick resistive films can be obtained as the TCR of the conducting component and the TCR of the barrier in the MIM unit indicate much larger magnitudes.

As an example, we consider the experimental results for RuO_2-based TRFs. For these films $|\alpha(T,v)| \ll \alpha_C(T,v)$ and $|\alpha(T,v)| \ll |\alpha_B(T,v)|$ or $|\alpha(T,v)| < |\alpha_B(T,v)|$. Therefore, the simple tradeoff between positive values of $\alpha_C(T,v)$ and negative values of $\alpha_B(T,v)$ cannot explain the experimental observation. We find the conditions which have to be fulfilled by the network parameters in order to approximate the data and we calculate the values of these parameters for some values of the volume fraction of conducting component and temperature.

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II. THE EQUIVALENT NETWORK

A. Physics of the elementary resistances

According to the results of the investigation of conduction mechanisms[2,5–9] we assume that the resistance of the network consists of two types of units: (i) contacts (constrictions) between two sintered conducting particles of equal diameters d, it is characterized by resistance R_C and TCR α_C; and (ii) tunneling barriers in the MIM units of the type, conducting particle-glass layer-conducting particle. The tunneling barrier itself is characterized by resistance R_B and TCR α_B, while the whole MIM unit is represented by the serial connection of constriction and tunneling barrier. Both conducting particles in the MIM unit are assumed to be of diameter d.

Let us first characterize unit (i). The constriction resistance R_C is determined by the resistivity of the conducting component ρ_C, by constriction radius a and by diameter d of the conducting particles in the unit (Fig. 1). We apply the Holm model of conducting sphere with hemispherical surfaces of current inlet and outlet at both ends of sphere diameter[10] in the estimation of the constriction resistance,

$$R_C = \frac{\rho_C}{\pi}\left(\frac{1}{a} - \frac{1}{d}\right). \tag{1}$$

As the values of the constriction radius a can be of the order of several nanometers, we take into account the scattering of electrons at the surface of the constriction. It is known that for the wire of radius y the resistivity (with the assumption of surface diffuse scattering) can be approximated by[11]

$$\rho_C = \rho_C^b(1 + 3\lambda/8y), \tag{2}$$

where ρ_C^b and λ denote the bulk resistivity and electron mean free path of the conducting component. In the case of the constriction (Fig. 1), the radius determining the magnitude of the surface-scattering dependent increment of resistivity varies between a and $d/2$. We have found that, in this case, the effective resistivity can be estimated by (see the Appendix)

$$\rho_C = \rho_C^b\left[1 + \frac{3\lambda}{4(d-s')}\sin^{-1}\left(1 - \frac{s'}{d}\right)\right], \tag{3}$$

where s' is determined by the values of d and a according to

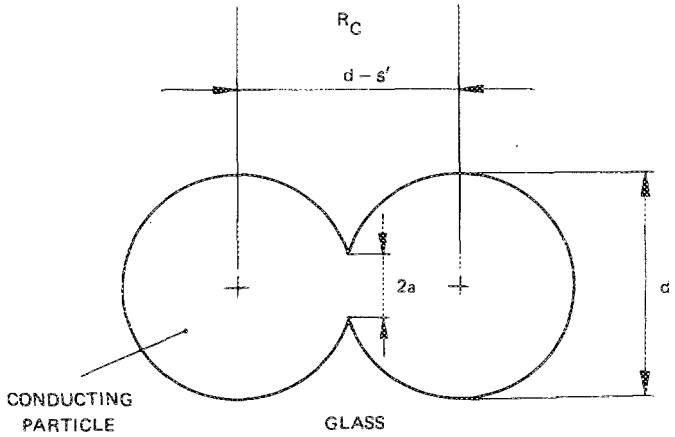

FIG. 1. Model of two adjacent conducting particles forming the sintered contact.

Fig. 1 (see also Fig. 9). In order to calculate the electron mean free path we use the following formula[11]:

$$\lambda = \frac{\hbar}{q^2}\left(\frac{3\pi^2}{n^2}\right)^{1/3}\frac{1}{\rho_C^b}, \tag{4}$$

where $\hbar$ is the Planck constant, q is the elementary charge, and n is the electron concentration in the conducting component.

In this paper we consider the experimental results obtained for RuO_2-based TRFs and therefore we recall resistivity versus the temperature characteristic of ruthenium dioxide given by Ryden *et al.*[12] (Fig. 2). This dependence can be approximated in the considered temperature range by the straight-line equation,

$$\rho_C^b(T) = \rho_C^b(T_i)[1 + \beta_C(T_i)(T - T_i)], \tag{5}$$

where

$$\beta_C(T_i) \equiv \frac{1}{\rho_C^b(T_i)}\left(\frac{d\rho_C^b}{dT}\right)_{T=T_i}. \tag{6}$$

T_i is a constant temperature at which the parameter of Eq. (5) are evaluated. From the data given in Fig. 2 it can be estimated that at $T_i = 293$ K,

$$\rho_C^b(293) \simeq 3.4\times10^{-7}\ \Omega\ \text{m}$$

and

$$\beta_C(293) \simeq 5810\times10^{-6}\ \text{K}^{-1}.$$

Equation (5) is valid if $T > T_i - 1/\beta_C(T_i)$, which for the above values of T_i and $\beta_C(T_i)$ gives $T > 121$ K. The upper limit of temperature up to which Eq. (5) together with the found values of the parameters can be used is greater than 600 K (Ref. 12). Since the experimental results for our RuO_2-based TRFs are given in the range 213 K$\leqslant T \leqslant$393 K (see Fig. 4), we can utilize in the following the estimated above-straight-line equation. Using Eqs. (1)–(5) we find

$$R_C(T) = \frac{\rho_C^b(T_i)}{\pi}\left(\frac{1}{a} - \frac{1}{d}\right)[1 + \beta_C(T_i)(T - T_i)] + R_C^L \tag{7}$$

and

$$R_C^b(T_i) = \frac{\rho_C^b(T_i)}{\pi}\left(\frac{1}{a} - \frac{1}{d}\right), \tag{8}$$

where

$$R_C^L = \frac{3\hbar}{4q^2(d-s')}\left(\frac{3}{\pi n^2}\right)^{1/3}\left(\frac{1}{a} - \frac{1}{d}\right)\sin^{-1}\left(1 - \frac{s'}{d}\right) \tag{9}$$

is the surface scattering dependent increment of the contact resistance. Using Eq. (7) we obtain

$$\alpha_C(T) \equiv \frac{1}{R_C(T)}\frac{dR_C(T)}{dT} = \frac{\beta_C(T_i)}{1 + \beta_C(T_i)(T - T_i) + R_C^L/R_C^b(T_i)}. \tag{10}$$

Using Eqs. (8) and (10),

$$R_C(T) = R_C(T_i)[1 + \alpha_C(T_i)(T - T_i)], \tag{11}$$

where

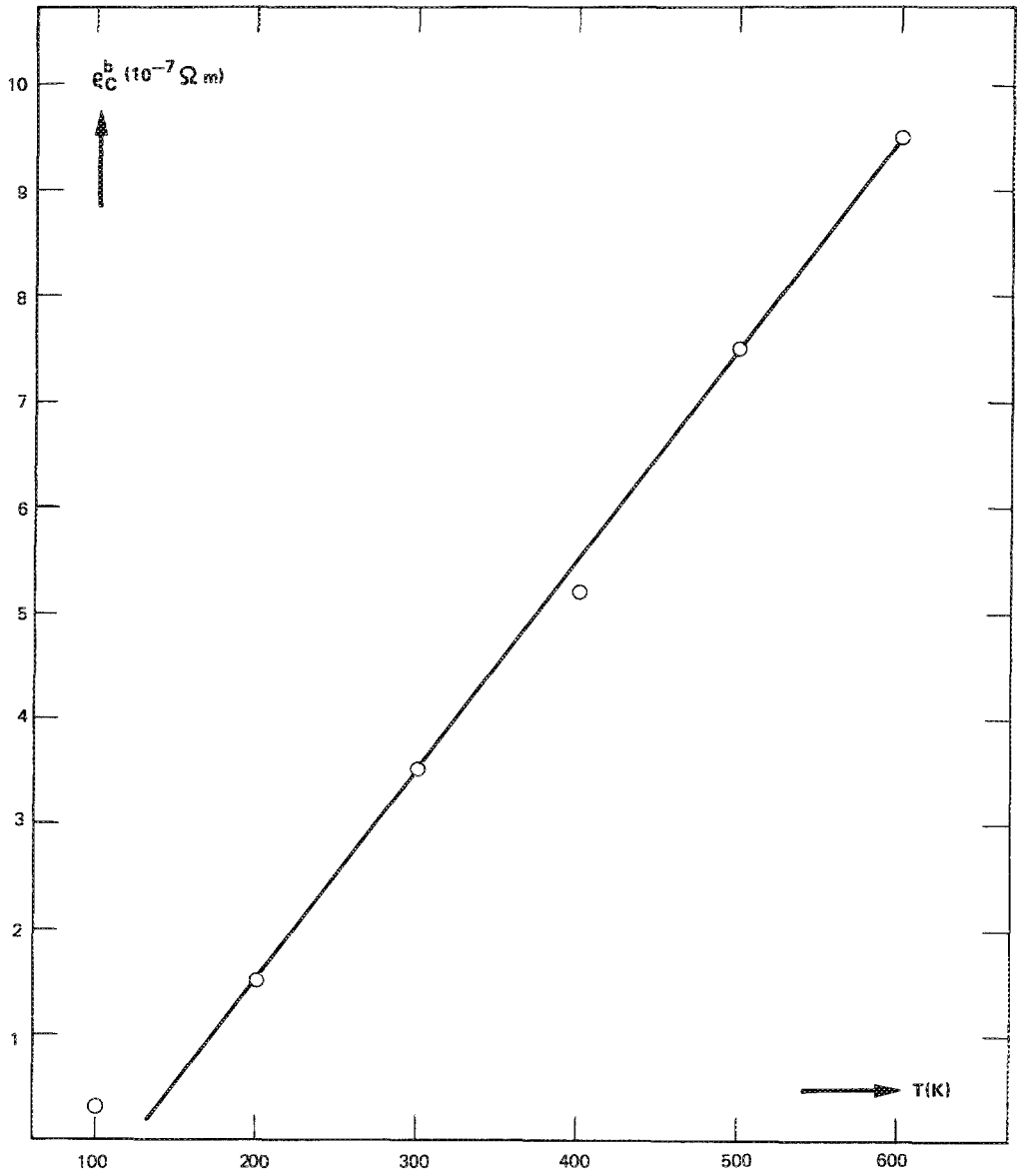

FIG. 2. Ruthenium dioxide bulk resistivity vs temperature; the points are taken from the curve published by Ryden *et al.* (see Ref. 12) and the straight line represents Eq. (5) for $T_i = 293$ K and $\beta_C = 5810 \times 10^{-6}$ K^{-1}.

$$R_C(T_i) = R_C^b(T_i) + R_C^L, \tag{12}$$

$$\alpha_C(T_i) = \beta_C(T_i) R_C^b(T_i) / [R_C^b(T_i) + R_C^L]. \tag{13}$$

As can be seen from Eqs. (11)–(13) the scattering of electrons at the constriction surface results in the increasing of the constriction resistance and the decreasing of its temperature coefficient.

In order to characterize unit (ii) it is necessary to know the mechanism of charge transport through the barrier. In this paper we assume that this mechanism is a thermally activated tunneling. This assumption is based on the fact that we consider the TRFs with low and medium resistance per square (see Sec. III), thus we are rather far from the percolation threshold of our system. We think, based also on the analysis given by Pike and Seager[2] and on our $1/f$ noise research,[13] that this mechanism is of greatest physical acceptance in the considered TRFs. The approach presented can be used as well with any other mechanism of the charge transport through the barrier e.g., with hopping conduction, provided that for this process the barrier resistance and its temperature coefficient can be evaluated. With the assumed mechanism, R_B and α_B are calculated for the simplified model of the MIM unit given in Fig. 3. The geometry of the MIM unit given in Fig. 3 has been assumed for the sake of simplicity of the tunneling area and the barrier resistance calculation [Eq. (16)]. Such geometry facilitates the direct definition of width s and of the barrier tunneling area. It was not the purpose of this paper to investigate the shape of conducting particles and the glass layers in the MIM units. Similar comments also concern the simplified model of two sintered conducting particles given in Fig. 1. We think that the results of the calculations presented for those simplified geometries are representative and useful from the point of view of the problem stated in the Introduction. The barrier resistance for thermally activated tunneling is[2]

$$R_B(T) = \tfrac{1}{2} R_B^0 \frac{\sin bT}{bT} \left[1 + \exp\left(\frac{E}{kT}\right)\right], \tag{14}$$

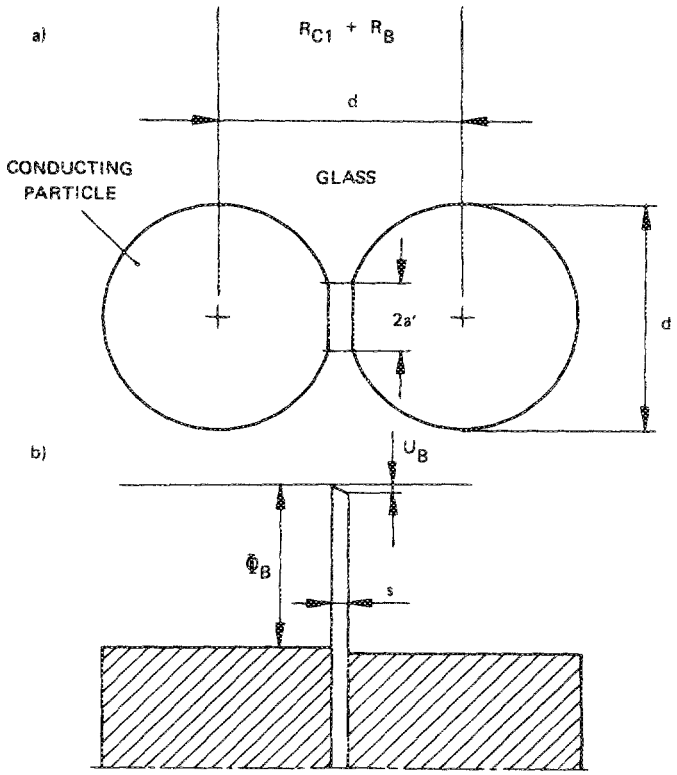

FIG. 3. A model of two adjacent conducting particles forming the MIM unit: (a) geometry and (b) band diagram.

where

$$b = (\pi k/\hbar)s(2m/q\phi_B)^{1/2}, \tag{15}$$

R_B^0 is the temperature-independent factor in the barrier resistance equation, k is the Boltzmann constant, E is the activation energy determining thermal equilibrium concentration of positively or negatively charged conducting particles in the film, m is the electron mass, and ϕ_B is the barrier height. If the voltage U_B [Fig. 3(b)] applied to the barrier is much smaller than the barrier height ϕ_B (Ref. 14),

$$R_B^0 = \frac{4\pi^2\hbar^2 s}{q^2 A(2mq\phi_B)^{1/2}} \exp\left(\frac{8mqs^2\phi_B}{\hbar^2}\right)^{1/2}. \tag{16}$$

From Eq. (14) we have

$$\alpha_B(T) = \frac{1}{R_B(T)}\frac{dR_B(T)}{dT} = b\cot(bT) - \frac{1}{T} - \frac{E}{kT^2}\left[1+\exp\left(-\frac{E}{kT}\right)\right]^{-1}. \tag{17}$$

The activation energy E for the process of thermally activated tunneling has been defined in different ways until now.[2,15–17] It is often formulated in terms of the charging energy,[17]

$$E_c = \frac{q^2}{2\pi\epsilon\epsilon_0 d}\frac{1}{K(d,s)}, \tag{18}$$

where ϵ_0 is the permittivity of vacuum, ϵ the relative permittivity of glass, and $K(d,s)$ is a function of d and s whose form depends on the shape and arrangement of the conducting particles and on interaction between the pair of charges. Half of the charging energy is assigned to each particle, so the activation energy is $E = E_c/2$.

The total resistance of the MIM unit is a serial connection of the barrier resistance R_B and of the constriction resistance R_{C1}, which is described by equations similar to Eqs. (11)–(13) with the parameters $R_{C1}(T_i)$, $\alpha_{C1}(T_i)$, $R_{C1}^b(T_i)$, and R_{C1}^L. The values of these parameters differ only by a small percentage from the values of $R_C(T_i)$, $\alpha_C(T_i)$, $R_C^b(T_i)$, and R_C^L (see the Appendix).

B. The resistance and TCR of the network

We define the network of M identical chains connected in parallel. Each chain consists of a number K_C of resistors characterized by resistance R_C and TCR α_C, and of a number K_B of resistors characterized by resistance R_B and TCR α_B. In the network there are $N_C = K_C M$ and $N_B = K_B M$ of the two described types of resistors, respectively.

In order to relate the parameters of the network to the structure of the films we assume that the number N_C of the resistors described by Eqs. (7)–(13) equals $\frac{1}{3}$ of the total number N of the conducting particles in the giant cluster of the film. The number N can be estimated by the formula

$$3N_C(v) = N(v) \simeq (6Vv/\pi d^3)g(v), \tag{19}$$

where V is the volume of the film, and $g(v)$, is the percolation probability.[4] Since the values of s' and s are small enough compared with d we can still approximate in Eq. (19) the shape of conducting particles given in Figs. 1 and 3 by the sphere of diameter d. The assumption of $N = 3N_C$ is justified by the fact that the constriction resistance for the model given in Fig. 1 is, in our case, only 2.5% smaller than the constriction resistance in Fig. 3 and that $N_B \ll N_C$ (see the Appendix).

It can be shown from the above definition of the network that its resistance and TCR are

$$R_N(T,v) = \frac{N_C(v)R_C(T) + N_B(v)R_B(T,v)}{M^2(v)}, \tag{20}$$

$$\alpha_N(T,v) = \alpha_C(T)\Big/\left(1+\frac{N_B(v)R_B(T,v)}{N_C(v)R_C(T)}\right) + \alpha_B(T,v)\Big/\left(1+\frac{N_C(v)R_C(T)}{N_B(v)R_B(T,v)}\right), \tag{21}$$

where

$$\alpha_N(T,v) = \frac{1}{R_N(T,v)}\frac{dR_N(T,v)}{dT}. \tag{22}$$

We have assumed that in the first approximation the constriction resistance and its temperature coefficient are both independent of the volume fraction of conducting component. This is not a substantial limitation for the range of values of the volume fraction of the conducting component (0.105 to 0.203) considered in this paper (see Sec. III). On the basis of Eq. (20) the effective constriction resistance $R_m(T,v)$ and the effective barrier resistance $R_b(T,v)$ can be defined as

$$R_m(T,v) = N_C(v)R_C(T)/M^2(v), \tag{23}$$

$$R_b(T,v) = N_B(v)R_B(T,v)/M^2(v). \tag{24}$$

Similarly from Eq. (21) the constriction part γ_C and the barrier part γ_B of the network TCR can be introduced as

$$\gamma_C(T,v) = \alpha_C(T)\Big/\left(1+\frac{N_B(v)R_B(T,v)}{N_C(v)R_C(T)}\right) = \alpha_C(T)\Big/\left(1+\frac{R_b(T,v)}{R_m(T,v)}\right), \tag{25}$$

$$\gamma_B(T,v) = \alpha_B(T,v)\Big/\left(1+\frac{N_C(v)R_C(T)}{N_B(v)R_B(T,v)}\right) = \alpha_B(T,v)\Big/\left(1+\frac{R_m(T,v)}{R_b(T,v)}\right). \tag{26}$$

Now Eqs. (20) and (21) take the form

$$R_N(T,v) = R_m(T,v) + R_b(T,v), \tag{20a}$$

$$\alpha_N(T,v) = \gamma_C(T,v) + \gamma_B(T,v). \tag{21a}$$

From Eq. (21) it can be seen that the TCR of the network α_N is a superposition of TCRs α_C and α_B. Temperature coefficients of both elementary resistances are divided in Eq. (21) by expressions being temperature and volume fraction dependent. Both of these expressions take values greater than unity forming, in effect, some weights which attenuate the contributions of α_C and α_B in the resultant TCR of the network. The degree of this attenuation depends also on the total number of constrictions and constriction resistance product as well as on the total number of barriers and barrier resistance product.

III. EXPERIMENTAL PROCEDURE AND RESULTS

Our network should indicate the characteristics $R_N(T,v)$ and $\alpha_N(T,v)$ offering a good approximation of the ones observed in real TRFs. As an example, we have taken into account the data obtained for RuO_2-based films. These films were made of four pastes having the following values of the volume fraction of RuO_2 /v/: 0.105, 0.120, 0.156, 0.203 (Ref. 9). All the pastes were produced from the same starting materials. The average particle size of RuO_2 powder has been estimated by the BET surface area method as equal to 12 nm. The glass powder has indicated the average particle size as 1.6 μm, relative permittivity 8.3, and composition as 65% of PbO, 25% of SiO_2, 10% of B_2O_3 (% by weight).[13] The films were screen printed on alumina substrates and fired in a continuous air atmosphere tunnel-type furnace at a peak temperature equal to 800 °C; the period of time the films were at this temperature was 10 min. All films are squares 5×5 mm. Their average thicknesses and resistances have been estimated as 12.9, 18.3, 13.7, 19.0 μm and 1244.1, 435.24, 138.12, 26.462 Ω for the values of v: 0.105, 0.120, 0.156, 0.203, respectively. The Pd-Ag type of conductive films have been used as contacts. Six or 12 films have been produced from each paste and the characteristics of resis-

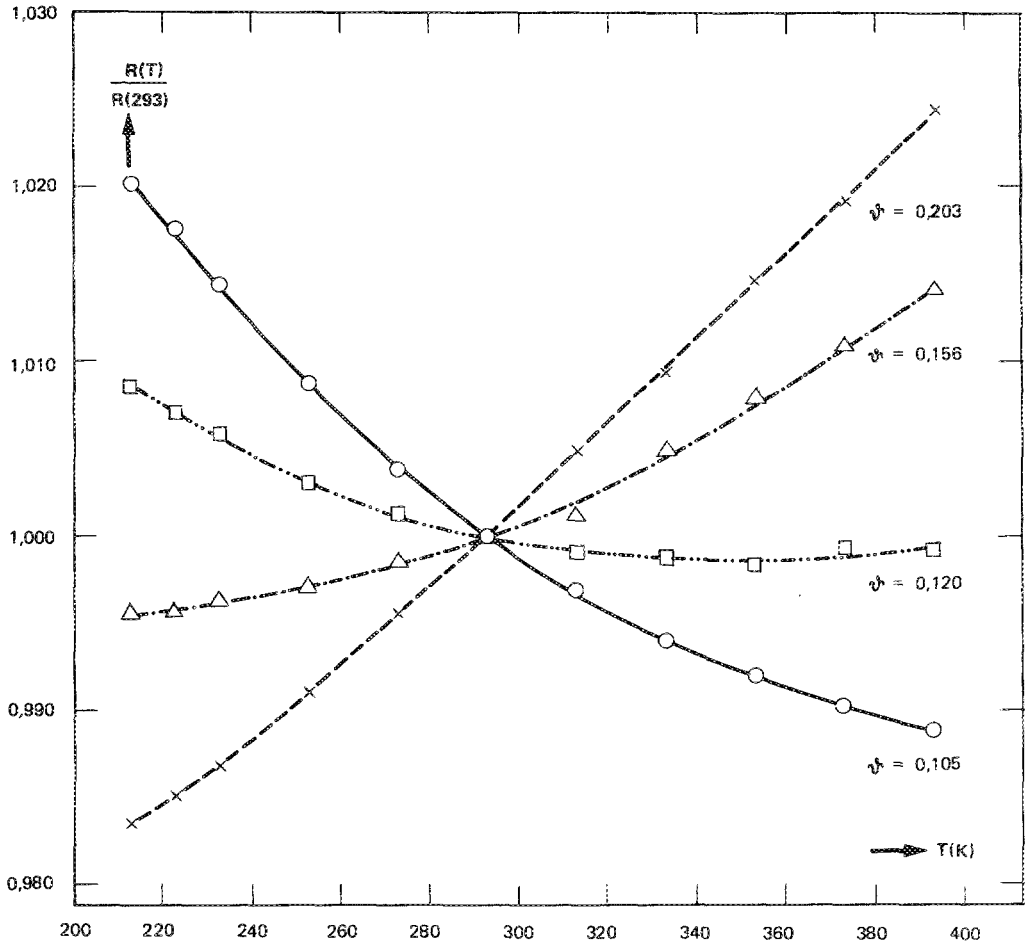

FIG. 4. Resistance vs temperature of RuO_2-based TRFs with different values of RuO_2 volume fraction; points represent the experimental data and the lines are drawings of the function $R_N(T,v)$ [Eq. (27)] for the optimum values of parameters B, C, E given in Table I; all data are normalized with respect to the value of $R(T,v)$ at $T = 293$ K.

tance versus temperature have been estimated for each film in the temperature range from 213 up to 393 K. The characteristics of all films produced from one particular paste have shown the same type of behavior and TCR values scattered within $\pm 20\%$ limits. Four characteristics of resistance versus temperature at four considered values of v have been evaluated in terms of average values of resistance normalized against resistance at $T = 293$ K; these characteristics are shown in Fig. 4.

IV. CALCULATION OF THE NETWORK PARAMETERS

We have approximated the obtained characteristics of resistance versus temperature using the approximating function $R_N(T,v)$ composed in terms of Eqs. (11)–(13), (14), and (20).

$$R_N(T,v) = C(v)\left[1 + \alpha_C(T_i)(T - T_i)\right] + B(v)\,\frac{\sin bT}{bT}\left(1 + \exp\frac{E(v)}{kT}\right), \quad (27)$$

where

$$C(v) = N_C(v)\left[R^{\,b}_C(T_i) + R^{\,L}_C\right]/M^2(v) = R_m(T_i,v), \quad (28)$$

$$B(v) = N_B(v)R^{\,0}_B/2M^2(v). \quad (29)$$

First, the values of $R^{\,b}_C(T_i)$, $R^{\,L}_C$, and b have been calculated with the help of Eqs. (8), (9), and (15). In this calculation the following data have been used: $T_i = 293$ K, $a = 1.75$ nm (Ref. 6), $\rho_C(T_i) = 3.4\times10^{-7}\ \Omega$ m (see Fig. 2), $d = 12$ nm, $s' = 0.5218$ nm, $n = 6.4\times10^{28}\ \mathrm{m}^{-3}$ (Ref. 18), $m = m_0$ (i.e., the rest mass of an electron), $s = 0.5$ nm, and $\phi_B = 1$ V. The last two values are based on the similar equivalent network evaluation made from the viewpoint of resistance and relative power spectral density of $1/f$ noise.[13] In the result $R^{\,b}_C(T_i) = 52.82\ \Omega$, $R^{\,L}_C = 10.28\ \Omega$, and $b = 6.934\times10^{-4}\ \mathrm{K}^{-1}$ have been found. Using these values as well as $T_i = 293$ K and $\beta_C(T_i) = 5810\times10^{-6}\ \mathrm{K}^{-1}$ and Eq. (13), the optimum values of the parameters $B(v)$, $C(v)$, and $E(v)$ have been evaluated by computer calculation of the minimum of the quality factor,

$$Q(B,C,E,v) = \sum_{j=1}^{P}\left\{R_N\left[B(v),C(v),E(v),T_j\right] - R(T_j,v)\right\}^2, \quad (30)$$

where $R_N[B(v),C(v),E(v),T_j]$ is given by Eq. (27). $R(T,v)$ represents the resistance versus temperature data obtained from the experiments for discrete values of temperature T_j and volume fraction v, while $P = 11$ is the number of temperature values T_j used in the measurements (see Fig. 4). The calculated optimum values of the parameters and the values of the quality factor Q have been summarized in Table I.

TABLE I. The optimal values of the parameters B, C, and E of Eq. (27) for RuO_2-based films having resistance vs temperature characteristics given in Fig. 4. R denotes the average values of the film resistances found from the measurements, and Q the quality factor [see Eq. (30)].

v	$C(\Omega)$	$B(\Omega)$	E(meV)	$R(\Omega)$	$Q(\Omega^2)$
0.105	27.27	573.1	3.269	1244.1	2.467
0.120	14.96	201.3	2.466	435.24	1.46×10^{-1}
0.156	8.612	62.23	2.303	138.12	4.034×10^{-2}
0.203	1.873	12.12	1.082	26.462	1.116×10^{-4}

On the basis of Eqs. (28) and (29) we have

$$N_B(v) = 2B(v)N_C(v)R_C(T_i)/C(v)R^{\,0}_B, \quad (31)$$

$$M(v) = \left[N_C(v)R_C(T_i)/C(v)R^{\,0}_B\right]^{1/2}. \quad (32)$$

Now with the help of Eqs. (12), (16), (19), (31), (32), and the values of v, $B(v)$, and $C(v)$ given in Table I, $R^{\,0}_B$, $N_C(v)$, $N_B(v)$, and $M(v)$ have been estimated. In this estimation the abovementioned values of ϕ_B, s, v, and d have been used and in addition $A = \pi(a')^2$, where $a' = 1.714$ nm results from Fig. 3 for $s = 0.5$ nm, $V = L^2t$, where $L = 5$ mm is the length and t the thickness of the film (see Sec. III); $g(v)$ has been assumed as equal to 0.9 for $v = 0.105$ and 0.120, while as equal to 1 for $v = 0.156$ and 0.203 (Ref. 4). From the calculation $R^{\,0}_B = 2.879\times10^5\ \Omega$ has been obtained among others. Next, using Eqs. (10) and (17) the temperature coefficients $\alpha_C(T)$ and $\alpha_B(T)$ at four v values and $\beta_C(T)$ have been evaluated and shown in Fig. 5. For comparison the constriction part $\gamma_C(T,v)$ and the barrier part $\gamma_B(T,v)$ of the network TCR as well as the resultant $\alpha_N(T,v)$ have been calculated with the help of Eqs. (25), (26), and (21a) [see Figs. 6(a)–6(d)]. As can be seen from Figs. 5 and 6, $\gamma_C(T,v)$ is approximately 10 to 75 times smaller than the respective $\alpha_C(T)$ and approximately 11 to 100 times smaller than the respective $\beta_C(T)$. On the other hand, the absolute value of $\gamma_B(T,v)$ is only slightly smaller than the respective absolute value of $\alpha_B(T,v)$. The characteristics of $\gamma_C(v)$, $\gamma_B(v)$, and the resultant $\alpha_N(v)$ all at $T = 293$ K have been shown in Fig. 7. Figures 6 and 7 illustrate how the tradeoff between the constriction part $\gamma_C(T,v)$ and the barrier part $\gamma_B(T,v)$ results in small absolute values of $\alpha_N(T,v)$. In Fig. 8 the network resistance R_N, the effective barrier resistance R_b, and the effective constriction resistance R_m, have been shown versus volume fraction of RuO_2. In Table II we have shown the resistance fraction of the effective barrier resistance (fractional barrier resistance) $\xi = R_b/(R_b + R_m)$, the ratio of effective barrier resistance to the effective constriction resistance $\psi = R_b/R_m$, the number of barriers per one chain N_B/M and the offset voltage $U_{\mathrm{off}} = (N_B/M)kT/q$, above which the network starts to be strongly nonlinear. As can be seen from Fig. 8 and Table II, the network resistance is strongly barrier dominated. On the basis of Table II we can also observe that both the number of barriers per one chain as well as the offset voltage U_{off} decrease as the volume fraction of RuO_2 increases.

V. DISCUSSION

It has been shown in the previous sections how the parameters of the equivalent network can be calculated on the one hand on the basis of the equations derived from the physical model of the conduction mechanism and on the other hand on the basis of experimental data obtained for RuO_2-based TRFs. Equations (20)–(22) and (27) offer a very good description of the experimental characteristics of resistance and TCR of the films versus temperature and composi-

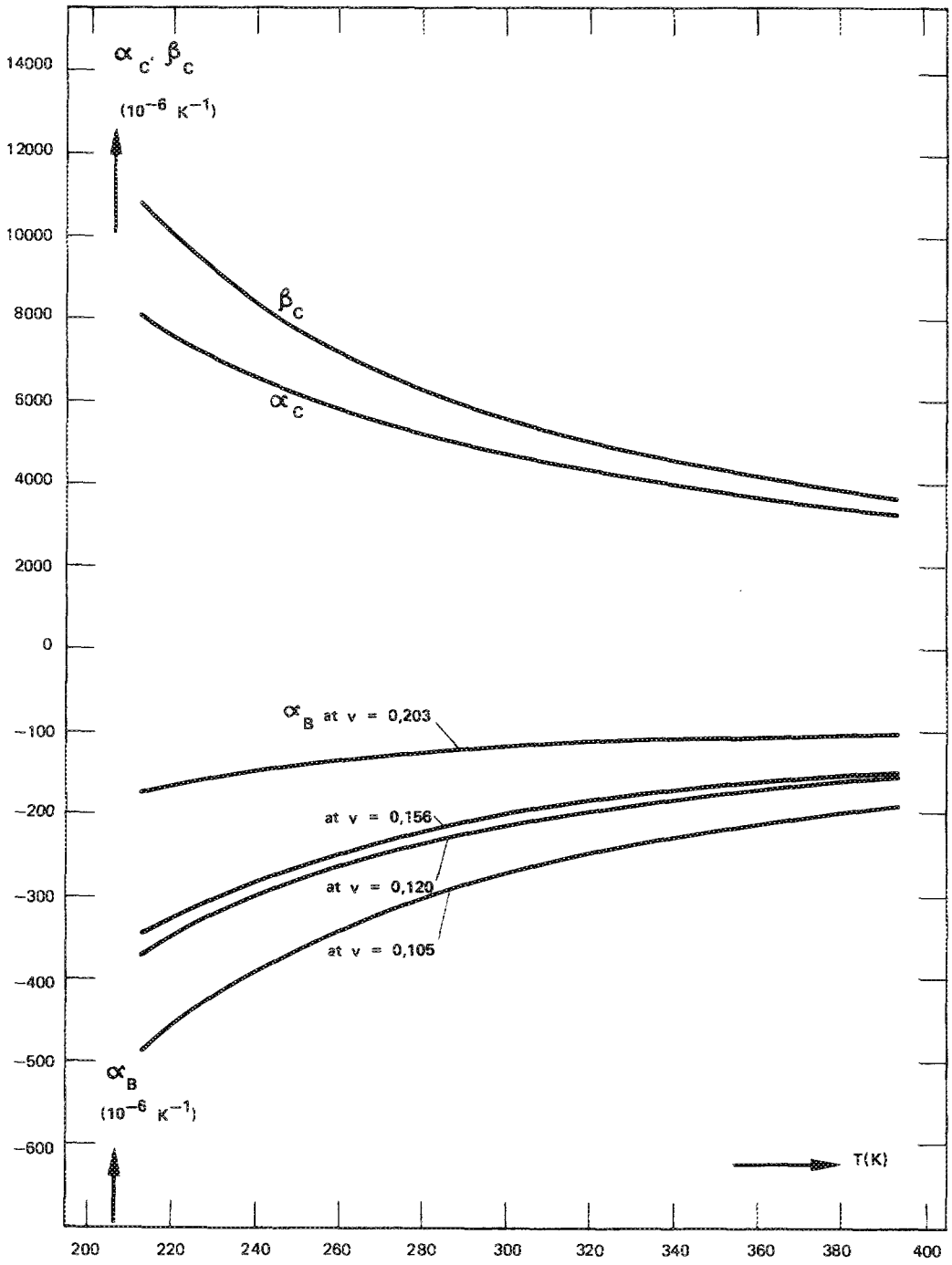

FIG. 5. Temperature coefficient of constriction resistance $\alpha_C(T)$ [Eq. (10)], bulk TCR of RuO_2 $\beta_C(T)$ [Eq. (10) with $R_C^L = 0$] and temperature coefficient of barrier resistance $\alpha_B(T,v)$ [Eq. (17)] vs temperature. Different course of $\alpha_B(T)$ at different v values results from different values of activation energy E obtained from the approximation procedure (see Table I).

tion. In particular, this can be concluded from Fig. 4 where the experimental data are shown together with the drawings of the approximating function [Eq. (27)] for the calculated optimum values of the parameters B, C, and E (Table I). The measured and the calculated values of resistance differ by no more than 0.05% at any pair of values of T and v and the accuracy of the approximation at any particular experimental point is limited by the scattering of the experimental results rather than by the quality of approximation function and/or procedure. In addition, the values of the network parameters, both assumed and calculated can, in general, be accepted from the viewpoint of technology and physics of RuO_2-based TRFs. However, discussion of some values of the network parameters is necessary.

From the previous investigation of the approximation procedure[9] we infer that the assumption of the constant value of b is essential in Eq. (27). Otherwise, if the value of b is calculated as the fourth variable parameter, one obtains the values of barrier width s [see Eq. (15)] that cannot be accepted from the point of view of the thermally activated tunneling process. Therefore in this paper we have assumed acceptable values of $s = 0.5$ nm and $\phi_B = 1$ V on the basis of our $1/f$ noise research.[13] Using these values of s and ϕ_B we have calculated, with the help of Eq. (15), the value of b and

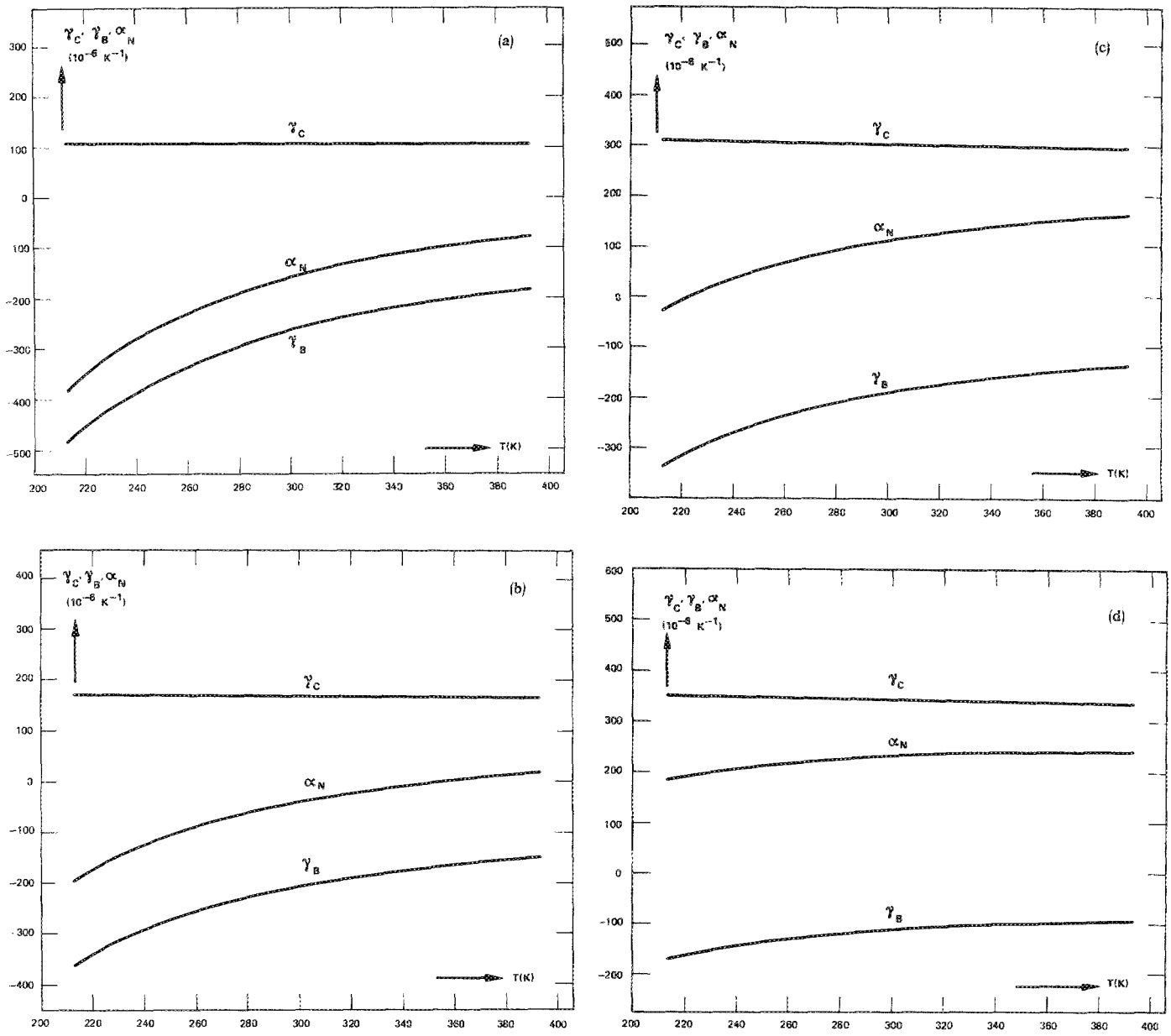

FIG. 6. The constriction part $\gamma_C(T)$ [Eq. (25)] and the barrier part $\gamma_B(T)$ [Eq. (26)] of the network TCR, and the resultant TCR of the network itself, α_N [Eq. (21)] all vs temperature at four different values of RuO_2 volume fraction: (a) $v = 0.105$, (b) $v = 0.120$, (c) $v = 0.156$, (d) $v = 0.203$; the α_N values are calculated as differential TCR [Eq. (22)] of $R_N(T,v)$ given by Eq. (27) so they fit the experimental data of Fig. 4.

have used this value as a constant parameter in Eq. (27). The obtained good accuracy of the approximation of our experimental results is an introductory justification of the assumed values of s and ϕ_B. Furthermore, we have stated that the value of E must be composition dependent; i.e., must be the variable parameter in order to obtain satisfactory quality of the approximation of resistance versus temperature characteristics (Fig. 4). Consequently, in the approximation process we have calculated the optimum values of three variable parameters of Eq. (27): B, C, and E (see Table I). However, the value of activation energy E can also be directly calculated using Eq. (18) as $E = E_c/2$. Using $\epsilon = 8.3$ (Ref. 13), $d = 12$ nm, and $K(d,s) = 3$ (Ref. 2) we find $E \simeq 4.82$ meV independent of v. As can be seen from Table I the values of E calculated from the approximation process are definitely smaller than the given value and moreover they decrease with v increasing. Some explanation of the observed divergence of the values of E can be found if we admit the dependence of $K(d,s)$ on the film composition. The explicit form of this dependence has not been known until now and the problem of its determination was not considered in this paper. Some indication concerning this dependence could, however, be inferred on the basis of the considered results.

From Fig. 8 and Table II we infer that the network resistance is strongly barrier dominated. The estimated values of $R_b/(R_b + R_m)$ are very close to 1. These values are definitely greater than the values of the fractional barrier resistance

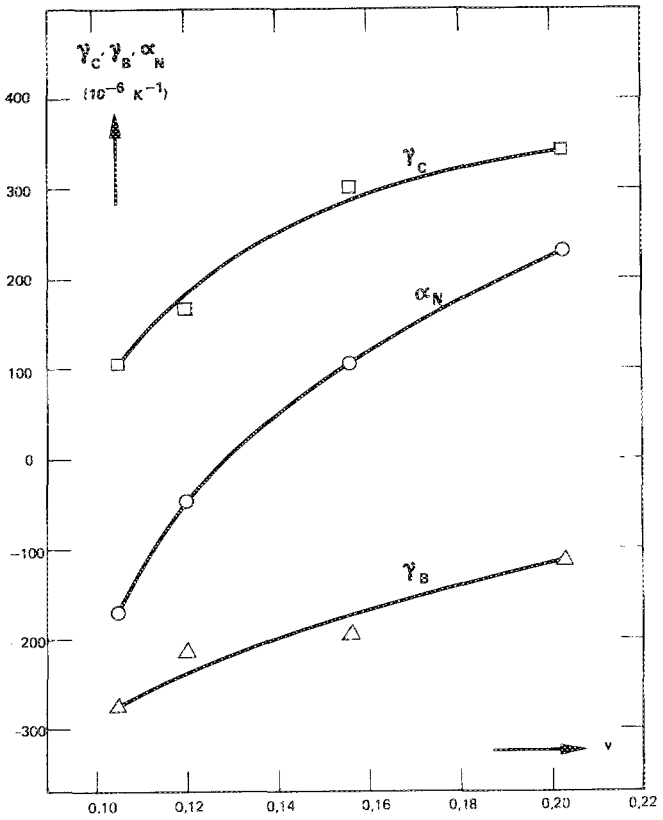

FIG. 7. Two parts of the network TCR: constriction part $\gamma_C(v)$ [Eq. (25)] and the barrier part $\gamma_B(v)$ [Eq. (26)] as well as the resultant TCR of the network itself [Eq. (21)] vs volume fraction of RuO_2 all at $T = 293$ K. The points represent the values of the respective quantity found from the experiment or calculated for the network. The lines drawn visualize the behavior of the calculated quantities vs v.

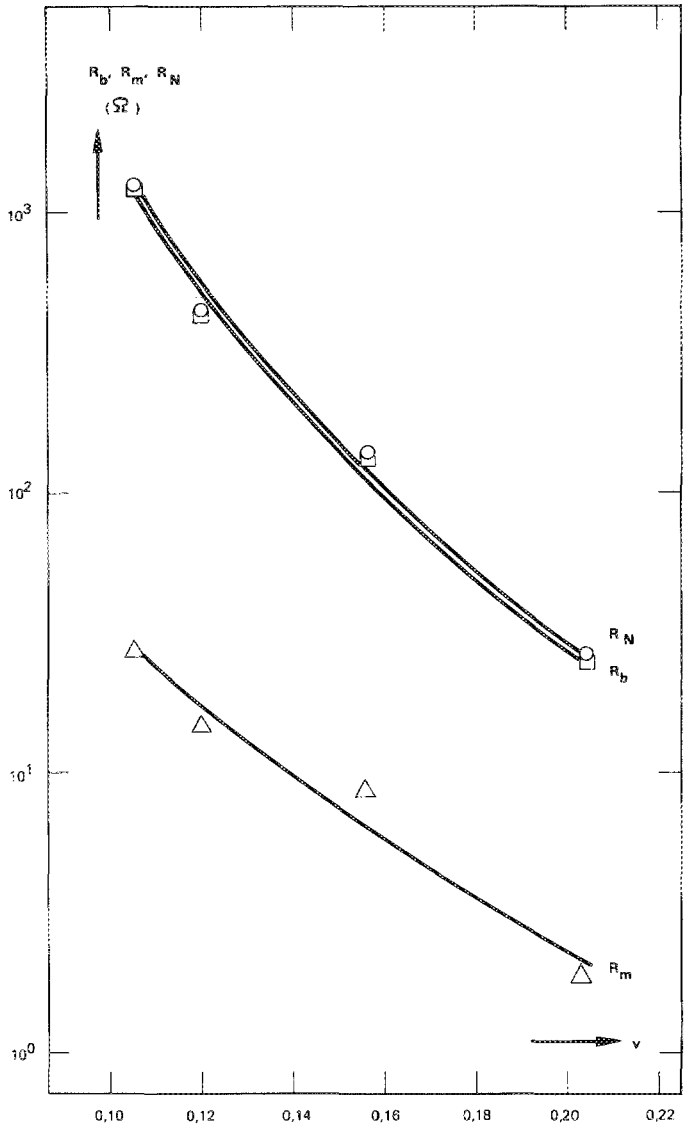

FIG. 8. The effective constriction resistance $R_m(v)$ [Eq. (23)], the effective barrier resistance $R_b(v)$ [Eq. (24)], and the resultant resistance of the network $R_N(v)$ [Eqs. (20) or (20a)] vs volume fraction of RuO_2 all at $T = 293$ K. The points represent the values of the respective resistance calculated for the network or resistance measured for the films since the measured and calculated values differ by less than 0.05%. The lines drawn visualize the behavior of the respective resistance vs v.

given by Pike and Seager in the range 0.7–0.75 (Ref. 2). From Table II we also see that the values of the offset voltage U_{off}, above which the network starts to be strongly nonlinear, are quite acceptable from the viewpoint of the electric endurance of TRFs with these resistances (see Table I) and dimensions (5 mm × 5 mm).

From the data shown in Figs. 5 and 6 it is seen that the constriction part of the network TCR $\gamma_C(T,v)$ is approximately 10 to 75 times smaller than the temperature coefficient of constriction resistance $\alpha_C(T)$ and approximately 11 to 100 times smaller than the bulk TCR of the conducting component $\beta_C(T)$. Thus the magnitude of the TCR of the conducting component is attenuated in two steps in its contribution in the resultant TCR of the network. First, the magnitude of $\beta_C(T)$ is decreased by the effect of electrons scattering at the surface of the constriction [Eq. (10)]. Second, the TCR of constriction $\alpha_C(T)$ is attenuated by the inherent property of the network [Eq. (25)]. The latter attenuation is generally stronger than the former in medium and high temperatures. However, the surface-scattering dependent attenuation of the TCR of the conducting component increases strongly with T decreasing. On the other hand, the magnitude of the barrier part of the network TCR $\gamma_B(T,v)$ is only few percent smaller than the magnitude of the temperature coefficient of the barrier resistance $\alpha_B(T,v)$. The sum

$$\gamma_C(T,v) + \gamma_B(T,v) = \alpha_N(T,v)$$

evaluates the small values of the resultant TCR of the network. This answers our question stated in the Introduction of how the network TCR $\alpha_N(T,v)$ is related to the tem-

TABLE II. Some physical parameters of the equivalent network at $T = 293$ K.

v	$\xi = \frac{R_b}{R_b + R_m}$	$\psi = \frac{R_b}{R_m}$	$\frac{N_B}{M}$	$\frac{N_B}{M}\frac{kT}{q}$ (V)
0.105	0.978	44.6	2.03×10^4	512
0.120	0.966	28.1	1.23×10^4	309
0.156	0.938	15.0	5.18×10^3	131
0.203	0.929	13.1	2.92×10^3	73.6

perature coefficients of two elementary resistances. The answer is inferred from Eqs. (21), (21a), (25), (26), and their evaluation from our experimental data. Thus we find that the effect of the attenuation of the temperature coefficients of the elementary resistances is the property of the network. This general property of the network composed of a large number of resistors of different types has been quantitatively determined for the technological and physical characteristics of our RuO_2-based TRFs. Thus we postulate that the property of the attenuation of the temperature coefficients of elementary resistances in their contribution to the resultant TCR is the general property of TRFs. The following facts should be considered in further research of TRFs in this area: (i) the randomness of the structure and thus the randomness of the network, (ii) the existence of the spectrum of values of each parameter instead of singular values as considered in this paper, and (iii) the possibility of other mechanisms of electric charge transport through the barrier with special attention to conduction by hopping of electrons via weakly localized states within the glass layer.

Fact (i) has already been considered in our next two papers.[7,8] The model of a very narrow band of weakly localized states in the intergranular material has been presented by Hill[19] and also considered by Ansell.[20] The quantitative elaboration of such a model could be utilized and introduced into the equivalent network proposed in this paper. It is clear that the effect of attenuation of temperature coefficients of the elementary resistances due to the many particle structure of the network would also be found with that model and equations similar to Eq. (21) would be valid.

APPENDIX: ESTIMATION OF THE VALUES OF THE CONSTRICTION RESISTANCES R_C AND R_{C1}

Resistivity of the wire of radius y for the case of surface diffuse scattering can be estimated by [see Eq. (2)][11]

$$\rho_C/\rho_C^b = 1 + 3\lambda/8y, \tag{A1}$$

where the notations used are the same as those in Sec. II A. According to Fig. 9 the radius of the constriction varies between a and $d/2$ and this type of variation continues throughout the chain formed of sintered-type and isolated-type pairs of adjacent conducting particles. We introduce a weight for the magnitude of the surface-scattering-dependent increment of the resistivity of constriction with the radius y. This weight equals $2\,dx/(d-s')$. Thus the effective increment of the constriction resistivity can be approximated by

$$\frac{\Delta\rho_C}{\rho_C^b} = \frac{3\lambda}{4(d-s')}\int_0^l \frac{dx}{[d^2/4-(x-1)^2]^{1/2}}, \tag{A2}$$

where $l = (d-s')/2$. After calculation we obtain

$$\frac{\Delta\rho_C}{\rho_C^b} = \frac{3\lambda}{4(d-s')}\sin^{-1}\left(1-\frac{s'}{d}\right) \tag{A3}$$

and Eq. (3) in Sec. II A. Using Eqs. (8), (9), and (12) we find at $T_i = 293$ K, $R_C^b(T_i) = 52.82\ \Omega$, $R_C^L = 10.28\ \Omega$ and $R_C(T_i) = 63.10\ \Omega$. In this calculation we have used: $a = 1.75$ nm and $s' = 0.5218$ nm estimated according to Fig. 9 and the same values of the remaining quantities as in Sec. IV.

Similar calculations have also been performed for the constriction of the form given in Fig. 3 with the same final equation for the resistivity increment as Eq. (A3) but with s' replaced by $s = 0.5$ nm. Using Eqs. (8), (9), and (12) with s' and a replaced by s (as above) and $a' = 1.714$ nm, respectively, and with the same values of the remaining quantities we have found at $T_i = 293$ K, $R_{C1}^b(T_i) = 54.13\ \Omega$, $R_{C1}^L = 10.57\ \Omega$, and $R_{C1}(T_i) = 64.69\ \Omega$.

The presented calculation shows that the difference between the values of the parameters describing the temperature dependencies of R_C and R_{C1} are relatively small. In

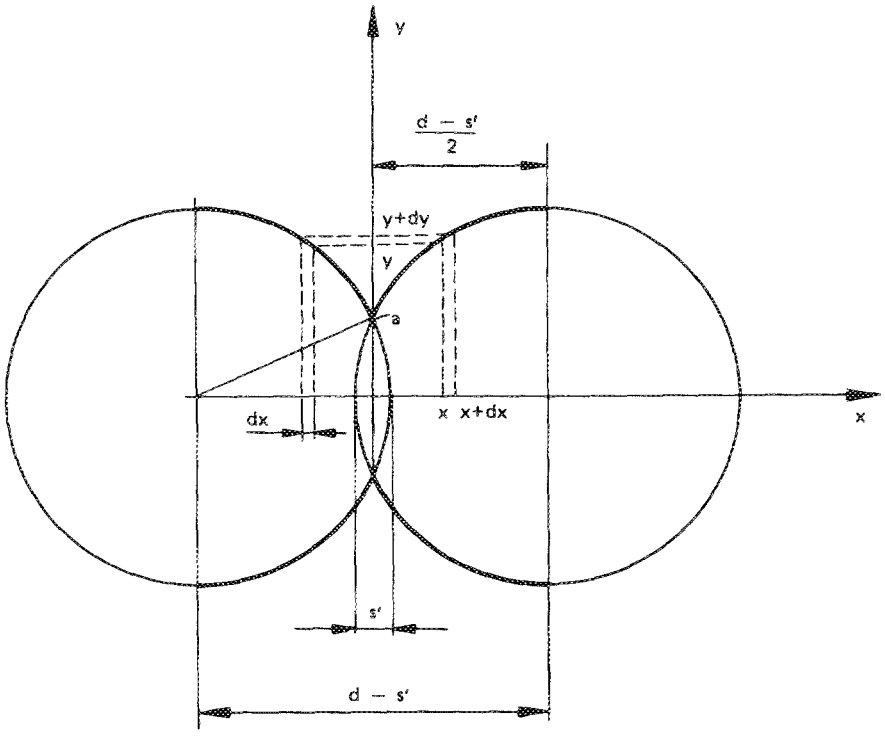

FIG. 9. The geometry of the simplified constriction model utilized in the estimation of the surface scattering dependent increment of the constriction resistivity ρ_C.

the network we have exactly $N_C - N_B$ constrictions of the type given in Fig. 1, described by Eqs. (11)–(13); there are also N_B constrictions of the form shown in Fig. 3, described by Eqs. (11)–(13) with $R^{b}_{C1}(T_i)$, R^{L}_{C1}, $R_{C1}(T_i)$, and $\alpha_{C1}(T_i)$ introduced instead of the respective quantities without index "1." Since $N_B \ll N_C$, the contribution of the "barrier type constriction" $[R_{C1}(T)]$ is strongly attenuated in the resultant characteristic of the network. Therefore, in the analysis we have considered only one type of temperature dependence of constriction resistance $R_C(T)$ given by Eqs. (11)–(13) with $R^{b}_{C}(T_i)$, R^{L}_{C}, $R_C(T_i)$, and $\alpha_C(T_i)$ calculated according to Figs. 1 and 9. This approximation has negligibly changed the estimated values of the network parameters; however, it has substantially simplified the parameter calculation.

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