SUPERIORITY OF THE CK1 FITTING MODEL FOR DISPERSED CONDUCTIVE SYSTEMS

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Here, some reasons for such superiority are briefly summarized, with reference numbers to published papers, available for downloading in pdf format, that provide details and proof of the various assertions. The reference numbers are those in the Serially Ordered list of my publications appearing in my web site: http://jrossmacdonald.com.

1. The CK1 and its simplified, nearly universal version, the CUN, have been used to fit many experimental data sets for ionic glasses, polycrystalline materials, single crystals, and supercooled liquids, such as CKN. Such fits are generally superior to those using other models [e.g., 226,229,235,237,239,248]. Estimates of the values of the characteristic shape parameter of the CK1 model, β_1, are particularly close to 1/3 for disordered and single-crystal materials when a single species of charge is mobile.

2. Unlike the widely used but flawed [232,248] original modulus formalism (OMF) K1 model of Mohnihan, Ngai, and others, the composite CK1 model separates conductive-system and dielectric-system effects by including, along with the conductive-system K1 model, a separate parallel capacitance parameter represented by C that models the bulk dipolar dielectric constant of the material, $\varepsilon_{D\infty}$, one independent of frequency in the usual experimental range. Then an effective dielectric constant arising solely from mobile charge, $\varepsilon_{C1\infty}$, also appears, and the total high-frequency dielectric constant is given by $\varepsilon_{\infty} = \varepsilon_{C1\infty} + \varepsilon_{D\infty}$, although the response of the first term begins to decrease at sufficiently high frequencies before the second one begins to approach n^2.

3. The K1 model for fitting and analyzing dispersed frequency response conductive-system data is the only one independently derived by both macroscopic and

microscopic theoretical analyses. The general microscopic model follows from a continuous-time, random-walk hopping model [229], and becomes the K1 when its temporal correlation factor is of stretched exponential form. Thus, the model is indirectly derived from the assumption of stretched-exponential temporal response, itself a paradigmatic result of many different theoretical approaches [208,212, 229,235,239].

4. The CK1 model nearly always yields appreciably better fits of full complex data than other models such as the CK0 one or the Universal Dynamic Response power-law one often ascribed to Jonscher [221]. The K0 model is just the direct one-sided Fourier transform of stretched exponential temporal response to the frequency domain. It may be used to calculate K1 response but more accuracy is achieved if such calculation involves the pertinent K1 distribution of relaxation times [212,239]. Accurate calculation of K0 and K1 response for any pertinent β_1 value is a feature of the LEVM/LEVMW complex nonlinear least squares fitting and inversion computer program [179,220], freely available for downloading from the above web site.

5. The K1 is the only model for which values of its shape parameter, β_1, have been derived from constraint theory and depend on the effective dimensionality of the space in which mobile charges move [233,235,249]. For 3D motion, its value is 1/3, and estimates from data fitting for a large number of materials lead to close approximations to this value, defining the CUN, a semi-universal K1 model with β_1 fixed at 1/3. Further, in agreement with its derivation, such β_1 values are found to be independent of exogenous variables such as temperature and ionic concentration, a remarkable property.

6. When the CUN model provides as good or better fit of data than the CK1 one, the conductive-system part of the model involves only two free parameters: a scale parameter ρ_0, the dc resistivity, and a placement parameter, τ_0, the characteristic

relaxation time of the K1 model with its value of β_1 fixed at 1/3. Then when any electrode and/or nearly-constant-loss effects are absent or have been removed from the data, plots of $\sigma'(\omega)/\sigma_0 \equiv \rho_0\sigma'(\omega)$ *vs* $\omega\tau_0$ for different materials and different exogenous conditions will all be the same, obviating the need for any other kind of scaling. For the UN model itself, $\rho(\omega)/\rho_0$ *vs* $\omega\tau_0$ response is independent of ρ_0 and τ_0 values, as are the results of transformation of such response to any of the other immittance levels.

7. Although the CK1 model and the OMF K1 one both use the K1 model, data fitting with the OMF leads to variation of its β_1 value with temperature and ionic concentration, interpreted by Ngai as implying variable ion-ion correlation. But this approach and such a conclusion are invalid for both theoretical and experimental reasons [232,239]].

8. Both the CK0 model with its β_0 shape parameter fixed at 2/3 and the K1 model with $\beta_1=1/3$ lead to a limiting-high-frequency power-law exponent or log-log slope of the real part of the conductivity, $\sigma'(\omega)$, of 2/3, but their approaches to this limiting value differ appreciably [235,243]. Although many power-law fits of experimental data at high frequencies have led to slope values very close to 2/3, the slope is often larger and may be close to unity or even greater [227], requiring the presence in the composite fitting model of an independent nearly-constant-loss process or of an electrode-polarization model that leads to high- as well as low-frequency effects [225,226,228,237,249].

9. A great many data sets for different conductive-system materials have been found to satisfy the empirical Barton, Nakajima, Namikawa (BNN) relation. The UN model leads to a value of the BNN parameter p of about 1.65, close to most observed values [234,235,251] and thus provides a theoretical justification for the empirical relation.