On two incompatible models for dispersion in ionic conductors

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The two models considered are the widely used 1973 original modulus formalism (OMF) of Moynihan and associates, and the later corrected modulus formalism (CMF). Both approaches involve a dispersive frequency-response model derived from Kohlrausch stretched-exponential temporal response, the KWW1 model, also termed the K1. A brief summary of the derivation of this model is followed by consideration of the small but crucial differences between OMF and CMF analysis approaches and the reasons why the OMF and an inferred physical basis for its behavior, variable correlation between mobile ions, are inappropriate. After discussions of some prior criticisms of the OMF approach, results of accurate least-squares fitting of experimental frequency-response data to OMF and CMF models for a variety of ionic materials illustrate these differences and demonstrate a crucial inconsistency of the OMF, one that critically falsifies it. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1636832]

I. INTRODUCTION

Conductive-system dispersive response involving mobile charge may be conceptually associated with the effects of three processes. These are (1) electrode effects that are particularly important at low frequencies[1] but may not be negligible at very high ones;[2,3] (2) ionic hopping effects, usually significant at mid-range frequencies;[2–4] and (3) nearly constant loss effects primarily evident at sufficiently low temperatures over the usual frequency range or at high frequencies for higher temperatures.[5–9]

Three different kinds of models have been proposed for describing these responses. A useful summary of some of the pertinent history of attempts to characterize the situation appears in Ref. 6. We shall consider here only models for the above behaviors that may be associated with mobile charge effects. The first and most desirable would be a fully microscopic model that accounted for all the above processes, since they are all associated with thermally activated mobile charge in conductive-system materials. Unfortunately, this many-body problem involving all interactions is currently insoluble. A detailed continuum approach including Coulomb interactions and electrode reactions[10] does not include either distributed-process effects or lead to nearly constant loss and so disagrees with most experimental results for solid materials.

A second approach involves semimicroscopic models whose $\log$–$\log \sigma'(\omega)$ conductivity slope continuously increases toward a value of unity until a plateau is reached, and so involve a kind of nearly constant loss at sufficiently high frequencies.[11–13] No account of electrode effects is included in these approaches. Unfortunately, although apparent theoretical defects inherent in the mismatch-and-relaxation model of Funke and associates[11] have been pointed out,[13] they have neither been explicitly recognized nor directly re-

solved. A recent empirical modification of this approach[12] seems, however, to avoid some of its defects. The semi-microscopic symmetric hopping model of Dyre and Schrøder[14] ignores Coulomb interactions and yields response similar to that of the mismatch-and-relaxation model,[6,11] but leads to nonphysical low-frequency-limiting response.[15] For both of these models, their mathematical complexity makes fitting and the estimation of values of model parameters difficult, and thus no complex-nonlinear least squares (CNLS) fitting of data to estimate their parameters seems to have been published so far.

The third approach[2–9,16–24] involves a composite model involving separate parts: one accounting for ionic hopping; a parallel contribution representing the effect of the endemic bulk dielectric constant, $\epsilon_{D\infty}$; possibly a part describing nearly constant loss; and finally a series response model to account for electrode effects. For fitting most limited-range data, only two or three of these parts are usually required and excellent data fits are usually found using appropriate models. Here we will deal with data that do not extend to high enough frequencies or low enough temperatures to require a nearly constant loss contribution.

Why does it matter that there appear to be two incompatible hopping models for fitting and representing the dispersive frequency response of ionic conductors? It matters because they both cannot be correct, yet most analysis is still carried out using the theoretically incorrect and experimentally inconsistent one, while the other approach is ignored. The two models considered here are the 1973 original modulus formalism (OMF) of Moynihan and associates,[25,26] and the corrected modulus formalism (CMF) first discussed in 1994 and 1996.[27,28] To the list of 19 OMF references cited by Ngai and León in 1999,[29] a few more recent representative ones are those of Refs. 5, 8, 23, 24, and 30–32.

Why has the OMF continued to be used up to the present time without significant challenges to the CMF? One might speculate that the reason is that although it apparently cannot

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be shown that the CMF model and its physical interpretation are inappropriate, replacing the OMF by the CMF would eliminate the main experimental support for the proposed physical basis of the OMF: variable ionic coupling (e.g., Refs. 24, 33–36). To do so would, however, call into question all the OMF data interpretations of the last 30 years based on the assumption of such variable coupling. Perhaps it is thus understandable that such a change has, so far, been unpalatable.

In Sec. II I define and compare some predictions of the two incompatible models. In Sec. III some prior challenges to the OMF approach are discussed and compared with earlier and later CMF analyses, all dealing with high-frequency-limiting dielectric quantities and their effects. Section IV summarizes and illustrates a crucial experimental falsification of the OMF, and Section V lists important conclusions.

II. ANTITHESES

It seems paradoxical that although the OMF and CMF are both conductive-system dispersive-response models, they are strongly antithetical and differ greatly in their physical interpretations, yet their analytical forms are only slightly different. A derivation of the OMF begins with the assumption of a stretched-exponential temporal response for an electric field decay function, $\phi_0(t)$,[26]

$$\phi_0(t) = \exp[-(t/\tau_o)^{\beta_0}], \quad 0 < \beta_0 \le 1. \tag{1}$$

Its single-sided Fourier transform leads to a frequency-response model called the KWW0 or K0,[2,4,37–40] where a $k = 0$ or 1 index has been set to 0 for β_k and Kk.

The K0 high-frequency-limiting dielectric response associated entirely with mobile ions, $\epsilon_{C0}(\infty) \equiv \epsilon_{C0\infty}$, is identically zero, but this is not the case for the K1 model,[21,22,27,28,40] derived directly from the K0 model.[4,26,28,40,41] It is also significant that Fourier transformation of K1-model frequency response to the time domain *does not* lead to stretched-exponential behavior,[38,40] a fact not mentioned in OMF analyses. Although both the OMF and the CMF involve the K1 model, they formally differ only in their identification of its high-frequency-limiting dielectric response, $\epsilon_{C1}(\infty)$, and the consequences of that identification. Users of the OMF express this quantity as

$$\epsilon_\infty = \sigma_0 \langle \tau \rangle_{01}/\epsilon_V = \epsilon_{Ma} \langle x \rangle_{01} = \epsilon_{Ma} \beta_{1O}^{-1} \Gamma(\beta_{1O}^{-1}), \tag{2}$$

where we identify the OMF β_1 as β_{1O} to distinguish it from that of the CMF, β_{1C}. Here the Maxwell quantity ϵ_{Ma} is

$$\epsilon_{Ma} \equiv \sigma_0 \tau_o / \epsilon_V; \tag{3}$$

$x \equiv \tau/\tau_o$; τ_o is the characteristic relaxation time of the K0 or K1, as in Eq. (1); ϵ_V is the permittivity of vacuum; $\Gamma(\ )$ is the Euler gamma function, and β_{1O} is the fractional exponent associated with K1, quite different from β_0. In Eq. (2), the 01 subscript indicates that $\langle x \rangle_{01}$ is the normalized mean of x over the K0 distribution of relaxation times involving β_{1O} rather than β_0.[2,4,40]

For the CMF, on the other hand, $\epsilon_{C1}(\infty) \equiv \epsilon_{C1\infty}$, and it may be expressed, using the K1 dispersion model, as[2–4,22,40]

$$
\begin{aligned}
\epsilon_{C1\infty} &= \epsilon_{Ma}/\langle x^{-1} \rangle_1 \\
&= \epsilon_{Ma} \langle x \rangle_{01} \\
&= \epsilon_{Ma} \beta_{1C}^{-1} \Gamma(\beta_{1C}^{-1}) \\
&= [\gamma N(qd)^2/(6k_B \epsilon_V)]/T = A/T, \tag{4}
\end{aligned}
$$

where N is the maximum mobile charge number density; γ is the fraction of charge carriers of charge q that are mobile; and d is the rms single-hop distance for the hopping entity.

The term involving N in Eq. (4), not included in the OMF, follows[22,40] from the important microscopic stochastic transport analysis of Scher and Lax,[42] a continuous-time, random-walk hopping model. Since this model and the K1 have been shown to be fully isomorphic in form,[22,40] the K1 response is consistent with both from the original macroscopic model of Ref. 26 and with the microscopic analysis of Ref. 42. Therefore, the N term in Eq. (4) provides a microscopic physical interpretation of $\epsilon_{C1\infty}$, along with the macroscopic one involving ϵ_{Ma} in this equation. The part involving the gamma function is only appropriate in the absence of cutoff of the K1 distribution of relaxation times and so does not apply for the cutoff dispersion model.[38]

We expect that, as usual, the quantities in the square brackets of Eq. (4) are temperature independent, so the fitting parameter A is itself independent of temperature.[2] Then it follows that in the usual case where τ_o is thermally activated, $T\sigma_0$ is activated with the same activation energy in the absence of cutoff, a standard result but one not accepted by all practitioners in the present field. Although the N term of Eq. (4) shows that as the ionic concentration approaches zero, $\epsilon_{C1\infty} \to 0$, requiring that $\epsilon_{Ma} \to 0$ as well, in accordance with CMF fit results,[2,15] the situation is different for the OMF expression of Eq. (2). In this case, OMF fits show that both ϵ_∞ and ϵ_{Ma} approach the same constant value, one later identified herein as $\epsilon_{D\infty}$, the bulk dielectric constant of the material.

Because $\epsilon_{C1\infty}$ is a pure conductive-system quantity arising solely from charge motion, the CMF approach must include a free-fitting parameter, ϵ_x, to account for the effect of $\epsilon_{D\infty}$, present in all experimental data. The resulting CMF composite model is designated the CK1. For the CK1 the total high-frequency-limiting dielectric constant is $\epsilon_\infty = \epsilon_{C1\infty} + \epsilon_{D\infty}$. The fitting of data for a variety of materials using the CK1 leads to $\beta_{1C} \simeq 1/3$ estimates, substantially independent of temperature and ionic concentration.[2,4]

III. LIMITING DIELECTRIC QUANTITIES AND OMF CRITICISMS

Because the separate existence of $\epsilon_{C1\infty}$ is only acknowledged by those such as the present author who make a distinction between the OMF interpretation of the dispersive response and that of the CMF, the ϵ_∞ quantity appearing in OMF and in other fitting models must be interpreted as $\epsilon_{D\infty}$, a result consistent with the usual non-CMF definitions of this quantity (e.g., Refs. 25, 26, 34, 43). Thus, in the OMF and CMF *models*, the interpretation of ϵ_∞ is significantly different. But experimental data always involve the effects of a nonzero $\epsilon_{D\infty}$. Therefore, an analysis of such data with either

approach leads to estimates of ϵ_∞ that are identified as $\epsilon_{D\infty}$ in the OMF situation and as $\epsilon_{C1\infty} + \epsilon_{D\infty}$ in the CMF one. Clearly, when a K1 fitting model is appropriate, the second expression is the appropriate one.

Experimental data fits show, in fact, that the CMF CK1 fitting is superior to OMF K1 fitting,[2,4,15] and the quantity A in Eq. (4) is usually temperature independent, allowing a consistent discrimination between the estimation of both $\epsilon_{C1\infty}$ and $\epsilon_{D\infty}$. To distinguish between the $\epsilon_\infty(\equiv\epsilon_{D\infty})$ quantity that appears in all models but the CMF and an estimate of ϵ_∞ obtained from fitting experimental data, I shall hereafter denote the former by $[\epsilon_\infty]$, while ϵ_∞ itself will refer to both the CMF CK1-model quantity and to experimental estimates of the total high-frequency-limiting dielectric constant.

In a recent summary dealing with conflicting points of view concerning a dispersive response, Ngai and co-authors[44] made the wise statement, "...a proper approach to the dynamics of ions in glasses, crystals and melts is still a matter of genuine scientific debate. We need to emphasize the experimental facts that critically falsify a model... ." One would think that a genuine scientific debate would be one that recognized and discussed all plausible criticisms of a particular model such as the OMF, including those appearing in earlier publications on the CMF, but no such work is explicitly mentioned in the attempted justification for the use of $\epsilon_\infty = [\epsilon_\infty]$ in the present OMF, Eq. (2).[24,44] Instead, in Ref. 24 Ngai and co-authors merely mention "misguided attacks on the use of the electric modulus representation of data." This debatable statement is itself a misnomer since it is not the representation of *data* at the electric modulus level that is the issue but instead the use of the electric modulus *formalism*, an approach that fits data expressed only in a $M''(\omega)$ form. There is no such limitation for CMF model fitting since experimental data may be expressed in terms of any one of the four immittance spectroscopy levels and any model may be used to fit data at any of these levels.[13]

The gist of the Ngai defense of the presence of $[\epsilon_\infty]$ in the OMF is a comparison of this quantity with its "exact analog," the high-frequency limit, G_∞, of the rheological complex dynamic modulus $G(\omega)$.[24,44] Since a detailed rebuttal of this defense appears in Ref. 45, it need only be summarized here. The above comparison is invalid because it compares two disparate quantities, G_∞ and $[\epsilon_\infty]$, rather than the proper quantities G_∞ and $\epsilon_{C1\infty}$. Here, as usual, G_∞ is taken as a model quantity associated with mobile defects, just as a nonzero $\epsilon_{C1\infty}$ arises solely from mobile charges in the K1 model.

The following discussion provides some historical background concerning criticisms of the OMF. In 1991, Dyre[46] made the important point that experimental $M''(\omega)$ data always include effects arising from the presence of $[\epsilon_\infty]$, and that therefore such data should not be fitted by models that involve only mobile-charge effects. Next, in 1994 Boukamp and Macdonald[27] first introduced the $\epsilon_{C1\infty}$ quantity, independently reiterated Dyre's limitation, and explicitly showed why CMF fitting should be used in place of $M''(\omega)$ OMF fitting.

Then in a valuable paper published in 1995, Sidebottom, Green, and Brow[47] compared fit results for a wide variety of materials using the OMF as well as a power-law model[21] (involving the log–log slope exponent n) for $\sigma'(\omega)$ data. They found β_{1O} values of the order of 0.58 and $\beta_n \equiv 1-n$ estimates of 0.33 ± 0.05. This result is in full agreement with β_{1C} estimates obtained from later CMF CK1 fits carried out at any immittance level.[2,4,37] In 1996, explicit differences between the OMF and CMF approaches were illustrated in detail,[28] and many subsequent publications involving CMF and OMF fit comparisons have strengthened the case against the OMF.

In 1998, Moynihan attempted in Ref. 48 to summarize and justify criticisms of the "use of the electrical modulus...for data analysis." Again, the OMF itself was meant, but no reference was made to the various earlier papers dealing with the CMF [e.g., Refs. 19, 27, 28, 37]. He explained the large differences between OMF and power-law results by apparently suggesting that $M''(\omega)$ and $\sigma'(\omega)$ data might involve some different microscopic processes and that therefore no single theory might apply to both. As discussed in the next section, the CMF does so. Moynihan also listed as one of the main advantages of the OMF for ionic conductors that "it suppresses...low frequency electrode impedance effects." In fact, plotting of data in $M''(\omega)$ form does hide, but does not eliminate, electrode polarization effects, and fitting models that include a part to account for electrode effects can yield exactly the same estimates of the electrode parameters whether the fitting is carried out for data at the $M''(\omega)$ level or at any of the other immittance levels.[13]

Finally, it is worth mentioning that in 2001 Sidebottom, Roling, and Funke[43] illustrated Dyre's[46] criticism that the shape of $M''(\omega)$ is sensitive to the value of $[\epsilon_\infty]$ and showed, for example, that synthetic $M''(\omega)$ data without any contribution from ϵ_∞ no longer involves a peak but increases indefinitely as the frequency increases. These authors ignored the work of the present author that demonstrates the virtues of replacing the OMF by the CMF. Examples of the difference between their criticisms of the OMF, which, as usual, do not acknowledge the existence of $\epsilon_{C1\infty}$, and CMF results are provided by Fig. 2 of Ref. 4 and Fig. 3 of Ref. 49. There, subtraction of the effects of $\epsilon_{D\infty}$, *not* ϵ_∞, from a CK1 fit of experimental data led to a pure K1 $M''(\omega)$ response with much higher peaks than those of the original data, but not to an indefinite increase in $M''(\omega)$. The resulting peaks are associated with a nonzero value of $\epsilon_{C1\infty}$, an appreciably smaller value than that of ϵ_∞ for the datasets considered. The main conclusion of these authors that the use of the OMF "should be discouraged," should be modified to the following: *the OMF should be replaced by the CMF, at least until a more appropriate model than the CMF becomes available.*

IV. EXPERIMENTAL FALSIFICATION OF THE OMF

It is noteworthy that although there have been many criticisms of the OMF approach over the years, as discussed in part above, only those concerned with the CMF show how the general OMF approach should be reinterpreted and augmented to yield a viable model for dispersive data fitting and

TABLE I. A comparison of OMF and CMF $M''(\omega)$- and $\sigma'(\omega)$-level β_1 and CKO $[\beta_0]$ fit estimates.

Material/(Ref.)	T (K)	x_c	OMF-M'' [CK0-M'']	OMF/CMF-σ' [CK0-σ']
Na$_2$O·3SiO$_2$ (49)	273	⋯	0.47	0.33
Li$_2$O·Al$_2$O$_3$·2SiO$_2$ (39)	297	⋯	0.46	0.33
0.02K$_2$O·0.98GeO$_2$ (15)	602	0.02	0.96	0.32
0.2K$_2$O·0.8GeO$_2$ (15)	414	0.2	0.52	0.30
			[0.54]	
0.88ZrO$_2$·0.12Y$_2$O$_3$ (4)	503	0.12	0.55	1/3 fixed
			[0.52]	[0.54]
0.88ZrO$_2$·0.12Y$_2$O$_3$ (2)	503	0.12	0.51	0.32

interpretation. CMF analysis has usually involved full CNLS simultaneous fitting of both parts of complex data, but nearly all other dispersive data analyses have dealt only with either $M''(\omega)$ or $\sigma'(\omega)$, but not with full $M(\omega)$ or $\sigma(\omega)$ complex-response data. Part of the reason may have been that no closed form is available for either a K0 or K1 response for arbitrary β. Since 1997, however, the free LEVM CNLS computer program[50] has included accurate algorithms for calculating these functions, allowing both the generation of synthetic data and the accurate fitting of experimental data.

An explanation of why even simple power law fitting of $\sigma'(\omega)$ data has yielded more reasonable log–log slope parameter estimates than has the OMF is that $\sigma'(\omega)$ and $\epsilon''(\omega)$ are the only ones of the eight real and imaginary parts of the four immittance functions that do not involve separate capacitative effects associated with the bulk dielectric quantity $\epsilon_{D\infty}$. Therefore, the presence and magnitude of $[\epsilon_\infty] \equiv \epsilon_{D\infty}$ may strongly affect the shape and magnitude of $M''(\omega)$ data but not that of the associated $\sigma'(\omega)$ response.

It is one thing to state that one should emphasize the experimental facts that critically falsify a model and an entirely different matter to actually do so. The above considerations provide a definitive test for doing this, however. Note that there is no formal difference between the OMF K1 model and the CMF CK1 one for fits of $\sigma'(\omega)$, ones carried out with the CK1 free parameter ϵ_x taken either fixed or zero. Therefore, OMF and CMF fits of $\sigma'(\omega)$ data should and do lead to the same parameter estimates since they both involve fits of just the K1 model. Although it has long been known that OMF fits of the same data in $M''(\omega)$ form yield quite different and inconsistent results from those of $\sigma'(\omega)$ fits, it has only relatively recently been emphasized that since CMF fits of $M''(\omega)$ data or of any of the other seven individual immittance parts or of complex data expressed in any of the four immittance levels are all consistent and yield the same, or nearly the same, parameter estimates as those for $\sigma'(\omega)$ fits, such results critically falsify the OMF model and all its results and interpretations.

Some results of such comparisons for several different materials, temperatures, and ionic concentrations, are presented in Table I. Note that the OMF/CMF results of column five are not a part of ordinary OMF fitting. Most of the present OMF and CMF fits were carried out with composite models that included a part to account for electrode polarization effects.[2,4,19,49] More information is available in the references cited in the table.

The large differences between the $(\beta_1)_{\text{OMF}} \equiv \beta_{1O}$ values of column four and the $(\beta_1)_{\text{CMF}} \equiv \beta_{1C}$ ones of column five are evident. No values for CMF fits of data in $M''(\omega)$ or other immittance forms were included because they were generally negligibly different from those shown in column five. A synthetic CMF dataset with β_{1C} fixed at 1/3 was used for the OMF and CK0 fittings of Ref. 4, row five of the table. As expected, like the CMF results, little differences appear for CK0 fits carried out with data expressed in different forms. A comparison of the Ref. 4 and Ref. 2 results for the same material demonstrates the effect of keeping β_{1C} fixed at 1/3 and of leaving it free to vary when fitting the original experimental data. The present results and many others not included here clearly indicate that CMF fits are consistent and OMF ones are not.

CMF fits at any immittance level usually lead to estimates of $\beta_{1C} \approx 1/3$ that are nearly independent of temperature and of relative mobile-ion concentration, x_c,[2,4] but OMF $M''(\omega)$ fits yield β_{1O} estimates that are much larger and that approach unity as $x_c \to 0$. The CMF macro- and microscopic model involves no overt Coulomb interactions, consistent with its nearly constant value of β_{1C}. The above variation of β_{1O}, however, has been interpreted as arising from the decreasing correlation between ions as their separation increases. We see that this apparently plausible explanation is unsupported by the CMF model and by the experimental constancy of β_{1C} over appreciable temperature and concentration ranges. Since the OMF approach is neither theoretically nor experimentally consistent, one must conclude that the Ngai variable-correlation physical interpretation of the basis of the OMF is unsupported. It is also perhaps significant that Li ions intercalated into β-Ta$_2$O$_5$ "tended to distribute uniformly, rather than to attract each other and form clusters... ."[51]

V. SUMMARY

The OMF and the CMF approaches are antithetical because the CMF takes proper account of $\epsilon_{D\infty}$ and the OMF does not. They are antithetical because the OMF fitting of experimental data leads to disparate and inconsistent estimates of β_{1O} from fitting of the data in $M''(\omega)$ and in $\sigma'(\omega)$ forms, while such CMF fitting leads to very nearly equal and fully consistent estimates of β_{1C} close to 1/3.

Further, CMF complex nonlinear least squares fits of experimental data are always appreciably better than are OMF ones, and CMF fits, unlike OMF ones, yield estimates of K1-model parameters that all represent a pure conductive-system response. The K1 model is supported by both macroscopic and microscopic analyses, and the CMF approach, using K1, leads to results that fail to justify the common physical interpretation of OMF fitting results, one involving ion–ion correlations that decrease as the ionic concentration decreases. For all these reasons, the OMF model should be replaced by the CMF one.

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