

# Communication

## Evaluation of Matusita Equation and Its Modified Expression for Determining Activation Energy Associated with Melt Crystallization

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Both the Matusita equation and the modified Matusita equation for estimating the activation energy associated with non-isothermal crystallization were critically evaluated. The derivation for melts crystallization on cooling indicates that, unlike for the crystallization that occurs on heating, the term  $1 - \exp(-\Delta G/RT)$  in the basic rate equation of crystal growth and the term  $\int_0^{T_s} \exp(-E/RT)dT$  depending on the initial temperature of the cooling process cannot be neglected. It is demonstrated that both the Matusita equation and its modified expression are only valid to estimate the activation energy associated with the crystallization that occurs on heating, but are inapplicable for the melt crystallization that occurs on cooling. It is suggested that the isoconversional methods of Friedman and Vyazovkin should be alternative to determine effective activation energy for melt crystallization that occurs on cooling.

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Various models have been developed to estimate the activation energy associated with the non-isothermal crystallization from thermal analysis data, including Kissinger equation,<sup>[1]</sup> Ozawa equation,<sup>[2]</sup> modified Ozawa-Chen equation,<sup>[3]</sup> and Matusita equations.<sup>[4,5]</sup> Among these models, Kissinger equation<sup>[1]</sup> and Matusita equations<sup>[4,5]</sup> are the most widely used approaches to determine the activation energy for the crystallization that occurs on heating.<sup>[6–10]</sup> Moreover, they are applied frequently for the non-isothermal crystallization of

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polymer melts<sup>[11–15]</sup> and metallurgical slags<sup>[16,17]</sup> that occurs on cooling.

Matusita and Sakka<sup>[4]</sup> emphasized that the physical meaning of the crystallization activation energy determined by the Kissinger equation<sup>[1]</sup> is obscure because the crystallization of glass is advanced by both nucleation and crystal growth, rather than an  $n$ -th order reaction. Therefore, they proposed the following generalized expression to determine the activation energy for crystal growth:<sup>[4]</sup>

$$\ln\left(\frac{\beta^n}{T_p^2}\right) = -\frac{mE}{RT_p} + \text{constant} \quad [1]$$

where  $\beta$  is the heating rate,  $E$  is the activation energy for crystal growth,  $R$  is the ideal gas constant,  $T_p$  is the crystallization peak temperature,  $n$  and  $m$  are the numerical factors that depend on the crystallization mechanism and summarized in Reference 18.

A few years later, Matusita *et al.*<sup>[5]</sup> derived a more precise equation (*i.e.*, Eq. [2]) than Eq. [1] by improving the accuracy of mathematical approximation in deriving the generalized expression to estimate the activation energy for crystal growth

$$\ln[-\ln(1-x)] = -n \ln \beta - 1.052 \frac{mE}{RT} + \text{constant} \quad [2]$$

where  $x$  is the volume fraction of crystallized phase at a given temperature  $T$ . In general, both Eq. [1] and Eq. [2] are called Matusita equation. To facilitate the evaluation of Matusita models in this article, Eqs. [1] and [2] are hereinafter designated as the Matusita equation and the modified Matusita equation, respectively.

Since the crystallization mechanism is involved in Matusita equation and its modified expression, these two models have been widely used to determine the activation energy for crystal growth. It should be stressed that both the Matusita equation and the modified Matusita equation were originally derived from the expressions for the crystallization that occurs on heating. However, many practical processes of non-isothermal crystallization proceed on cooling, such as some kinds of polymer melts, and mold fluxes in continuous casting of steel. Therefore, it is highly needed to ascertain that whether the Matusita equation and the modified Matusita equation are applicable in estimating the activation energy for melts crystallization.

In a series of papers,<sup>[4,5,19–21]</sup> Matusita and co-workers made a detailed derivation for their models, *i.e.*, Eqs. [1] and [2]. Matusita equation was derived from the basic equation of crystal growth rate<sup>[19]</sup>

$$U = \frac{dr}{dt} = \beta \frac{dr}{dT} = U_0 \exp(-E/RT) [1 - \exp(-\Delta G/RT)] \quad [3]$$

where  $U_0$  is the pre-exponential factor,  $\Delta G$  is the crystallization free energy, and  $r$  is the radius of a crystal particle. If the temperature is much higher than that of maximum growth rate in the heating process, then the

temperature dependence of the term  $1 - \exp(-\Delta G/RT)$  is negligibly small compared with that of the term  $\exp(-E/RT)$ .<sup>[19]</sup> Thus, Eq. [3] can be rewritten as

$$U = U_0 \exp(-E/RT) \quad [4]$$

During the heating of a glass from room temperature  $T_r$  to a certain temperature  $T$ , the nucleation and crystal growth will occur successively. The radius  $r$  of crystal particle can be calculated by substituting Eq. [4] into Eq. [3].

$$r = \int_{T_r}^T \frac{U}{\beta} dT = \frac{U_0}{\beta} \left[ \int_0^T \exp(-E/RT) dT - \int_0^{T_r} \exp(-E/RT) dT \right] \quad [5]$$

Because the upper limit of the integral  $\int_0^{T_r} \exp(-E/RT) dT$  in Eq. [5] is much smaller than that of the term  $\int_0^T \exp(-E/RT) dT$  during glass crystallization, the term  $\int_0^{T_r} \exp(-E/RT) dT$  is negligibly small compared with the term  $\int_0^T \exp(-E/RT) dT$ . Therefore, Eq. [5] can be simplified as the following equation.

$$r = \frac{U_0}{\beta} \int_0^T \exp(-E/RT) dT \quad [6]$$

Because the integral term  $\int_0^T \exp(-E/RT) dT$  in Eq. [6] cannot be expressed by compounds of elementary functions, a rough approximation is made to obtain the Matusita equation, and then Eq. [6] is approximated to

$$r = \frac{U_0}{\beta} \int_0^T \exp(-E/RT) dT \cong \frac{r_0}{a} \exp(-E/RT) \quad [7]$$

In the derivation by Matusita and Sakka,<sup>[19]</sup> the case where the crystal growth was three dimensional and controlled by interface reaction was taken as an example to derive the generalized Matusita equation. In this case, the crystallization rate  $\frac{dx}{dt}$  is expressed as

$$\frac{dx}{dt} = (1-x)N4\pi r^2 \frac{dr}{dt} \quad [8]$$

where  $N$  is the number of nuclei formed per unit volume and  $x$  is the relative degree of crystallinity.

Inserting Eqs. [3], [4], and [7] into [8], the following equation can be obtained.

$$\frac{dx}{dt} = 4\pi NU_0(1-x) \frac{r_0^2}{\beta^2} \exp\left(-\frac{3E}{RT}\right) \quad [9]$$

In a more generalized expression, Eq. [9] is expressed as

$$\frac{dx}{dt} = A\beta^{-(n-1)}(1-x)^k \exp\left(\frac{-mE}{RT}\right) \quad [10]$$

where  $k = 1$  for bulk nucleation and  $k = \frac{2}{3}$  for surface nucleation.

The crystallization rate reaches the maximum at temperature  $T_p$  where the second derivative of the relative degree of crystallinity  $x$  equals zero.

$$\frac{d}{dt} \left( \frac{dx}{dt} \right) = 0 \quad [11]$$

By solving equation set of Eqs. [10] and [11], the following equation can be derived.

$$\frac{\beta^n}{T_p^2} = \frac{AkR}{mE} (1-x)^{k-1} \exp\left(-\frac{mE}{RT_p}\right) \quad [12]$$

Taking the logarithm of Eq. [12], Matusita equation can be derived as Eq. [1]. The activation energy  $E$  can be evaluated from the slope of the linear fitted plot of  $\ln(\beta^n/T_p^2)$  vs  $1/T_p$  from Eq. [1].

In a few years' time, Matusita *et al.*<sup>[5]</sup> applied a closer approximation by employing Doyle's  $p$ -function<sup>[22]</sup> to integrate the term  $\int_0^T \exp(-E/RT) dT$  in Eq. [6]. As a consequence, Eq. [6] was approximated to Eq. [13]

$$r = \frac{U_0 E}{\beta R} \exp\left(-5.330 - 1.052 \frac{E}{RT}\right) \quad [13]$$

Taking the integration of Eq. [8], the following equation is obtained.

$$-\ln(1-x) = \frac{4}{3} \pi N r^3 \quad [14]$$

For a heating process, the radius of a crystal particle  $r$  in Eq. [14] was replaced by Eq. [13]. It gives

$$\begin{aligned} -\ln(1-x) &= \frac{4}{3} \pi N \left( \frac{U_0 E}{\beta R} \right)^3 \\ &\times \exp\left(-3 \times 5.330 - 3 \times 1.052 \frac{E}{RT}\right) \\ &+ \text{constant} \end{aligned} \quad [15]$$

In a more generalized expression, Eq. [15] is expressed as

$$-\ln(1-x) = K_1 \beta^{-n} \exp\left(-1.052 m \frac{E}{RT}\right) + \text{constant} \quad [16]$$

Equation [16] can be rewritten as Eq. [2] for estimating the activation energy for crystal growth. Because the crystallized fraction  $x$  at the peak temperatures  $T_p$  on differential scanning calorimetry (DSC) [or differential thermal analysis (DTA)] curves is almost constant irrespective of cooling rate  $\beta$ ,<sup>[5]</sup> Eq. [2] is available at the peak temperature  $T_p$ . The activation energy can be calculated from the slope  $-1.052 m E_g/nR$  provided that the numerical factors  $n$  and  $m$  are known.

However, for the case of crystallization from the melt in a cooling process, the crystallization commences from

a elevated temperature. The relationship  $|\Delta G| \ll RT$  cannot be met in this case.<sup>[23]</sup> Therefore, Eq. [3] cannot be simplified in the Arrhenius form as Eq. [4] under this condition. In addition, the radius  $r$  of crystal particle in this case can be expressed by the following equation:

$$r = \int_{T_s}^T \frac{U}{\beta} dT = \frac{U_0}{\beta} \left[ \int_0^T \exp(-E/RT) dT - \int_0^{T_s} \exp(-E/RT) dT \right] \quad [17]$$

where  $T_s$  is the starting temperature of the cooling process. Because  $T_s$  is greater than any certain temperature  $T$ , the value of the term  $\int_0^{T_s} \exp(-E/RT) dT$  in Eq. [18] is larger than  $\int_0^T \exp(-E/RT) dT$  in Eq. [17]. Therefore, the term  $\int_0^{T_s} \exp(-E/RT) dT$  cannot be neglected, and consequently, Eq. [17] cannot be approximately simplified as Eq. [6] as Eq. [5] for the heating process.

It can be concluded that when the crystallization occurs from melt in a cooling process, the term  $1 - \exp(-\Delta G/RT)$  in the basic rate equation of crystal growth and the term  $\int_0^{T_s} \exp(-E/RT) dT$  depending on the starting temperature of the cooling process cannot be neglected. In this case, the radius  $r$  of a crystal particle should be expressed as

$$r = \frac{U_0 \exp(-5.330)}{\beta R} \times \left\{ \begin{array}{l} \left[ E \exp\left(-1.052 \frac{E}{RT}\right) - E \exp\left(-1.052 \frac{E + \Delta G}{RT}\right) - \Delta G \exp\left(-1.052 \frac{E + \Delta G}{RT}\right) \right] \\ - \left[ E \exp\left(-1.052 \frac{E}{RT_s}\right) - E \exp\left(-1.052 \frac{E + \Delta G}{RT_s}\right) - \Delta G \exp\left(-1.052 \frac{E + \Delta G}{RT_s}\right) \right] \end{array} \right\} \quad [18]$$

Consequently, Eq. [18] cannot be simplified as Eq. [7] or Eq. [13], resulting in the failure in obtaining generalized expression Eq. [1] or Eq. [2] for melts crystallization based on the above derivation. Therefore, it can be concluded that the application of Eq. [1] or Eq. [2] in determining the activation energy for crystallization that occurs on cooling is questionable.

For the cooling process, the cooling rate  $\beta$  will be expressed with negative numbers. Because both the Matusita equations and the Kissinger equation require taking the logarithm of  $\beta$ , they cannot be directly applied to crystallization of melts that occurs on cooling ( $\beta < 0$ ). This problem has been bypassed by dropping off the minus sign for melts crystallization in other studies.<sup>[11–17]</sup> Actually, there have been questionable reports<sup>[16, 17]</sup> regarding positive activation energy for melt crystallization determined by the Matusita equations, where the negative signs of cooling rates were deliberately dropped in calculation. Similarly, some researchers<sup>[11–15]</sup> just dropped the negative sign of the cooling rates in calculation using Kissinger equation, and consequently, they obtained positive values of

activation energy for melts crystallization, which has been proved to be erroneous by later research.<sup>[24]</sup> Vyazovkin<sup>[24]</sup> made a detailed assessment of the Kissinger equation and demonstrated that the Kissinger equation is inapplicable to the processes that occur on cooling. On the contrary, in recent years, various studies have demonstrated that the activation energy for the cooling process should be negative; the crystallization that occurs during the cooling process shows anti-Arrhenius behavior.<sup>[25]</sup> This effective activation energy does not have the usual meaning of an energy barrier in this case but reflects the temperature dependence of the temperature coefficient of the crystallization rate.

According to Hoffman–Lauritzen theory,<sup>[26]</sup> the crystallization rate passes through a maximum for a given temperature,  $T_{\max}$ . If the crystallization temperature  $T_c$  lies in the range of  $T_{\max}$  to melting temperature  $T_m$ , the crystallization will follow an anti-Arrhenius behavior that is characterized by negative values of the temperature coefficient represented by the effective activation energy in crystallization rate equation (for heating process, *i.e.*, Eq. [9]). The crystallization rate is controlled by a nucleation rate whose temperature coefficient is negative. Below  $T_{\max}$ , one could observe the regular Arrhenius behavior that is characterized by positive values of the temperature coefficient. Under this condition, the crystallization rate becomes controlled by

diffusion, which has positive values of the effective activation energy. At the temperature corresponding to maximum crystallization rate  $T_{\max}$ , the activation energy for crystallization is close to zero.

It has been found that there are three different crystallization kinetic regimes. According to Hoffman–Lauritzen theory,<sup>[26]</sup> regime I is strongly dependent on the nucleation rate because the primary crystalline is formed from a single nucleus and grows linearly. As the crystallization kinetic regime transits from regime I to regime II, it leads to a downward break in the crystallization growth rate because the rate of substrate completion becomes approximately of the same order as the surface nucleation rate during regime II. At higher supercooling, the surface nucleation rate becomes very fast and the rate of crystal growth is then governed by the rate of substrate completion. This latter regime is commonly called regime III. The regime II→III transition is accompanied by an upswing break in the growth rate curve. The Hoffman–Lauritzen theory also gives a dependence of the linear growth rate  $U$ , which can be expressed as

$$U = U_0 \exp\left(\frac{-E_x}{R(T - T_\infty)}\right) \exp\left(\frac{-K_g}{T\Delta T f}\right) \quad [19]$$

where  $E_x$  is the effective activation energy for crystallization at a given relative degree of crystallinity  $x$ ,  $T_\infty$  is a hypothetical temperature where motion associated with viscous flow ceases that is taken 30 K (30 °C) below the glass transition temperature  $T_g$ ,  $\Delta T = T_m - T$  is the undercooling,  $T_m$  is the equilibrium melting temperature, and  $f = 2T/(T_m + T)$  is the correction factor. The kinetic parameter  $K_g$  associated with the nucleation process has the following form

$$K_g = \frac{\eta b \sigma \sigma_e T_m}{\Delta h_f k_B} \quad [20]$$

where  $b$  is the surface nucleus thickness,  $\sigma$  is the lateral surface free energy,  $\sigma_e$  is the fold surface free energy,  $\Delta h_f$  is the heat of fusion per unit volume of crystal,  $k_B$  is the Boltzmann constant, and  $\eta$  takes the value 4 for crystallization regimes I and III, and 2 for regime II.

The parameters of the Hoffman–Lauritzen equation can be evaluated by fitting the  $E_x$  vs  $T$  dependence to the following equation, according to the method proposed by Vyazovkin and Sbirrazzuoli:<sup>[27]</sup>

$$E_x(T) = \frac{\bar{U}T^2}{(T - T_\infty)^2} + \frac{K_g R(T_m^2 - T^2 - T_m T)}{(T_m - T)^2 T} \quad [21]$$

where  $\bar{U}$  is the parameter associated with diffusion. It can be demonstrated that the second term  $\frac{K_g R(T_m^2 - T^2 - T_m T)}{(T_m - T)^2 T}$  in Eq. [20] is negative in the temperature range between  $0.618T_m$  and  $T_m$ . Furthermore, the absolute value of this term quickly increases when approaching  $T_m$ . Based on these facts, it can be predicted that the effective activation energy  $E_x$  has large negative values at low extents of crystallization that correspond to small undercooling.

The differential isoconversional method of Friedman<sup>[28]</sup> and the advanced integral isoconversional method of Vyazovkin<sup>[24]</sup> can be considered as alternatives to estimate the activation energy for melts crystallization. The Friedman equation is expressed as

$$\ln\left(\frac{dx}{dt}\right)_{x,i} = -\frac{E_x}{RT_{x,i}} + \text{constant} \quad [22]$$

where  $T_{x,i}$  is the set of absolute temperatures related to a given relative degree of crystallinity at different cooling rates and the subscript  $i$  is the ordinal number of individual cooling rate.

Vyazovkin method has been designed to treat the kinetics that occurs under arbitrary variation in temperature. For a series of  $n$  experiments carried out under different temperature programs  $T_i(t)$ , the activation energy is determined at any particular value of relative degree of crystallinity  $x$  by finding  $E_x$ , which minimizes the following function

$$\Phi(E_x) = \sum_{i=1}^n \sum_{j \neq i}^n \frac{J[E_x, T_i(t_x)]}{J[E_x, T_j(t_x)]} \quad [23]$$

Friedman equation and Vyazovkin equation have been extensively applied to evaluate the effective activation energy for the non-isothermal melt crystallization that occurs on cooling.<sup>[29–33]</sup>

The current communication presents a commentary on the Matusita equation and the modified Matusita equation. The derivation of the Matusita equation and its modified expression for melts crystallization on cooling indicates that, unlike for the crystallization that occurs on heating, the basic rate equation of crystal growth cannot be simplified as the Arrhenius form, and the term  $\int_0^{T_s} \exp(-E/RT)dT$  depending on the initial temperature of the cooling process cannot be neglected. It is demonstrated that both Matusita equation and modified Matusita equation are inapplicable to estimate the activation energy associated with the melt crystallization that occurs on cooling. The isoconversional methods developed by Friedman and Vyazovkin are recommended to determine effective activation energy for melts crystallization that occurs on cooling. The isoconversional method of Friedman has been successfully employed to determine the effective activation energy for non-isothermal crystallization of lime-alumina-based mold fluxes that occurs on cooling in the authors' more recent study.<sup>[34]</sup>

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