Modeling Non-isothermal Recrystallization Phase Fraction

Michael Lanahan

Objective

This document provides specifics regarding the derivation of the models for non-isothermal recrystallization from discrete formulations in recent literature[2;3;4]. The primary intent is to explicitly demonstrate the connection between the discrete models and the notion of *Additivity* in reactions[1;6]. While no prior knowledge of the physical phenomenon is assumed, a basic understanding of calculus and differential equations will aid comprehension. I first specify the isothermal models, then the non-isothermal physical assumptions and their implied mathematical counterparts made when extending to the non-isothermal case. Finally, I present the consequential form of the non-isothermal recrystallization fraction models.

Contents

1 Supporting Material

1.1 Symbol Definitions

T	Fixed temperature
$T(t)$	Temperature function
t	Time
$X(t,T)$	*Isothermal* phase fraction
$Y(t)$	*Non*-isothermal phase fraction
A_1, A_2, B_1, B_2	Arrhenius process parameters
n	JMAK exponent
ν	GL exponent
$b(T)$	JMAK rate
$t_{inc}(T)$	JMAK incubation time
$B(T)$	GL rate
$M(T)$	GL starting time
τ	Effective time.

1.2 Definitions

Definition 1.1 (Arrhenius Process). The *Arrhenius equation* is a formula for the temperature dependence of reaction rates. Recrystallization is a solid phase change and hence a reaction. The speed of this reaction is dependent on temperature. The mathematical model for the rate k is:

$$k(T) = A \exp\left(\frac{B}{T}\right) \tag{1}$$

Definition 1.2 (Scheil Additivity[1]). To find the time τ needed to attain a certain fraction X of transformation with time-temperature path (or temperature *history*):

$$\int_0^\tau \frac{dt}{\tau_X(T(t))} = 1 \qquad (2)$$

Where $\tau_X(T(t))$ is the time to transform the fraction X isothermally at current *isotherm* $T(t)$.

Theorem 1.1 (Sufficient Condition for Scheil Additivity). *If the isothermal reaction rate $X'(t)$ can be written as a product of functions $h(T)$ and $g(X)$ then $X(t)$ is additive.*

$$X'(t) = h(T)g(X)$$

Definition 1.3 (Additivity). More generally, consider an isothermal rection $X(t, T)$ and a non-isothermal reaction counterpart $Y(T(t))$ of the same process. $Y(t)$ is additive if it satisfies:

$$Y'(t) = X'(t = \tau_X(Y(t), T = T(t))$$

Theorem 1.2 (Sufficient Condition for Additivity). *Let X be the reaction product. If*

$$X'(t) = f(X, T) \qquad (3)$$

*Then X is **additive**. For proof see[6].*

2 Isothermal Recrystallization Fraction Models

I provide the Arrhenius process forms of $b(T), t_{inc}(T), B(T), M(T)$ here for completeness. Importantly, these functional forms have no influence on the final expressions for non-isothermal recrystallization.

2.1 JMAK Model

The (modified) Johnson–Mehl–Avrami– Kolmogorov (JMAK) model includes both an incubation phase and recrystallization phase with $t < t_{inc}(T)$ the recrystallization fraction $X(t, T) \equiv 0$.

$$X(t, T) = 1 - \exp\left(-b^n(t - t_{inc})^n\right) \qquad (4)$$

$$b(T) = A_1 \exp\left(\frac{B_1}{T}\right) \qquad (5)$$

$$t_{inc}(T) = A_2 \exp\left(\frac{B_2}{T}\right) \qquad (6)$$

2.2 Generalized Logistic Model

The Generalized Logistic (GL) model is the solution to Richard's differential equation, and generalizes the logistic function. The equation presented here is simplified for the use case.

$$X(t, T) = \frac{1}{(1 + e^{-B(t-M)})^{1/\nu}} \qquad (7)$$

$$B(T) = A_1 \exp\left(\frac{B_1}{T}\right) \qquad (8)$$

$$M(T) = A_2 \exp\left(\frac{B_2}{T}\right) \qquad (9)$$

$$(10)$$

3 Non-isothermal Process

3.1 RXMAT

The *RXMAT model*, coined by[2] and pioneered by[5], used by[3;4] for non-isothermal recrystallization begins with the JMAK model. The temperature in each discrete time interval $\Delta t_i = t_{i+1} - t_i$ is approximated as constant. The effective time, τ, is computed using the approximately constant temperature $T(t_i + \Delta t_i)$ and the recrystallization fraction $Y(t_i)$. The contribution to $Y(t_i)$ leading to $Y(t_{i+1})$ is computed over the $[\tau, \tau + \Delta t_i]$ using the expression for isothermal recrystallization, as the temperature is assumed approximately constant in this window.

Let t_n denote a discrete time series of length n. The non-isothermal recrystallization has two phases: (1) incubation and (2) recrystallization.

3.1.1 Incubation Phase

Let the cumulative incubation fraction be:

$$\rho(t_n) = \sum_{i=1}^n \frac{\Delta t_i}{t_{inc}(T(t_i))} \qquad (11)$$

If $\rho(t_n) < 1$ the material is in the incubation phase and $Y(t_n) = 0$. At $\rho(t_n) = 1$ the recrystallization phase begins.

3.1.2 Recrystallization Phase

The following two-step procedure computes the non-isothermal recrystallization fraction.

Step 1: Invert the JMAK equation and retrieve the effective time τ.

$$\tau = -\frac{(\ln(1 - Y(t_i)))^{1/n}}{b(T(t_i + \Delta t_i))} \qquad (12)$$

Step 2: Compute the contribution over Δt_i to recrystallization fraction Y.

$$Y(t_i + \Delta t_i) = \qquad (13)$$
$$1 - \exp\left(-b(T(t_i + \Delta t_i))^n(\tau + \Delta t_i)^n\right) \qquad (14)$$

3.2   Assumptions

Eq. 14 implies that the non-isothermal rate of recrystallization is equal to the rate of isothermal recrystallization at the same recrystallization fraction $Y(t)$ and temperature $T(t)$. From the development of the RXMAT model, assuming temperature is constant over the window $[\tau, \tau + \Delta t_i]$ implies that to first order the temperature is constant, and the rate of change of temperature has no effect on the isothermal recrystallization.

Assumptions:

1. The *rate* of non-isothermal recrystallization is equal to the rate of isothermal recrystallization at the same phase fraction $Y(t)$ and temperature T.
2. Temperature is essentially constant over a small enough time interval: non-isothermal recrystallization is not a function of $T'(t)$.

Note that the assumption of *General Additivity* is sufficient to meet the RXMAT model assumptions.

4   Non-isothermal Formulations

4.1   Formulae

Here τ is used as the integrated variable.

4.1.1   Non-isothermal JMAK Model

The non-isothermal recrystallization phase fraction from the JMAK model is:

$$Y(t) = \begin{cases} 0 & t \le t_0 \\ Y^\dagger(t - t_0) & t > t_0 \end{cases} \qquad (15)$$

$$t_0 = \rho^{-1}(1) \qquad \rho = \int_0^t \frac{d\tau}{t_{inc}(T(\tau))} \qquad (16)$$

$$Y^\dagger = 1 - \exp\left\{-\left(\int_0^t b(T(\tau))d\tau\right)^n\right\} \qquad (17)$$

The incubation process is Scheil additive, while the recrystallization process is additive in the general sense.

4.1.2   Non-isothermal GL Model

The non-isothermal recrystallization phase fraction from the GL model is:

$$Y(t) = \left(\frac{1}{1 + C\exp\left(-\int_0^t B(T(\tau))d\tau\right)}\right)^{1/\nu} \qquad (18)$$

$$C = \left(\frac{1}{Y_0^\nu} - 1\right)\exp(B_0 M_0)$$

$$Y_0 = \left(\frac{1}{1 + \exp(B_0 M_0)}\right)^{1/\nu}$$

$$B_0 = B(T(0)) \qquad M_0 = M(T(0))$$

4.2   Interpretation

Notably, the forms of these equations closely resemble their isothermal counterparts. The terms:

$$B(T) \qquad b(T)$$

are replaced with

$$\int_0^t B(T(\tau))d\tau \qquad \int_0^t b(T(\tau))d\tau$$

which clearly reduce to the isothermal case for constant temperature. This means that the effect of non-isothermal recrystallization can be computed simply by the cumulative rate weighted by the amount of time spent at each rate.

An immediate and important consequence is that the *order* that temperatures occur in do not matter for the phase fraction at any particular time; only the temperature values matter.

4.3   Proofs

In this section I obtain the forms of the non-isothermal (modified) JMAK and GL models beggining with the discrete RXMAT equations. An equivalent procedure (and indeed implied by the assumptions of the RXMAT model) would be to begin with the definition of General Additivity (defined in Section 1).

4.3.1 Non-isothermal JMAK Model

Proof. We begin with the discrete model defined in Eq. 11. Eq. 16 follows from the straightforward application of a Riemann integral.

Proceeding to the recrystallization phase, I write $b(t)$ in lieu of $b(T(t))$ for parsimony. The strategy is to use Eq. 14 and express $Y'(t)$, the non-isothermal recrystallization fraction *rate* as a differential equation, and then solve that differential equation. Simplifying the expression for Eq. 12:

$$\tau = -\frac{(-W(t))^{1/n}}{b(t)}$$
$$W(t) = -\ln\left(1 - Y(t)\right)$$

Expansion of the discrete terms in Eq. 14 to first order yields:

$$b(t + \Delta t)^n \approx b(t)^n + nb(t)b'(t)\Delta t = b(t)^n$$
$$(\tau + \Delta t)^n \approx \tau^n + n\tau^{n-1}\Delta t$$

as $b'(t) = b'(T(t)) = T'(t)b'(T(t))$ and $T'(t) \approx 0$ Multiplying and factoring returns:

$$b(t+\Delta t)^n (\tau + \Delta t)^n \approx (b(t)\tau)^n + nb(t)^n \tau^{n-1}\Delta t$$
$$= W(t) + C\Delta t \quad C = nb(t)(b(t)\tau)^{n-1}$$

Substitution of the expansion into Eq. 14.

$$Y(t + \Delta t) \approx 1 - \exp\left(-W(t) - C\Delta t\right) =$$
$$Y(t) + \Delta t C(1 - Y(t))$$

The last equality follows from substitution of $W(t)$'s definition.

$$\frac{Y(t + \Delta t) - Y(t)}{\Delta t} \approx C(1 - Y(t))$$
$$Y'(t) = \lim_{\Delta t \to 0} \frac{Y(t + \Delta t) - Y(t)}{\Delta t}$$
$$Y'(t) = C(1 - Y(t))$$

Reconfiguring C from it's definition.

$$C = nb(t)(b(t)\tau)^{n-1} = nW(t)^{\frac{n-1}{n}} b(t)$$
$$= nb(t)\left(-\ln\left(1 - Y(t)\right)\right)^{\frac{n-1}{n}}$$

And so:

$$Y'(t) = nb(t)(1 - Y(t))\left(-\ln\left(1 - Y(t)\right)\right)^{\frac{n-1}{n}}$$

Making use of the expression $W(t)$, and applying the chain rule for $W'(t) = \frac{Y'(t)}{1 - Y(t)}$ before substituting these into the differential expression above yields:

$$W'(t) = nb(t)W(t)^{\frac{n-1}{n}}$$
$$W(t) = \left(\int_0^t b(\tau)d\tau\right)^n$$

By separation of variables, and assuming that the initial condition is 0. The final form (Eq. 17) follows straightforwardly from the definition of $W(t)$ and making explicit the functional form of $b(t) = b(T(t))$.

$\square$

4.3.2 Non-isothermal GL Model

Proof. We can invert the isothermal JMAK equation to obtain it's effective time, depending upon the current recrystallization fraction and temperature, again denoting $B(T(t)) = B(t)$, this leads to:

$$\tau = M(t) - \frac{1}{B(t)}\ln\left(Y(t)^{-\nu} - 1\right)$$

The discrete update rule analogous to the RX-MAT equations is:

$$\tau = M(t) - \frac{1}{B(t + \Delta t)}\ln\left(Y(t)^{-\nu} - 1\right)$$
$$Y(t + \Delta t) =$$
$$\left[1 + \exp\left(-B(t + \Delta t)(\Delta t + \tau - M(t + \Delta t))\right)\right]^{-\frac{1}{\nu}}$$

Substitution of τ into the above expression leads to the following argument in the exponential:

$$\Delta t + \tau - M(t + \Delta t) =$$
$$\Delta t + M(t) - \frac{1}{B(t + \Delta t)}\ln\left(Y(t)^{-\nu} - 1\right) - M(t + \Delta t) =$$
$$(\Delta t + M(t) - M(t + \Delta t)) - \frac{1}{B(t + \Delta t)}\ln\left(Y(t)^{-\nu} - 1\right)$$

and multiplication by the rate $B(t)$ produces:

$$B(t + \Delta t)(\Delta t + M(t) - M(t + \Delta t)) - \ln\left(Y(t)^{-\nu} - 1\right)$$

Let $P = B(t+\Delta t)(\Delta t + M(t) - M(t+\Delta t))$. Substitution of this expression back into the discrete $Y(t + \Delta t)$ leaves:

$$Y(t + \Delta t) = \left[1 + (Y(t)^{-\nu} - 1)\exp\left(-P\right)\right]^{-\frac{1}{\nu}}$$

Focusing our attention on P, to first order:

$$M(t) - M(t + \Delta t) \approx -M'(t)\Delta t \approx 0$$

as $M'(t) = M'(T(t)) = T'(t)M'(T(t))$. So the leading order behavior of $P \approx B(t)\Delta t$. Substitution into the equation yields:

$$Y(t+\Delta t) \approx \left[1 + (Y(t)^{-\nu} - 1)\exp\left(-B(t)\Delta t\right)\right]^{-\frac{1}{\nu}}$$

The remainder of the proof involves setting up the exact expression for $Y'(t)$. To make the steps more concise, I adopt $f(x) = Y(t+\Delta t)$ and $a = Y(t)^{-\nu}$ and $x = B(t)\Delta t$. Then the above equation may is written:

$$f(x) = \left(1 + (a-1)e^{-x}\right)^{-\frac{1}{\nu}}$$

$$Y'(t) = \lim_{x\to 0}\frac{f(x) - f(0)}{\Delta t} =$$

$$\lim_{x\to 0}\frac{f(x) - f(0)}{x}\frac{x}{\Delta t} = f'(0)B(t)$$

$$f'(0) = \frac{1}{\nu}(a-1)a^{-\frac{1}{\nu}-1}$$

And substitution of the definitions just defined produces

$$Y'(t) = \frac{B(t)}{\nu}Y(t)\left(1 - Y(t)^{\nu}\right)$$

Unsurprisingly, we have arrived at Richardson's equation with a coefficient that is not constant, but instead a function of time. Separation of variables, as in the constant coefficient case, leads to the form specified by Eq. 18. I'll sketch these steps:

$$\frac{dY}{Y(1-Y^{\nu})} = \frac{B(t)}{\nu}dt$$

$$\frac{1}{\nu}\ln\left(\frac{Y^{\nu}}{1-Y^{\nu}}\right) = \frac{1}{\nu}\int B(t)dt + C$$

Using the substitution $u = Y^{\nu}$ and a partial-fraction decomposition for the left-hand side.

$$\frac{Y^{\nu}}{1-Y^{\nu}} = K\exp\left(\int B(t)dt\right) \quad K = e^{\nu C}$$

Then isolate $Y(t)$ as:

$$Y(t) = \left[1 + \frac{1}{K}\exp\left(-\int B(t)dt\right)\right]^{-\frac{1}{\nu}}$$

which is equivalent to Eq. 18 after some fiddling with the initial condition K. □

References

[1] John Cahn. Transformation Kinetics During Continous Cooling. *Acta Metallurgica*, 4:572–576, 1956.

[2] A. Durif, M. Richou, G. Kermouche, and J-.M. Bergheau. Numerical study of the influence of tungsten recrystallization on the divertor component lifetime. *International Journal of Fracture*, 230(1):83–98, July 2021.

[3] F. M. B. Fernandes, S. Denis, and A. Simon. Mathematical model coupling phase transformation and temperature evolution during quenching of steels. *Materials Science and Technology*, 1(10):838–844, October 1985. Publisher: SAGE Publications.

[4] Changjun Li, Dahuan Zhu, Rui Ding, Zongxiao Guo, Chuannan Xuan, and Junling Chen. Numerical analysis of recrystallization behaviors for W monoblock under cyclic high heat flux. *Nuclear Materials and Energy*, 32:101227, September 2022.

[5] W. Pumphrey. Inter-relation of hardenability and isothermal transformation data. 1948.

[6] Michael T. Todinov. Alternative approach to the problem of additivity. *Metallurgical and Materials Transactions B*, 29(1):269–273, February 1998.