

## Electronic Supplementary Information (ESI)

### Crystallization and Relaxation Dynamics of Amorphous Loratadine under Different Quench-cooling Temperatures

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## Theoretical Background

### Non-isothermal cold crystallization kinetics

Avrami equation is used to describe the non-isothermal crystallization kinetics:

$$X_t = 1 - \exp(-K(t - t_0)^n) \quad (S1)$$

where  $X_t$  is the degree of crystallization,  $K$  is the crystallization rate constant depending on the nucleation and growth rate,  $t_0$  is the induction time of crystallization and  $n$  is the Avrami exponent depending on the nucleation rate and the dimensionality of the crystallization.<sup>1-4</sup>

Given the non-isothermal character of the investigated process, Jeziorny<sup>4</sup> suggested that the rate parameter should be corrected and given by  $\lg Z_t = \lg K / \Phi$  where  $Z_t$  is same as  $K$  as mentioned above, meaning the crystallization rate constant.

In the DSC measurements,  $X_t$  is defined by the follow equation:

$$X_t = \frac{\int_{t_0}^t \frac{dH}{dt} dt}{\int_{t_0}^{t_\infty} \frac{dH}{dt} dt} = \frac{A_t}{A_\infty} \quad (S2)$$

where  $dH/dt$  is the rate of heat evolution,  $t_0$  and  $t_\infty$  are respectively the time when crystallization starts and ends and  $A_t$  and  $A_\infty$  are the areas of partially and fully crystallized materials under normalized DSC curve, respectively.

Moreover, the non-isothermal crystallization kinetics of amorphous drugs could be analyzed through the logarithmic form of primary crystallization based on eq S2:

$$\lg[-\ln(1 - X(t))] = \lg Z_t + n \lg(t - t_0) \quad (S3)$$

### Molecular dynamics

Imaginary complex permittivity could be described by Havriliak–Negami (HN)<sup>5,6</sup>, which is employed as:

$$\varepsilon^*(\omega) = \varepsilon_\infty + \sum_k \left( \frac{\Delta \varepsilon}{\left(1 + (i\omega\tau_{HN_k})^{\alpha_{HN_k}}\right)^{\beta_{HN_k}}} \right) \quad (\text{S4})$$

where  $\varepsilon_\infty$  is the high frequency limit of the real part  $\varepsilon'(\omega)$ ,  $k$  sums over different relaxation processes (primary or secondary processes),  $\omega = 2\pi f$  is the angular frequency,  $\tau_{HN}$  is the characteristic relaxation time that is related to the frequency of maximal loss  $f_{\max}$ ,  $\Delta\varepsilon$  is the dielectric relaxation strength of the process under investigation;  $\alpha_{HN}$  and  $\beta_{HN}$  are fractional shape parameters ( $0 < \alpha_{HN} \leq 1$  and  $0 < \alpha_{HN}\beta_{HN} \leq 1$ ) describing the symmetric and asymmetric broadening of the dielectric spectrum. And for secondary relaxation process ( $\beta$ -relaxation), when  $\beta_{HN} = 1$  the HN function reduces to Cole–Cole distribution function.<sup>7</sup>

## Fragility parameter

Fragility parameter ( $m_p$ ) is calculated as:<sup>8</sup>

$$m_p \equiv \left. \frac{d \log \tau_\alpha}{d(T_g/T)} \right|_{T=T_g} \quad (\text{S5})$$

This parameter is often used to predict the tendency of glass-forming liquids toward crystallization. The values of  $m_p$  for different materials typically range between  $m_p = 16$  and about 200.<sup>9</sup> The liquids characterized by small values of  $m_p$  are classified as strong systems ( $m < 30$ ), whereas large values of  $m_p$  are a feature of fragile materials ( $m > 100$ ).<sup>10</sup> When  $m_p$  falls within the  $30 < m_p < 100$  range, the super-cooled liquid is classified as intermediate glass-former.<sup>11</sup> The strong liquids usually have a strong thermodynamic stability and a small tendency toward crystallization near  $T_g$ . In contrast with the low values found for the strong liquids, it varies rapidly at the temperature near  $T_g$  for fragile glass-formers.

In DSC test, the thermodynamic fragility could be calculated by the following equation:

$$m_p = 56 \frac{T_g \Delta C_p(T_g)}{\Delta H_m} \quad (\text{S6})$$

where  $\Delta C_p(T_g)$  is the heat capacity at  $T_g$  and  $\Delta H_m$  is the enthalpy of fusion.<sup>12,13</sup>



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