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Mathematical Modelling of Water Absorption Kinetics Using Empirical and Phenomenological Models for Millets

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Abstract: Steeping is one of the major pre-treatment which can reduce anti-nutritional factors without losing dietary fibre and polyphenols unlike in dehulling. In the current study water absorption characteristics of pearl millet and finger millet during steeping at temperature 10°C, 20°C, 30°C, 40°C and 50°C were calculated. Two empirical and one phenomenological model were used to simulate water absorption kinetics. Both the Peleg model and Omoto model were found adequately capable to predict water uptake of pearl millet and finger millet under the designed experimental conditions with regression coefficient more than 0.96. Due to comparatively high variation in grain volume during the hydration process and longer process time, sigmoidal model cannot be utilized for calculation of effective diffusivity and activation energy. Peleg's rate constant shows an inverse relationship with steeping temperature. The activation energy was calculated by substituting effective diffusivity with reciprocal of Peleg's rate constant in Arrhenius equation and was found 25.97 kJ/mol and 32.36 kJ/mol respectively for pearl millet and finger millet.

Keywords: Hydration Kinetics, Millet, empirical model, phenomenological model, Peleg model, Omoto model

I. INTRODUCTION

Millet is a group of cereal crops grown in different parts across the globe. Its adaptability to severe climatic conditions has added to its popularity in temperate countries. India is among the leading countries which produce and utilise millet as a popular cereal crop, other than wheat and rice. Its production is around 16.8% of total cereal production in India. Major varieties of millets grown in India are Sorghum, Pearl millet, Foxtail millet, Little millet, Finger millet, Proso millet, Barnyard millet and Kodo millet. Pearl (*Pennisetum glaucum*) and finger (*Eleusine coracana*) millet comprise of about 20% and 4% of total Indian millet production respectively. Besides containing considerable proportions of starch, proteins and non-starch carbohydrates, millet grains are generally assumed to be also rich in polyphenols and trace elements [1],[2],[3],[4],[5]. The nutritive value of millet has been found to be hindered by a number of anti-nutritional factors, namely tannin, oxalate, phytate, protease and amylase inhibitors [6],[7]. Post-harvest pre-milling treatments, namely dehulling, steeping, germination, fermentation, heat-moisture treatment can significantly decrease or remove these anti-nutritional compounds, which are mostly located in the hull [4],[8],[9],[10],[11]. Dehulling has been the most common practice for removal of antinutritional factors [12]. However, simultaneous removal of important fibre and polyphenolic compounds is a major drawback of dehulling. Tempering or steeping in water is another pre-treatment applied prior to millet flour milling process [9]. It is a slower process, in which water absorption and intra-endosperm moisture migration are controlled by the diffusion process. During the steeping process, the prevailing aqueous atmosphere solubilises the phenolic compounds present in the grains and facilitate their leaching out. The change in colour of the steep liquor is evident during steeping [13],[14]. Water absorption kinetics during steeping has been extensively studied for traditional cereals, pulses and legumes [15],[16],[17],[18]. Pre-set conditions during water absorption like temperature, steeping time and water to grain ratio affect the water absorption capacity, grain volume change as well as water absorption rate. Which could further affect the final product quality or next process [17],[19].

Mathematical modelling is one of the most common practices for process designing and optimization. Despite the presence of various advanced statistical tools and programming, mathematical models dominate the field due to their simplicity and ease of usage [20],[21]. Mathematical models have been applied successfully in studies of hydration kinetics of rice [22],[23], barley [24], corn [23], soybean [25], chickpea [26] and various other beans [15],[18], and seeds [27]. Empirical models are the most widely used mathematical models for hydration kinetics studies as they are very simple and mostly focused on water uptake rate[28]. Peleg's equation is the most common empirical model, which is non-exponential, two-way parameter equation[29]. Initial lag phase during hydration cannot be explained by the Peleg's equation, which generally occurs due to formation of the coating layer. The sigmoidal model was proposed for better understating of such phenomena [15],[28]. A phenomenological model considers such mass transfer phenomena like diffusion, convection, and water concentration inside the grain. Phenomenological model could be based upon lumped parameters like water distribution inside the grain or distributed parameters like concentration gradient inside the grain [16],[22],[25],[30]. Omoto model is based upon lumped parameter considering uniform water concentration inside the grain [31].

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No work on phenomenological modelling of steeping of millet could be found in the currently available literature. However, industrial process optimization of millet steeping would be benefitted by researches through mathematical approaches. Process and quality parameters like moisture content, temperature, density and diffusion can be suitable variables for applying the mathematical approach for steeping process modelling. In this study, two empirical mathematical models, namely Peleg's model and sigmoidal model and one phenomenological model of Omoto were compared for studying hydration kinetics during steeping of pearl and finger millet.

II. MATERIALS AND METHODS

A. *Sample preparation*

Pearl millet and finger millet were procured from the Indian Council of Agricultural Research, Pusa, New Delhi. They were manually cleaned to separate foreign matter and screened to get rid of particulate dust and lighter particles. The seeds were then stored at 4 °C in sealed polypropylene (PP) bags until further use.

B. *Physical properties*

A hundred kernels from each sample were randomly selected and measured in three perpendicular directions for length (L), width (W) and height (H) using a digital calliper (Mitutoyo) with an accuracy of 0.01 mm. The geometric mean diameter (D_g), surface area (S), sphericity (φ), and volume (V_g) were calculated using the following relationships [28]:

$$D_g = (LWH)^{\frac{1}{3}} \quad (1)$$

$$S = \frac{\pi (WH)^{0.5} L^2}{2L-(WH)^{0.5}} \quad (2)$$

$$\emptyset = \frac{D_g}{L} \quad (3)$$

$$V_g = \frac{\pi WHL^2}{6[2L-(WH)^{0.5}]} \quad (4)$$

C. *Steeping procedure*

Twenty grams of sample was soaked in 80 ml distilled water and incubated at 10°C, 20°C, 30°C, 40°C and 50°C in a temperature-controlled water-bath with the temperature accuracy of ±1 °C. Steeping was continued till collection of representative samples at 30th, 60th, 120th, 180th, 240th, 300th, 360th, 420th, 480th, 720th, 960th, 1200th and 1440th min. The excess water was removed by placing samples immediately on a blotting paper and weighed for determination of water uptake. Water absorption capacity (WAC, %) of the samples were calculated using the following equation [17]:

$$WAC = \frac{W - W_o}{W_o} X\,100 \quad (5)$$

Where W is sample grain weight at time t, W_o is initial sample grain weight

D. *Mathematical Modelling*

The empirical model proposed by Peleg is one of the most common mathematical models which is used for the understanding of water absorption and desorption. It is most extensively used to explain water absorption and desorption behaviour of both cereal and non-cereal grains. It is a two-parameter sorption equation, which could be stat as:

$$M_t = M_0 \pm \frac{t}{K_1+K_2 t} \quad (6)$$

Where t is steeping time (hours), M_t is moisture content (dry basis) at time t, M_o is initial moisture content (dry basis), and K_1 & K_2 are the constants. K_1 is Peleg rate constant (1/h%) which is related to sorption rate at the beginning of water absorption, while K_2 is Peleg capacity constant which shows maximum water holding capacity at the end of water absorption process, the time when the sample is in equilibrium with surrounding [32].

The same equation could also be re-written as:

$$\frac{t}{M_t - M_o} = K_1 + K_2 t \quad (7)$$

The above equation is like a linear equation $y = mx + c$. Hence a plot between $t/(M_t - M_o)$ versus steeping time t, could give the values of constant K_1 and K_2.

While the Peleg model works well for homogenous models, it is not that much accurate for heterogeneous materials like grains. Water absorption behaviour of the outer bran layer is significantly different from the endosperm. According to Paquet-Durand et.al [33], Peleg model for water sorption should be applied for the bran layer and endosperm individually and total water sorption should be equal to the sum of both of these corresponding water sorption. They provided following modified Peleg model for water sorption:

$$M_t = \frac{t}{K_{1b}+K_{2b}t} + \frac{t}{K_{1e}+K_{2e}t} \quad (8)$$

Where t is steeping time (hours), M_t is moisture content (dry basis), K_{1b} & K_{2b} are Peleg's constant for the outer bran layer and K_{1e} & K_{2e} are Peleg's constant for endosperm. Here initial moisture content M_o is assumed to be zero because here water absorbed in comparison to initial moisture content is considered.

Apart from Peleg's model, following sigmoidal model was also utilized for the understanding of basic diffusivity of water during hydration [28]:

$$M_t = \frac{M_s}{1+\exp\,[-k.(t-\tau)]} \quad (9)$$

Where t is steeping time (min), M_t is moisture content (dry basis) at time t, M_s is saturation moisture content (dry basis), k is a constant rate of rehydration (min^{-1}) and τ is soaking time (min) at which grain moisture content is half of saturation moisture content.

Peleg's model and sigmoidal model, being empirical models ignore the elementary stages of mass transfer and provide no information regarding the water transportation mechanism. Both models ignore basic grain dimension and physical changes occurring during the hydration process. To overcome these shortcomings, Omoto et al. [31] proposed a phenomenological model which consider theoretical assumption as well as the elementary stage of mass transfer. Considering water concentration as uniform inside the grain initially, Omoto proposed following mass transfer balance equation:

$$\frac{d(\rho_A V)}{dt} = N_A A \quad (10)$$

Here right side represents the rate of change of water mass inside the grain with respect to time t while the left side represents water mass flow flux. N_A is water flow (g/cm^2 h), A is the surface area of the grain, ρ_A is the concentration of water (g/cm^3) and V is grain volume (m^3)

Convective water mass flow could be written as

$$N_A = K_s(\rho_{eq} - \rho_A) \quad (11)$$

Where K_s is the overall mass transfer coefficient (cm/h), and ρ_{eq} is the equilibrium water concentration (g/cm^3) in the grain.

Assuming spherical geometry of the grain with constant volume above mass transfer equation (10) could be written as:

$$\frac{d(\rho_A V)}{dt} = \frac{3K_s}{r}(\rho_{eq} - \rho_A) \qquad (12)$$

Where r grain radius (cm) is The analytical solution of the above Omoto model assuming K_s as constant is presented as [16]:

$$\rho_A(t) = \rho_{eq} - (\rho_{eq} - \rho_{Ao}) \exp\left(-\frac{3K_s t}{r}\right) \qquad (13)$$

For statistical analysis of the results, XLSTAT (trial version) and Microsoft EXCEL were used. All the experiments were conducted in triplicate for all samples. The determination coefficient (R^2) and the root mean square error (RMSE) were used to assess the quality of approximation of considered model and experimental data.

III. RESULTS AND DISCUSSION

A. *Physical Properties*

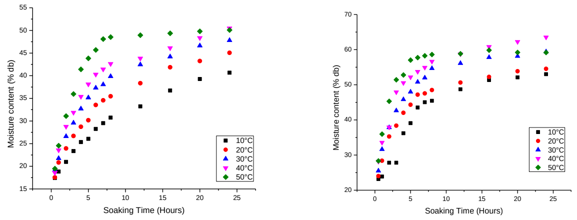
Major physical characteristics of pearl millet and finger millet grains are given in Table 1. The measured length, width, height and geometric mean diameter of the pearl millet grain was 3.363± 0.46, 2.265± 0.23, 2.08± 0.18 and 2.56±0.22 mm, respectively. The results are in a similar range with the previous studies that stated the average length, width and height of pearl millet grain was 2.8 to 3.7 mm, 1.7 to 3.3 mm and 1.4 to 2.1 mm respectively[34],[35]. Similarly, for finger millet grain, values were 1.72± 0.16, 1.7± 0.13, 1.61± 0.19, 1.675± 0.33 respectively, which are close to the range reported by Ramashia et al[36]. The 1000 grain weight was 10.58±0.29 gm and 3 ±0.06 gm respectively for pearl millet and finger millet kernels.

Table 1. Average Values Of Physical Analysis Of Pearl Millet And Finger Millet

Parameter	Pearl millet	Finger millet
Whole seed weight (g)	0.01058	0.003
Length (mm)	3.363	1.72
Width (mm)	2.265	1.7
Thickness (mm)	2.08	1.61
Geometric diameter (mm)	2.509302	1.675111
Degree of sphericity	0.74615	0.973901
Surface (mm^2)	16.93138	8.612186
Volume (mm^3)	6.125013	2.37465
Moisture content (d.b.)	11.706 %	12.007 %
True density (g/cm^3)	1.45	1.48

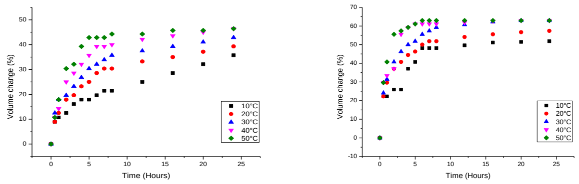
B. *Water absorption kinetics*

During the hydration process, the change in moisture content (on a dry weight basis) and volume (percentage) for pearl millet and finger millet were calculated at the mentioned five different steeping temperatures are shown in figure 1 and figure 2. As observed, the increase in water absorption is directly related to the temperature increase. Though at a higher temperature after initial hydration, water absorption is less compared to a lower temperature. This is due to a high rate of water diffusion at a higher temperature, which causes grain to reach its equilibrium stat faster. Similar behavior was observed in other studies [18],[19],[37]. As indicated in figure 1, as hydration progresses the initial rate of water absorption is higher and then slowly goes to down as grain moisture content approach towards equilibrium. There is a decrease in driving force which is responsible for water transfer inside the grain. A similar effect was also reported during water steeping of sorghum, rice, corn, and lentil [19],[23],[38].



(a) (b)

Fig 1. Effect Of Time And Temperature On Moisture Gain Of Pearl Millet Grain And Finger Millet Grain



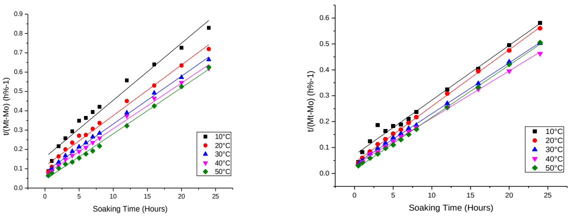
(a) (b)

Fig 2. Effect of time and temperature on volume change in Pearl millet grain and finger millet grain

C. *Fitting of model equations*

The experimental data of moisture content (dry basis) during water absorption was fitted to Peleg's equation. The plot between $t/(M_t - M_o)$ versus steeping time (t) allows the studying of Peleg's constants i.e. Peleg's rate constant (K_1) and capacity constant (K_2) (figure 3). The values of these Peleg constants at mentioned temperatures are presented in Table 2. The competence of the equation for describing the water absorption kinetics could be confirmed by the coefficient of determination (R^2) values, which are greater than 0.97 and 0.98 for pearl millet and finger millet respectively for the studied temperature range.

In present work, values of Peleg's rate constant (K_1) were inversely correlated to the temperature. It indicates that at higher temperatures there is an increase in water absorption rate, which gradually goes down with time (figure 4). This result is in agreement with previous studies on sorghum [39] and finger millet [37].



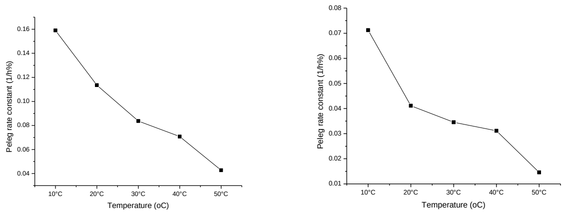
(a) (b)

Fig 3. Application of Peleg's equation to the experimental data on Pearl millet and Finger millet

It is also observed that the Peleg's capacity constant K_2 for pearl millet and finger millet kernels can be a function of temperature at a lower temperature (Figure 5). Similar results have been reported for chickpea [18], acha grain [29], amaranth grain [40], sorghum [39] and finger millet [37].

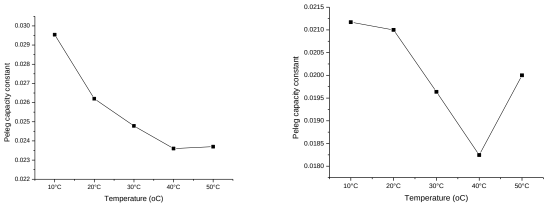
The constant K_2 decreased from 0.0295 to 0.0236 for pearl millet and from 0.0211 to 0.0182 for finger millet, while the steeping temperature (*T*) increased from 10 to 40°C (Table 2).

This is due to increase of temperature resulting into increase in water absorption capacity of pearl millet and finger millet. But constant K_2 increases slightly as steeping temperature increases further from 40°C to 50°C which could be the result of soluble solid loss during the steeping process for prolong duration at a higher temperature [26].



(a) (b)

Fig 4. Effect Of Steeping Temperature On The Peleg Rate Constant (K_1) (1/H%) On Pearl Millet And Finger Millet



(a) (b)

Fig 5. Effect of steeping temperature on the Peleg's Capacity Constant (K_2) ($\%^{-1}$) on Pearl millet and Finger Millet

The water absorption capacity of grain shows an inverse relation with capacity constant. As time processed to infinite, the saturation moisture content (M_s) could be calculated by equation 5 as:

$$M_s = M_0 + \frac{1}{K_2} \tag{14}$$

Table 2. Assessment of the parameters and goodness of fit for the Peleg model for Pearl millet and Finger Millet

Sample	T(°C)	K_1	K_2	M_s	M_i	R^2
Pearl millet	10°C	0.159	0.029	45.562	6.28	0.97
	20°C	0.114	0.026	49.877	8.8	0.98
	30°C	0.084	0.025	52.065	11.97	0.99
	40°C	0.071	0.024	53.939	14.14	0.99
	50°C	0.043	0.024	53.815	23.4	0.99
Finger millet	10°C	0.071	0.021	59.247	14.05	0.98
	20°C	0.041	0.021	57.809	24.33	0.99
	30°C	0.034	0.019	62.936	28.96	0.99
	40°C	0.031	0.018	66.818	32.11	0.99
	50°C	0.014	0.02	61.56	68.54	0.99

Saturation moisture content increases as steeping temperature increases from 10°C to 40°C due to increase in water absorption capacity of millets but it decreases as steeping temperature rises from 40°C to 50°C due to soluble solid loss. The value of saturation constant of pearl millet and finger millet kernel is given in table 2.

Initial rate of water absorption (M_i) could be calculated by deriving equation 5 with respect to time and evaluating moisture content at the very beginning of the hydration process. Initial water absorption rate is directly related to inverse of Peleg's rate constant. It increases rapidly as steeping temperature increases, especially when temperature increases from 40°C to 50°C (Table 2)

Table 3. Assessment of the parameters and goodness of fit for the Modified Peleg model.

Sample	T	K_{1b}	K_{2b}	K_{1e}	K_{2e}	R^2
Pearl millet	10°C	0.179	0.031	8.192	0.236	0.97
	20°C	0.242	0.027	0.044	0.157	0.99
	30°C	-0.067	0.422	0.114	0.025	0.99
	40°C	0.074	0.052	0.246	0.041	0.99
	50°C	0.076	0.036	0.129	0.061	0.97
Finger millet	10°C	0.086	0.02	-1.117	2.402	0.97
	20°C	-0.071	0.369	0.054	0.023	0.99
	30°C	-0.008	0.188	0.053	0.021	0.99
	40°C	-0.116	0.403	0.039	0.019	0.98
	50°C	0.061	0.06	0.029	0.028	0.98

Table 3 represent parameter values for modified Peleg equation. The R^2 values greater than 0.97 for both pearl millet and finger millet confirms the competence of the equation for describing the hydration kinetics of grains within the studied temperature range. There is no significant change in R^2 compared to Peleg equation. This could be due to the small size of the grain, and thinner bran layer which reduces the heterogeneity of structure

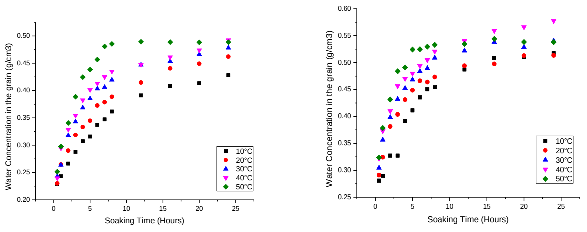
Table 4. Assessment of the parameters and goodness of fit for the Sigmoidal model.

Sample	T (°C)	π (min)	K (cm/min)	R^2	RMSE
Pearl millet	10°C	115±5	0.003	0.989	0.837
	20°C	100±5	0.004	0.983	1.153
	30°C	90±4	0.004	0.981	1.372
	40°C	80±4	0.005	0.96	2.111
	50°C	65±3	0.009	0.997	0.713
Finger millet	10°C	110±5	0.005	0.982	1.574
	20°C	56±2	0.006	0.98	1.647
	30°C	52.5±2	0.006	0.984	1.529
	40°C	51±2	0.006	0.968	2.297
	50°C	36±52	0.013	0.992	0.989

Constant rate of hydration (*k*) for the sigmoidal model is given in table 4 along with the coefficient of determination (R^2) for each temperature, which increases with increase in temperature. This increase in the value of the constant rate of hydration is relatively slow as temperature increase from 10°C to 40°C, with almost stagnant for the temperature change from 20°C to 30°C. But as temperature increase to 50°C from 40°C, the value of the rate of hydration constant increase by 80% for pearl millet and 100% for finger millet.

Effect of temperature and steeping time on water concentration inside the grain is given in figure 6. These data were fitted in Omoto model.

Table 5 represents the value of mass transfer coefficient also known as diffusion coefficient (K_S) and coefficient of determination (R^2) for each temperature. The R^2 values greater than 0.98 and 0.96 for pearl millet and finger millet respectively indicate a good fit for the model to experimental data. As temperature rises the value of diffusion coefficient also increases, which indicate the increased initial velocity of water desorption of grains at higher temperatures [30],[31]. Despite the importance of diffusion coefficient on water uptake rate during steeping, very less work has been reported for Omoto model.



(a) (b)

Fig 6. Effect Of Time And Temperature On Water Concentration In Pearl Millet Grain And Finger Millet Grain

Table 5. Assessment Of The Parameters And Goodness Of Fit For The Omoto Model

Sample	T (°C)	ρ_{EQ}	K_S (cm/min)	R^2	RMSE
Pearl millet	10°C	0.428	0.012986	0.988	1.545
	20°C	0.462	0.014366	0.985	0.021
	30°C	0.478	0.018021	0.994	0.020
	40°C	0.492	0.019033	0.989	0.026
	50°C	0.488	0.029505	0.986	0.011
Finger millet	10°C	0.517	0.011351	0.963	0.025
	20°C	0.513	0.018552	0.980	0.026
	30°C	0.540	0.019735	0.969	0.030
	40°C	0.577	0.017371	0.960	0.042
	50°C	0.538	0.033195	0.973	0.017

D. Activation energy

As per Fick's law, effective diffusivity is directly proportionate to the square of grain radius. Due to comparatively high variation in grain volume during the hydration process and longer process time, Fick's law of diffusion cannot be utilized for calculation of effective diffusivity and activation energy. However, Peleg rate constant could be linked to the diffusion coefficient as it shows a linear relationship with temperature. Sopade et.al. proposed instead of effective diffusivity, reciprocal of K_1 could be used in the Arrhenius equation temperature as [20]:

$$\frac{1}{K_1} = K_o \, exp\frac{E_a}{RT} \qquad (15)$$

Values of Peleg rate constant and temperature were fitted in the above equation for determination of constants. The coefficient of determination (R^2) for the above equation was 0.961 and 0.893 for pearl millet and finger millet respectively, which indicate a good fit for the model to experimental data for pearl millet. The predicted value of activation energy was 25.97 kJ/mol and 32.36 kJ/mol respectively for pearl millet and finger millet.

IV. CONCLUSION

The empirical and phenomenological models were evaluated for water absorption kinetics of pearl millet and finger millet as a function of hydration temperature. Gain in grain volume and water absorption rate remains high during the initial phase of hydration at a higher temperature while at lower temperature this increment is gradual. At higher temperature, the water absorption rate decreases with time. Both Omoto model and Peleg model could be utilized for describing the hydration process. Peleg rate constant decreases linearly as temperature increases while Peleg capacity constant shows a slight increase at a higher temperature. Diffusivity coefficient of Omoto model increases as hydration temperature increases with major increment at a higher temperature. While the hydration rate constant of the sigmoidal model remains almost similar at lower temperature and sudden increase at a higher temperature. Due to considerable small grain size, the change in grain radius during the hydration process is significant, hence first order equation based upon Fick's law and rate of hydration from the sigmoidal model could not be utilized for calculation of effective diffusivity. Although reciprocal of Peleg rate constant could be used in place of effective diffusivity in the Arrhenius equation for calculation of activation energy, as it is inversely related to hydration temperature.

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