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Thermal Evaluation of Food Processes: The Role of a Reference Temperature*

R. V. Nunes,[a] K. R. Swartzel[b,c] & D. F. Ollis[a]

[a]Departments of Chemical Engineering, [b]Food Science and [c]Biological and Agricultural Engineering, North Carolina State University, Raleigh, North Carolina 27695, USA

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ABSTRACT

Thermal evaluation methods for food processes are derived from either the Arrhenius or the Bigelow models, among them the thermal death time method (TDTM) with z = *10°C and the equivalent point method (EPM) are of particular interest. Incorporation of a reference temperature* (T_{Ref}) *into these two methods is examined for both low- and high-temperature thermal processing. Four examples are presented, covering batch and continuous operations. For* T_{Ref} = *121·11°C, the TDTM for a typical canning operation yields a processing time 7% larger than that of the EPM; by contrast, applying the TDTM to continuous processes may result in large underestimations of the processing time, i.e. between 30 to 40% lower than those of the EPM. To avoid such underestimations, a new* T_{Ref} = *145·0°C is proposed, which is obtained by setting the first derivative of the Arrhenius equation equal to 1/*z. *In this way, the design of thermal processes can be achieved with only small overestimations or negligible underestimations. In addition, the EPM makes it possible to evaluate easily* F *and* G *values for the Bigelow and Arrhenius models, respectively.*

INTRODUCTION

Preservation of foods by thermal processing is based upon reducing the probability of survival of both vegetative organisms and bacterial spores. These processes are either batch or continuous. The former, retort

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processes, are normally used in today's food industry. The latter, aseptic processing packaging (APP), are increasingly being used due to the promise of increasing shelf life, reducing product returns, reducing energy requirements throughout the marketing chain and reducing overall costs (Anon. 1980).

Food products are divided traditionally into two main groups: high- and low-acid foods. The former group have microbial contaminants, which are normally less heat resistant than spores. The latter group provide the proper environment for the growth of pathogens (i.e. *Clostridium botulinum*) and, therefore, must receive a more severe thermal process to assure inactivation of these spores. Usually, a desirable level of thermal treatment is established, based upon laboratory generated kinetic data for the pathogens. The thermal treatment required is then incorporated into the process design with established design techniques. Even though the concepts and mathematical steps of process evaluation are not complex, confusion still exists about the accuracy of using either the Bigelow or Arrhenius models (Jones, 1968; Ramaswamy *et al.*, 1989).

For more than 60 years, Bigelow's model has been the scientific basis for the design of thermal processes used by the low-acid canned food industry. Most of these thermal processes have a maximum temperature around 121°C (250°F). By contrast, the Arrhenius model has been recommended over a broader temperature range, i.e. 100 to 150°C (Simpson & Williams, 1974).

Newer designs for thermal optimization require increasing process temperature with decreasing holding time (Swartzel, 1986). Therefore, the most convenient methods of thermal evaluation need revision as we strive for process optimization.

Based upon Bigelow's model, the thermal death time method (TDTM) is widely used by microbiologists and food technologists. Recently, the TDTM has been reviewed and recommended for heat process engineering (Pflug & Odlaug, 1978; Pflug, 1987). Despite being widely used in today's sterilization process evaluation, the TDTM is only an approximation to Arrhenius kinetics (Lawrence & Block, 1968). Based upon the Arrhenius model, the inactivation of spores of *Cl. botulinum* yield an activation energy (E_a) in the range $320 < E_a < 350$ kJ/mol; in particular, $E_a = 343{\cdot}94$ kJ/mol has been reported in the literature (Levine, 1956; Jones, 1968) as an example of highly resistant spores for *Cl. botulinum.*

The integrated Arrhenius model has been widely used by biochemical engineers (Aiba *et al.*, 1973; Bailey & Ollis, 1986). For large temperature ranges, the most convenient description of the denaturation of *Cl.*

botulinum spores is given by the Arrhenius rate expression (Lawrence & Block, 1968; Simpson & Williams, 1974; Lin, 1980). Only recently have parameters for this model been reviewed by Norwig & Thompson (1986). Interestingly, they introduce a reference temperature and summarize the Arrhenius kinetic parameters for spore inactivation reactions.

Based on the Arrhenius model, Swartzel (1982, 1984, 1986) developed the equivalent point method (EPM) of thermal evaluation, which requires a unique pair of parameters: the equivalent time (t_E) and the equivalent temperature (T_E). Recently, Nunes & Swartzel (1990) have shown that weighted least squares regression (WLSR) provides excellent methodology for accurate estimation of t_E and T_E, useful for predicting spore inactivation and constituent changes.

This paper will focus on both the role of the reference temperature and differences in processing time for both the TDTM using Bigelow's model and the EPM using Arrhenius' model.

KINETIC MODELS

For a known thermal history, $T(t)$, two models are frequently used for evaluating processing times.

Bigelow's model

Bigelow (1921) introduced the concept of thermal death time (TDT) from which the TDTM is derived. Thus, the slope of TDT versus temperature plot characterizes the dependence of the reaction constant on temperature. This model approximates the temperature dependence of lethal rates by

$$L = 10^{(T(t) - T_{Ref})/z} \tag{1}$$

where L is the lethal rate (min at T_{Ref}), T is the process temperature (°C), T_{Ref} is the reference temperature (121·11°C), and z is the temperature change required to change the TDT by a factor of 10. For *Cl. botulinum*, a typical value for z is 10°C, which is widely used by microbiologists and food technologists (Pflug & Odlaug, 1978; Pflug, 1987).

For this model, the processing time, F, is evaluated by

$$F = \int_0^{t_R} 10^{(T(t) - T_{Ref})/z} dt \tag{2}$$

where t_R is the final processing time and F is the equivalent time, usually in minutes, at T_{Ref} (121·11°C). Therefore, by introducing the proper time–temperature profile, F is easily obtained by numerical integration.

Arrhenius model

In this model the temperature dependence of the rate constant is given by

$$k(T) = B \exp\left(-\frac{Ea}{RT}\right) \tag{3}$$

where B is the pre-exponential (or frequency) factor (1/s), Ea is the activation energy (J/mol), R is the universal gas constant (8·314 J/mol K), and T is the absolute temperature (K). Clearly, Ea has to be constant over the temperature range of interest. Not only does the rate constant depend on temperature, but also on other environmental conditions such as pH (Lin, 1980; Norwig & Thompson, 1986).

Equation (3) has been verified to give the temperature behavior of most reaction rate constants, within experimental accuracy, over fairly large temperature ranges (Fogler, 1986). For example, Kessler & Fink (1986) studied changes in heated and stored milk; they demonstrated that the Arrhenius model was valid over a surprisingly wide range of time and temperature (from 4 to 160°C). Simpson and Williams (1974) concluded that the most convenient description of the inactivation kinetics of *Cl. botulinum* spores is given by

$$k(T) = 2 \times 10^{40} \exp\left(-\frac{310{\cdot}11 \times 10^3}{8{\cdot}314\,T}\right) \tag{4}$$

for 373 K $< T <$ 423 K. However, higher Ea values should be used for highly resistant spores, i.e. $Ea = 343{\cdot}94$ kJ/mol (Levine, 1956; Jones, 1968).

The magnitude of Ea and B are so different that it is necessary to rescale the parameters by introducing a suitable reference temperature, T_{Ref} (Nelson, 1983, Haralampu *et al.*, 1985; Villadsen & Michelsen,

1987), as follows:

$$k(T) = B \exp\left(-\frac{Ea}{R\,T}\right) = k(T_{Ref}) \exp\left[-\frac{Ea}{R}\left(\frac{1}{T} - \frac{1}{T_{Ref}}\right)\right] \tag{5}$$

In thermal process evaluation, it has been customary to set $T_{Ref} = 121{\cdot}11°C$ (250°F). Interestingly, this rescaling reduces a large part of the inherent correlation between B and Ea (Villadsen & Michelsen, 1987).

Based upon the Arrhenius' model, there are two methods for thermal process evaluation: the Delta-Method (Deindoerfer & Humprey, 1959) and the EPM. The EPM is more powerful than the Delta-Method because it is not restricted to first-order reactions and allows for predictions over a large range of Ea, i.e. 50 to 340 kJ/mol (Swartzel, 1982, 1984; Sadeghi *et al.*, 1986).

In the EPM, the lethality of any thermal process is evaluated by

$$G_{Abs} = \frac{\ln\left(\frac{N_0}{N}\right)}{B} = \int_0^{t_R} \exp\left(-\frac{Ea}{R\,T}\right) dt = t_E \exp\left(-\frac{Ea}{R}\,\frac{1}{T_E}\right) \tag{6}$$

where G_{Abs} is the absolute thermal reduction relationship, N_0 and N are the initial and final concentration, t_E is the equivalent time, and T_E is the equivalent temperature. As suggested by Nunes & Swartzel (1990), a new G value is defined by introducing a reference temperature. It follows from eqns (5) and (6):

$$G = \int_0^{t_R} \exp\left[-\frac{Ea}{R}\left(\frac{1}{T} - \frac{1}{T_{Ref}}\right)\right] dt = t_E \exp\left[-\frac{Ea}{R}\left(\frac{1}{T_E} - \frac{1}{T_{Ref}}\right)\right] \tag{7}$$

where the relationship between G_{Abs} and G is given by

$$G_{Abs} = \exp\left(-\frac{E_a}{R\,T_{Ref}}\right) G \tag{8}$$

This equation allows for the evaluation of any constituent change provided that the kinetic parameters are known. For any chemical reaction, the final concentration changes can be evaluated from

$$M = B\,G_{Abs} = k(T_{Ref})\,G \tag{9}$$

where M is properly defined for zero-, first- and second-order reactions (Swartzel & Jones, 1984; Nunes & Swartzel, 1990).

All the calculations in this paper were carried out as described by Nunes & Swartzel (1990). In particular, WLSR was used to evaluate t_E and T_E.

MODEL COMPARISONS

Models

Both the Arrhenius and Bigelow models are the most widely used ones in thermal evaluation. For $T_{Ref} = 121{\cdot}11°C$, Fig. 1 shows a semilogarithmic plot of lethalities versus temperature for these two models. According to a log transformation of eqn (1), Bigelow's model with $z = 10°C$ results in a straight line with slope $1/z = 0{\cdot}1$ and ordinate $T_{Ref}/z = 12{\cdot}111$. By contrast, the Arrhenius model has a slight downward curvature as is concluded from the negative value of the second derivative of $\log(k)$ with respect to T. Clearly, both models approach each other only in the vicinity of the reference temperature, i.e. $T_{Ref} = 121{\cdot}11°C$.

At temperatures lower than T_{Ref}, lethality values, according to the Bigelow model, are greater than those predicted by the Arrhenius model with $Ea = 343{\cdot}94$ kJ/mol. By contrast, at temperatures higher than T_{Ref}, lethality values, according to the Arrhenius model, are greater than those given by the Bigelow model. In addition, if the Bigelow model is used, longer processing times (F values) will result provided that the process-

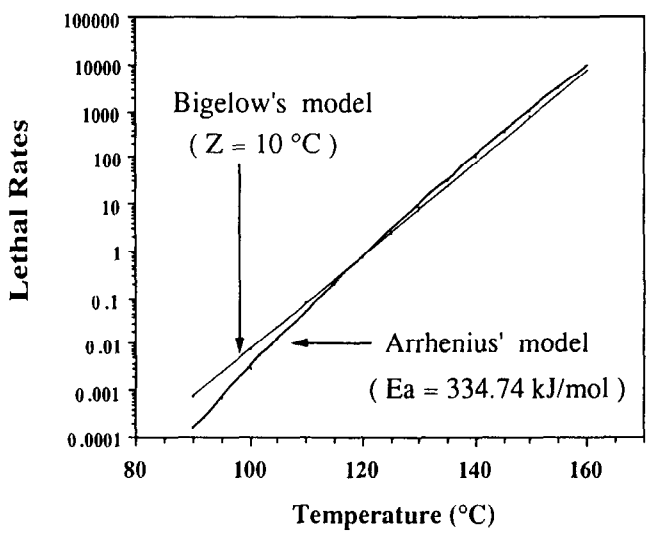

Fig. 1. Lethal rates according to Bigelow's and Arrhenius' models with $T_{Ref} = 121{\cdot}11°C$.

ing temperatures are lower than T_{Ref}, resulting in an overestimation when compared to those given by the EPM (G values). This overestimation in the processing time may explain why the incidence of botulism outbreaks has been dramatically reduced since 1940s (Pflug & Odlaug, 1978). On the other hand, processing temperatures greater than T_{Ref} result in an underestimation of the processing time. In Fig. 1, note that, even though the two lethality curves appear to be very close, their values differ by 40% at 150°C. On the other hand, less resistant spores have lower Ea values, i.e. $Ea = 310{\cdot}11$ kJ/mol (Simpson & Williams, 1974); in this case, lethalities according to the Bigelow model are larger than those given by the Arrhenius model over the entire range of temperatures. Therefore, the TDTM will result in overestimations of the processing time.

Based upon its simplicity, microbiologists and food technologists may still want to continue using Bigelow's model. As shown in Fig. 1, the two curves intersect at $T = 121{\cdot}11$°C. To avoid large differences in the processing time evaluated, using the Bigelow's model, there are two possible solutions. The former is to use two different z values, depending upon whether the process temperature is lower or higher than the reference temperature (121·11°C). Unfortunately, this solution will become complex because two F values are required. The latter is to use a different reference temperature; for example, that temperature where the Bigelow model is tangent to the Arrhenius model, avoiding their intersection. Mathematically, for the Arrhenius model, the slope of the tangent is given by

$$\frac{\mathrm{d}\log(k)}{\mathrm{d}T} = \frac{Ea}{2{\cdot}3026\,R\,T^2} \tag{10}$$

then the reference temperature (°C) is found by setting the slope equal to $1/z$, as follows

$$T_{Ref} = \sqrt{\frac{Eaz}{R\,2{\cdot}3026}} - 273{\cdot}16 \tag{11}$$

For $z = 10$, Table 1 shows how T_{Ref} increases with increasing Ea values.

Typical Ea values for thermally resistant spores are in the range 320 to 350 kJ/mol, resulting in a T_{Ref} between 140 and 150°C, respectively. Therefore, it is reasonable to set $T_{Ref} = 145{\cdot}0$°C, which according to Table 1 corresponds to $Ea = 334{\cdot}7$ kJ/mol. This particular Ea value avoids the need of restricting the analysis to $Ea = 343{\cdot}94$ kJ/mol; however, further research in low-acid foods is needed to find Ea values for

highly resistant spores at ultra high temperatures. Figure 2 shows Bigelow and Arrhenius models for this new T_{Ref}. Clearly, these two curves do not have any intersecting point.

In sterilization processes, spore inactivation is of major concern. The sterilization value (SV) is defined by:

$$SV = \log\left(\frac{N_0}{N}\right) \tag{12}$$

Thus, for the Bigelow model:

$$SV = \frac{F}{D} \tag{13}$$

TABLE 1
Reference Temperature for Different *Ea* values

Ea(kJ/mol)	$T_{Ref}(°C)$
318·0	134·41
326·4	139·74
334·7	144·99
343·1	150·19

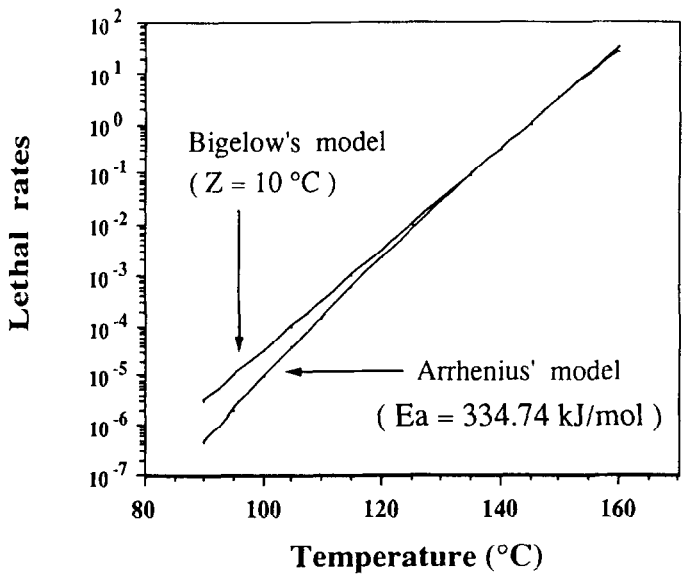

Fig. 2. Lethal rates according to Bigelow's and Arrhenius' models with $T_{Ref} = 145·0°C$.

where

$$D = \frac{2{\cdot}3026}{B \exp\left(\frac{-Ea}{R\,T_{\mathrm{Ref}}}\right)} = \frac{2{\cdot}3026}{k(T_{\mathrm{Ref}})} \tag{14}$$

Similarly, for the Arrhenius model:

$$\mathrm{SV} = \frac{G}{D} \tag{15}$$

Clearly, from either eqn (13) or (15), the percentage of error in the sterilization time (F or G) is equal to the percentage of error in SV.

Navankasattusas & Lund (1978) and Lund (1975) have shown that Ea and z are not independent parameters, as concluded from eqns (1) and (5):

$$z = \frac{2{\cdot}3026\,R\,T_{\mathrm{Ref}}\,T(t)}{Ea} \tag{16}$$

For example, to keep Ea constant ($Ea = 343{\cdot}94$ kJ/mol) different z values have to be used, i.e. $z = 8{\cdot}19$ at 100°C and $z = 9{\cdot}29$ at 150°C. Consequently, to obtain the same result by using either Bigelow's or Arrhenius' models, we need to use a z value that is a function of the process temperature, which is an undesirable feature of Bigelow's model. In addition, if equal processing times are used as a basis for comparing these methods, an erroneous relationship results as shown by eqn (5) in Jones (1968).

The EPM allows for a convenient way to evaluate both F and G values. The EPM provides a set of two parameters, t_{E} and T_{E}, that characterize the entire heating process. Thus, for Bigelow's model, equation (2) yields

$$F = t_{\mathrm{E}} 10^{(T_{\mathrm{Ref}} - T_{\mathrm{E}})/z} \tag{17}$$

The right-hand sides of eqns (7) and (17) allow for comparing these two methods, provided both t_{E} and T_{E} are known. Thus, we see that any process is associated with a particular z value such that:

$$z = \frac{2{\cdot}3026\,R\,T_{\mathrm{Ref}}\,T_{\mathrm{E}}}{Ea} \tag{18}$$

As concluded from eqns (7) and (17), the EPM allows for evaluating sterilization times for both the Bigelow and Arrhenius models; in addition, different spore inactivations are easily evaluated provided that either z or E_a values are known.

Numerical examples

Batch operation
Two examples are included in this section, as follows.

Example 1. A manufacturer has a food product that requires a thermal treatment of 5 min at 120°C. It is desired to find the sterilization time at 140°C; Jones (1968) refers to this problem as example 1. Equation (2) yields:

$$t_{R(140)} = \frac{5 \times 10^{120-121{\cdot}11/10}}{10^{140-121{\cdot}11/10}} = 3 \text{ s}$$

and eqn (7) yields:

$$t_{R(140)} = \frac{5 \times \exp\left[-\left(\frac{343{\cdot}941}{8{\cdot}314}\right) \times \left(\frac{1}{393{\cdot}16} - \frac{1}{394{\cdot}27}\right)\right]}{\exp\left[-\left(\frac{343{\cdot}941}{8{\cdot}314}\right) \times \left(\frac{1}{413{\cdot}16} - \frac{1}{394{\cdot}27}\right)\right]} = 1{\cdot}84 \text{ s}$$

Because T_{Ref} appears in both the numerator and denominator it is cancelled out. This example shows that the processing time at 140°C is independent of the reference temperature. Interestingly, the Bigelow model, with $z = 10$°C, yields a processing time 63% greater than that predicted by the Arrhenius model with $E_a = 343{\cdot}94$ kJ/mol.

Example 2. A typical canning heat treatment is reported by Pflug (1979); for this case, eqns (2) and (7) yield:

$$F = \int_0^{28} 10^{(T - T_{Ref})/10} dt = 8{\cdot}85 \text{ min}$$

$$G = \int_0^{28} \exp\left[-\left(\frac{343{\cdot}941}{8{\cdot}314}\right) \times \left(\frac{1}{T(t)} - \frac{1}{394{\cdot}27}\right)\right] dt = 8{\cdot}218 \text{ min}$$

For this example, the Bigelow model yields a processing time about 7% greater than that of the Arrhenius model. As discussed above, this low

error results from the fact that lethalities for both models are in very close agreement at temperatures close to T_{Ref}. At temperatures lower than 121·11°C, lethal rates have low numerical values and their contribution to the integral is not significant even though their difference is large.

Continuous operation

Two typical APP processes are selected in this section. These processes are divided into three parts, i.e. heating, holding and cooling as reported by Swartzel (1984) and Nunes and Swartzel (1990).

Example 3. A low temperature (L) process for which $t_E = 11{\cdot}0$ s and $T_E = 410{\cdot}0$ K.

Example 4. A high temperature (H) process for which $t_E = 14{\cdot}55$ s and $T_E = 422{\cdot}84$ K.

APP processes are carried out at temperatures between 130 and 150°C. Therefore, as discussed above, the TDTM with $z = 10$°C may yield rather large differences when compared to the EPM with $Ea =$ 343·94 kJ/mol. Table 2 shows F and G values corresponding to two reference temperatures, i.e. 121·11 and 145·0°C. First, for $T_{Ref} =$ 121·11°C, the processing time of treatments L and H result in underestimations of 33·3 and 39·7% when F values are compared to G values. Second, for $T_{Ref} = 145{\cdot}0$°C, the F value for treatment L results in an overestimation of the processing time of only 8·3%. Note that this overestimation has the same order of magnitude as that one in the canning example. On the other hand, the F value for treatment H results in a slight underestimation of only 1·1%, which represents an acceptable difference. Note that G values are evaluated with $Ea = 343{\cdot}94$ kJ/mol while, according to eqn (11), $T_{Ref} = 145{\cdot}0$°C corresponds to $Ea = 343{\cdot}1$ kJ/mol. This small underestimation can be reduced even more by increasing T_{Ref}. For APP processes, these examples show clearly how either underestimations or overestimations in processing time are introduced when F values ($z = 10$°C) are compared with G values using different values for T_{Ref}. In conclusion, to avoid large underestimations in APP processing time calculations it is recommended to set the reference temperature in the range 140 to 150°C. Consequently, more research is needed to validate the Arrhenius model and to obtain Ea values for highly resistant spores in different environments.

The sterilization value depends upon either $k(T_{Ref}) \times F$ or $k(T_{Ref}) \times G$. As shown above, F and G depend upon the time–temperature history and a characteristic parameter (z or Ea). On the other hand, $k(T_{Ref})$

TABLE 2
Processing Times for Different Reference Temperatures

	T_{Ref} = 121·11°C[a]		T_{Ref} = 145·0°C[b]	
	Treat. L	*Treat. H*	*Treat. L*	*Treat. H*
F[c]	8·0	176·0	1·96	43·06
G[d]	12·0	292·0	1·81	43·54

[a]Processing time (min) at 121·11°C.
[b]Processing time (s) at 145·0°C.
[c]TDTM with z = 10°C.
[d]EPM with Ea = 343·94 kJ/mol.

depends upon food composition, temperature, pH, etc. For example, for Spanish rice, the kinetic parameters depend on the pH; that is, at pH = 4, they are k(110°C) = 5·27/min and Ea = 306 kJ/mol and, at pH = 7·0, k(110°C) = 0·972/min and Ea = 334 kJ/mol (Xezones & Hutchings, 1965; Norwig & Thompson, 1986). Therefore, a decrease in acidity causes a slight increase in Ea, but a dramatic decrease in $k(T_{Ref})$, requiring a large increase in G values (or F values) in order to maintain constant SV values. In summary, the final thermal evaluation depends strongly upon both G (or F) and $k(T_{Ref})$.

CONCLUSIONS

The introduction of T_{Ref} makes possible a better understanding of the kinetic evaluation methods for continuous and batch processes. For a given thermal treatment, T_{Ref} is a scaling factor that allows for comparing different reaction rates. This scaling factor presents a simple method for calculating SV values by using eqn (15), which is analogous to the widely used eqn (13).

At present, it is believed that techniques derived from Bigelow's and Arrhenius' models are equivalent. These methods are very close to each other only at temperatures close to T_{Ref}. Thermal processes (e.g. canning) carried out with a maximum temperature of about 121°C may continue using the Bigelow's model with z = 10 and T_{Ref} = 121·1°C. By contrast, thermal evaluation of processes carried out in the range 130–150°C (i.e. APP) requires a higher value for T_{Ref}, for example T_{Ref} = 145·0°C. This new T_{Ref} is obtained by making the slope of the

tangent of the Arrhenius curve equal to $1/z$. Because of the widespread practice of using Bigelow's model, microbiologists and food technologists may find it easier to use different T_{Ref} values for different temperatures ranges. In summary, the proper selection of T_{Ref} makes it possible to avoid undesirable, large underestimations in thermal processes carried out at temperatures higher than 121°C.

The EPM has the main advantage of characterizing any heat treatment process by both t_E and T_E; these parameters allow for calculating the conversion of chemical reactions provided that the kinetic parameters are known. The EPM is a new tool that makes it possible to evaluate not only spore inactivation, but also many changes occurring in the food product. Furthermore, the EPM may be used to evaluate kinetic parameters, especially at high temperatures where thermal lags are conveniently accounted for. For chemical and biochemical reactions that follow the Arrhenius' model, it is better to characterize the temperature dependence of the rate constant by both $k(T_{Ref})$ and Ea, avoiding the use of z values. For any thermal process, the EPM yields both F and G values by using eqns (7) and (17); as a result, the effect of different parameters (z, Ea) on the thermal treatment is easily obtained. In conclusion, based upon G values, the EPM allows for comparisons of processing time not only among different thermal processes, but also between the Arrhenius and the Bigelow models.

Accurate thermal process evaluation requires accurate kinetics parameters. Obviously, from a practical point of view, safety is of major concern. Taking into account both $k(T_{Ref})$ and G (or F), SV should be evaluated allowing for the effect of both temperature and food properties (pH, viscosity, etc.). Clearly, more experimental evidence is needed, in particular for continuous systems at high temperatures, to explore the applicability of the new reference temperature in thermal process evaluation. In addition, the T_{min}, T_{max} concept developed by Ramaswamy *et al.* (1989) may be expanded, with the reference temperature approach presented in this paper, to minimize further transformational error, especially for the higher temperature ranges.

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