



# Derivation of multiple but finite number of imaginary eigenvalues for a two-layer diffusion-reaction problem

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## ABSTRACT

Diffusion-reaction problems occur commonly in heat and mass transfer analysis. In the particular case of a multilayer body under certain conditions, such problems are known to result in imaginary eigenvalues that indicate divergence in the temperature/concentration field at large times. For small values of the reaction coefficient, past work has derived conditions in which an imaginary eigenvalue may exist. This work generalizes this result by showing that more than one, but not infinite imaginary eigenvalues may exist under certain conditions in a one-dimensional two-layer diffusion-reaction problem. A systematic investigation of the eigenequation in the imaginary space is carried out in order to derive a closed form expression for the number of imaginary eigenvalues for any given problem. It is shown that, for other parameters being fixed, the larger the values of the reaction coefficients, the greater is the number of imaginary eigenvalues. However, unlike real eigenvalues, it is shown that the number of imaginary eigenvalues is never infinite. While presented for a two-layer body, similar techniques may be applied for a more general multilayer body, although the derivation of explicit equations may be more challenging. Besides its theoretical importance, accounting for imaginary eigenvalues in the problem discussed here is clearly important to ensure accuracy of the mathematical model. The present work contributes towards this by systematically identifying the number and nature of imaginary eigenvalues that occur in multilayer diffusion-reaction heat and mass transfer problems.

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## 1. Introduction

Heat and mass transfer in a multilayer body is commonly encountered in engineering processes and systems, including energy conversion [1], thermal management [2], drug delivery [3] and manufacturing [4]. While some such problems are governed only by diffusion, for which standard analytical solutions are available [5,6], many problems include additional complications, such as reaction and convection terms [3,7–9]. Consideration of this problem in one spatial dimension offers analytical simplification [3,7,8], whereas analysis in two or three dimensions may be necessary in specific cases [10–12]. Development of analytical solutions that account for such complications is important for accurate design and optimization of such systems, towards improved performance, safety and reliability.

The solution of multilayer transport problems is usually derived using separation of variables method, in which, the solution is written as an infinite series for each layer, and the boundary

and interface conditions are used to derive the eigenequation for the problem [5]. Orthogonality of eigenfunctions involves contributions from each layer, with additional complications when convection is present [8]. Past work has shown that such multilayer problems may admit imaginary eigenvalues [7,8,10,11], even when the problem is one-dimensional [7,8]. Such imaginary eigenvalues are associated with divergence of the temperature/concentration field at large times. This is seen, for example, in the thermal runaway phenomena in Li-ion cells, where an imbalance between temperature-dependent heat generation and dissipation results in a positive feedback loop, leading to uncontrolled temperature rise [13,14]. Imaginary eigenvalues are of much theoretical interest. Further, understanding and computing imaginary eigenvalues is also of much practical importance, since standard eigenvalue computation tools do not search in the imaginary space and ignoring imaginary eigenvalues may lead to inaccurate predictions [7].

In the recent past, an analysis of the multilayer diffusion-reaction problem has been presented [7], showing that under certain conditions, an imaginary eigenvalue may exist. The values of the reaction coefficients considered in this work were quite low, which is why, at most one imaginary eigenvalue was encountered. Based on the monotonically increasing nature of the eigenequa-

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Nomenclature	
$Bi$	Biot number
$h$	convective heat transfer coefficient ( $Wm^{-2}K^{-1}$ )
$i$	unit imaginary number, $i = \sqrt{-1}$
$k$	thermal conductivity ( $Wm^{-1}K^{-1}$ )
$\bar{k}$	non-dimensional thermal conductivity
$M$	number of layers
$N$	eigenvalue norm
$T$	temperature (K)
$z$	spatial coordinate (m)
$t$	time (s)
$\alpha$	diffusivity ( $m^2s^{-1}$ )
$\bar{\alpha}$	non-dimensional diffusivity
$\beta$	reaction coefficient ( $s^{-1}$ )
$\bar{\beta}$	non-dimensional reaction coefficient
$\gamma$	non-dimensional interface location
$\tau$	non-dimensional time
$\theta$	non-dimensional temperature
$\xi$	non-dimensional spatial coordinate
$\lambda$	non-dimensional eigenvalue
Subscripts	
$m$	layer number
$0$	initial value

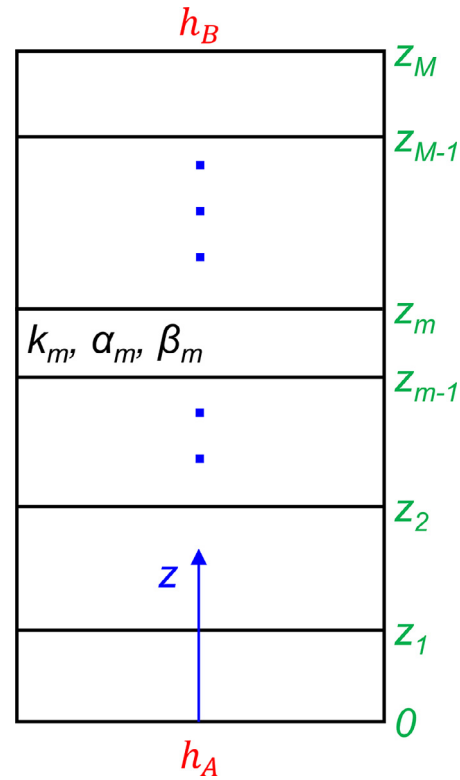


Fig. 1. Schematic of the one-dimensional multilayer body considered in this work with diffusion and linear reaction in each layer.

tion along the imaginary axis, a limiting condition for the existence of an imaginary eigenvalue was derived. It was shown that ignoring the imaginary eigenvalue may lead to significant inaccuracy in computing the temperature field.

In contrast with the work described above, more than one imaginary eigenvalues may exist when the reaction coefficient is large and positive. This paper presents theoretical analysis of a one-dimensional two-layer diffusion-reaction problem in order to determine the number of imaginary eigenvalues of the problem. Through a systematic investigation of the eigenequation along the imaginary axis, a closed-form expression for the number of imaginary eigenvalues for any given problem is derived. It is shown that more than one imaginary eigenvalues may exist, depending on a careful balance between heat/species generation due to the reaction term, and dissipation due to the diffusion term and boundary conditions. Unlike real eigenvalues, it is shown that the number of imaginary eigenvalues is not infinite. The impact of various problem parameters on the imaginary eigenvalues is examined.

## 2. Problem definition and background

Fig. 1 presents a schematic of a one-dimensional  $M$ -layer body of total thickness  $z_M$ . Interfaces between layers are located at  $z = z_m$  ( $m = 1, 2, \dots, M-1$ ). A heat transfer problem is considered here, although, due to heat and mass transfer analogy, results derived here also apply to mass transfer problems. Diffusion occurs within each layer, with a constant and uniform thermal diffusivity  $\alpha_m$  ( $m = 1, 2, \dots, M$ ). Linear, temperature-dependent heat generation/consumption occurs in each layer, with a reaction coefficient  $\beta_m$ . Positive and negative values of  $\beta_m$  correspond to exothermic and endothermic reactions, respectively. Boundary conditions at the two ends are characterized by heat transfer coefficients  $h_A$  and  $h_B$ , respectively. The initial temperature in each layer is  $T_{m,0}$ . All properties are assumed to be constant and uniform.

The following non-dimensionalization may be carried out:  $\theta_m = \frac{T_m - T_{amb}}{T_{ref} - T_{amb}}$ ,  $\xi = \frac{z}{z_M}$ ,  $\tau = \frac{\alpha_M t}{z_M^2}$ ,  $\gamma_m = \frac{z_m}{z_M}$ ,  $\bar{k}_m = \frac{k_m}{k_M}$ ,  $\bar{\alpha}_m = \frac{\alpha_m}{\alpha_M}$ ,  $\bar{\beta}_m = \frac{\beta_m z_M^2}{\alpha_M}$ ;  $\theta_{m,0} = \frac{T_{m,0} - T_{amb}}{T_{ref} - T_{amb}}$ ,  $Bi_A = \frac{h_A z_M}{k_M}$ ,  $Bi_B = \frac{h_B z_M}{k_M}$ , where  $T_m$  refers to the temperature field in the  $m^{\text{th}}$  layer, while  $T_{amb}$  and  $T_{ref}$  are

the ambient temperature and a reference temperature, respectively. Therefore, this problem is governed by the following energy conservation equation for the non-dimensional temperature rise in each layer [7]:

$$\frac{\partial \theta_m}{\partial \tau} = \bar{\alpha}_m \frac{\partial^2 \theta_m}{\partial \xi^2} + \bar{\beta}_m \theta_m \quad (m = 1, 2, \dots, M) \quad (1)$$

The following boundary and interface conditions apply:

$$-\bar{k}_1 \frac{\partial \theta_1}{\partial \xi} + Bi_A \theta_1 = 0 \quad \text{at } \xi = 0 \quad (2)$$

$$\frac{\partial \theta_M}{\partial \xi} + Bi_B \theta_M = 0 \quad \text{at } \xi = 1 \quad (3)$$

$$\theta_m = \theta_{m+1} \quad \text{at } \xi = \gamma_m \quad (4)$$

$$\bar{k}_m \frac{\partial \theta_m}{\partial \xi} = \bar{k}_{m+1} \frac{\partial \theta_{m+1}}{\partial \xi} \quad \text{at } \xi = \gamma_m \quad (5)$$

Eqs. (4) and (5) represent perfect thermal contact and energy conservation, respectively, at each interface ( $m = 1, 2, 3, \dots, M-1$ ). A non-zero initial temperature distribution  $\theta_m = \theta_{m,0}(\xi)$  is assumed at  $\tau = 0$ .

It can be shown [7] that a solution for this problem may be written as follows:

$$\theta_m(\xi, \tau) = \sum_{n=1}^{\infty} c_n [A_{m,n} \cos(\omega_{m,n} \xi) + B_{m,n} \sin(\omega_{m,n} \xi)] \exp(-\lambda_n^2 \tau) \quad (6)$$

$\times (m = 1, 2, \dots, M)$

Where  $\omega_{m,n} = \sqrt{\frac{\lambda_n^2 + \bar{\beta}_m}{\bar{\alpha}_m}}$ . A proof that Eq. (6) satisfies Eq. (1) exactly is presented in Appendix A. The coefficients  $A_{m,n}$  and  $B_{m,n}$  are determined by solving a set of algebraic equations obtained

from the boundary and interface conditions. Coefficients  $c_n$  are derived using quasi-orthogonality of eigenfunctions [5,7]. Finally, it has been shown [7] that an imaginary eigenvalue may be encountered under certain conditions, such as small  $Bi_A$  and  $Bi_B$ , and/or large  $\bar{\beta}_m$ .

### 3. Analysis of the number of imaginary eigenvalues

The goal of the present work is to determine the number of imaginary eigenvalues that may exist. A two-layer body is considered here for derivation of closed-form results. The eigenequation for this case has been shown to be given by [7]

$$f(\lambda) = \bar{k}_1 \omega_1 \frac{-\bar{k}_1 \omega_1 + Bi_A \cot(\omega_1 \gamma_1)}{\bar{k}_1 \omega_1 \cot(\omega_1 \gamma_1) + Bi_A} + \omega_2 \frac{-\omega_2 + Bi_B \cot(\omega_2(1 - \gamma_1))}{\omega_2 \cot(\omega_2(1 - \gamma_1)) + Bi_B} = 0 \quad (7)$$

where  $\omega_m = \sqrt{\frac{\lambda^2 + \bar{\beta}_m}{\alpha_m}}$  ( $m = 1, 2$ ). For analysis of this function in the imaginary space, one may substitute  $\hat{\lambda}^2 = -\lambda^2$ , resulting in the following eigenequation in the imaginary space

$$f(\hat{\lambda}) = \bar{k}_1 \hat{\omega}_1 \frac{\bar{k}_1 \hat{\omega}_1 + Bi_A \coth(\hat{\omega}_1 \gamma_1)}{\bar{k}_1 \hat{\omega}_1 \coth(\hat{\omega}_1 \gamma_1) + Bi_A} + \hat{\omega}_2 \frac{\hat{\omega}_2 + Bi_B \coth(\hat{\omega}_2(1 - \gamma_1))}{\hat{\omega}_2 \coth(\hat{\omega}_2(1 - \gamma_1)) + Bi_B} = 0 \quad (8)$$

where,  $\hat{\omega}_m = \sqrt{\frac{\hat{\lambda}^2 - \bar{\beta}_m}{\alpha_m}} = i\omega_m$  for  $m = 1, 2$ . Note that  $i = \sqrt{-1}$  is the unit imaginary number.

Imaginary eigenvalue analysis presented in the past work [7] proved that the eigenequation increases monotonically along the imaginary axis. Therefore, it was argued that the existence of an imaginary eigenvalue can be predicted on the basis of whether the eigenequation is positive (does not exist) or negative (exists) at the origin,  $\hat{\lambda}=0$ . While such analysis is accurate for relatively mild conditions (small  $\bar{\beta}_m$  and/or large Biot numbers), this may lose accuracy in harsher conditions (large  $\bar{\beta}_m$  and/or small Biot numbers) because the eigenequation may no longer be continuous, and despite being monotonically increasing, the eigenfunction may go to  $\pm\infty$ , resulting in additional eigenvalues at which the eigenequation crosses the  $\hat{\lambda}$  axis. This behavior is similar to the  $\tan$  function, which has an infinite number of roots despite being monotonically increasing, because it goes to  $\pm\infty$  an infinite number of times. As a result, in the present case, it may be possible for an imaginary eigenvalue to exist despite the eigenequation being positive at  $\hat{\lambda}=0$ . This is illustrated in Fig. 2, which plots the eigenequation as a function of  $\hat{\lambda}$  for multiple values of  $\bar{\beta}_2$ , while other problem parameters are held constant ( $\bar{k}_1 = 0.75$ ;  $\bar{\alpha}_1 = 0.75$ ;  $Bi_A = Bi_B = 0.5$ ;  $\gamma_1 = 0.6$ ;  $\bar{\beta}_1 = 0$ ). Fig. 2 shows that for a negative value of  $\bar{\beta}_2$ , the eigenequation starts above the  $\hat{\lambda}$  axis, at  $\hat{\lambda}=0$  and increases monotonically, resulting in no roots. At  $\bar{\beta}_2 = 10$  - a positive but relatively small value - the eigenequation starts below the  $\hat{\lambda}$  axis, increases monotonically and crosses the  $\hat{\lambda}$  axis only once, corresponding to one imaginary eigenvalue. For an even larger value of  $\bar{\beta}_2 = 30$ , it is found that while the eigenequation starts above the  $\hat{\lambda}$  axis and increases monotonically, however, it goes to  $+\infty$  and starts again at  $-\infty$ . As a result of this discontinuity, the eigenequation crosses the  $\hat{\lambda}$  axis once, resulting in one imaginary eigenvalue, despite starting above the  $\hat{\lambda}$  axis. Finally, for an even larger  $\bar{\beta}_2 = 180$ , the eigenequation attains an infinite value twice, resulting in two imaginary eigenvalues.

It is of interest to formally analyze this problem and predict the conditions under which imaginary eigenvalues exist, and if so, the number of such imaginary eigenvalues. Specifically, the threshold

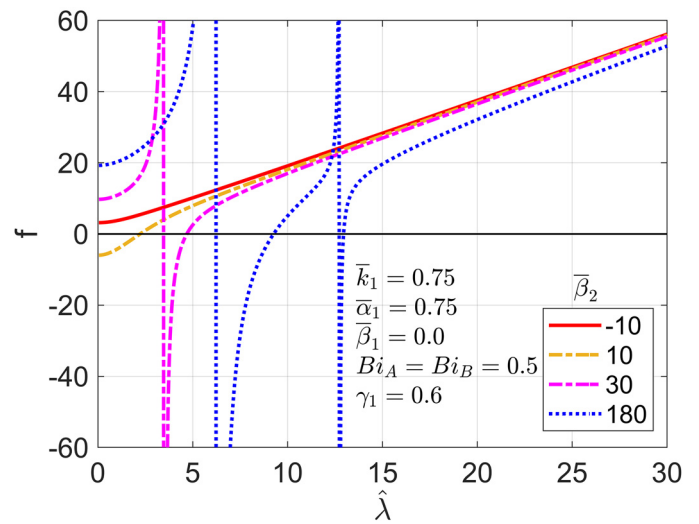


Fig. 2. Eigenequation plot in imaginary space for the two-layer problem. Curves are plotted for multiple values of  $\bar{\beta}_2$ . Other problem parameters are  $\bar{k}_1 = 0.75$ ;  $\bar{\alpha}_1 = 0.75$ ;  $Bi_A = Bi_B = 0.5$ ;  $\gamma_1 = 0.6$ ;  $\bar{\beta}_1 = 0$ .

values of the reaction coefficients that lead to a certain number of imaginary eigenvalues may be of much practical interest, for example in understanding the limits of thermal runaway in Li-ion cells. Towards this, one may proceed by separately examining two independent mechanisms that lead to the eigenequation plot crossing the  $\hat{\lambda}$ -axis, and, therefore, existence of an imaginary eigenvalue.

Firstly, if the value of the eigenequation at  $\hat{\lambda}=0$ , i.e.,  $f(0)$ , is negative, then the monotonically increasing nature of the eigenequation implies the existence of an imaginary eigenvalue. An expression for  $f(0)$  may be derived by inserting  $\hat{\lambda}=0$  in Eq. (8), resulting in [7]

$$f(0) = -\bar{k}_1 \sqrt{\frac{\bar{\beta}_1}{\alpha_1}} \frac{\bar{k}_1 \sqrt{\frac{\bar{\beta}_1}{\alpha_1}} - Bi_A \cot\left(\sqrt{\frac{\bar{\beta}_1}{\alpha_1}} \gamma_1\right)}{\bar{k}_1 \sqrt{\frac{\bar{\beta}_1}{\alpha_1}} \cot\left(\sqrt{\frac{\bar{\beta}_1}{\alpha_1}} \gamma_1\right) + Bi_A} - \sqrt{\frac{\bar{\beta}_2}{\alpha_2}} \frac{\sqrt{\bar{\beta}_2} - Bi_B \cot\left(\sqrt{\bar{\beta}_2}(1 - \gamma_1)\right)}{\sqrt{\bar{\beta}_2} \cot\left(\sqrt{\bar{\beta}_2}(1 - \gamma_1)\right) + Bi_B} \quad (9)$$

Therefore, the existence of an imaginary eigenvalue can be predicted simply by determining the sign of  $f(0)$  given by Eq. (9) above for the given set of problem parameters.

Now, regardless of whether  $f(0)$  is negative or not, additional imaginary eigenvalues may still exist. This is illustrated by the  $\bar{\beta}_2 = 30$  and  $\bar{\beta}_2 = 180$  curves in Fig. 2. In these cases,  $f(0)$  is positive, yet, one and two additional imaginary eigenvalues exist for these two cases, respectively, because the eigenequation attains an infinite value once and twice, respectively, along the imaginary axis. In order to investigate such additional imaginary eigenvalues, it is important to note that due to the monotonically increasing nature of the eigenequation, an additional imaginary eigenvalue is encountered if and only if the eigenequation attains an infinite value, i.e., the eigenequation crosses the  $\hat{\lambda}$  axis as many times as it attains an infinite value and thus must start at  $-\infty$  and cross the  $\hat{\lambda}$  axis once. Therefore, in order to determine the number of imaginary eigenvalues beyond the one that is predicted based on the sign of  $f(0)$ , it is important to determine under what conditions does the eigenequation become infinite, and if so, how many times.

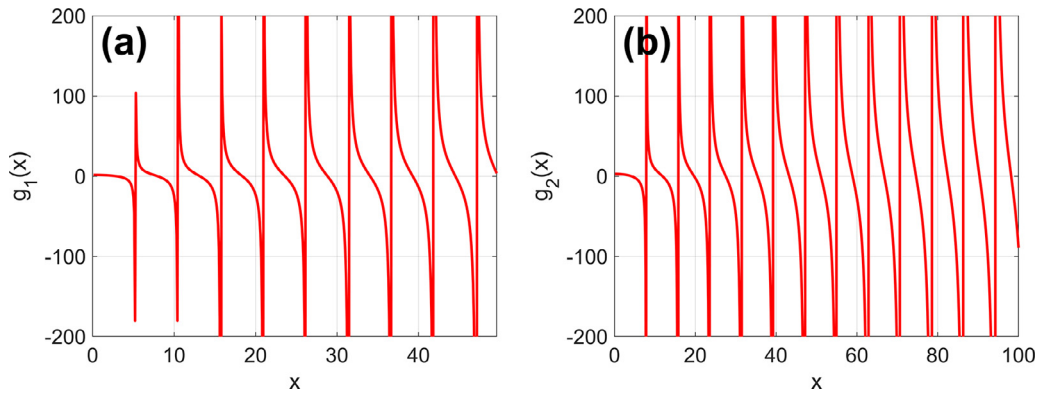


Fig. 3. Plots of functions (a)  $g_1(x)$  and (b)  $g_2(x)$ , showing the infinite number of roots in each case. Problem parameters are  $\bar{k}_1 = 0.75$ ;  $\gamma_1 = 0.6$ ;  $Bi_A = Bi_B = 0.5$ .

It can be seen that an infinite value is attained when one of the two functions appearing in the denominator in Eq. (8) become zero, i.e.

$$\bar{k}_1 \hat{\omega}_1 \coth \hat{\omega}_1 \gamma_1 + Bi_A = 0 \tag{10}$$

or

$$\hat{\omega}_2 \coth \hat{\omega}_2 (1 - \gamma_1) + Bi_B = 0 \tag{11}$$

It is helpful to separately consider the possibility of expressions in Eqs. (10) and (11) becoming zero. These may be interpreted as the separate contributions of the two layers in the problem. Focusing only on the first layer, i.e., Eq. (10), based on the definition of  $\hat{\omega}_1$  given above,  $\hat{\omega}_1$  may be either real or imaginary. When real, the function in Eq. (10) is always positive because  $x \coth x$  is a positive function for real  $x$ . This could occur, for example, when  $\bar{\beta}_1 < 0$ , i.e., the reaction term is endothermic in nature, and therefore, no thermal runaway due to heat accumulation is expected. On the other hand, when  $\hat{\omega}_1$  is imaginary, it is helpful to simplify by substituting  $\hat{\omega}_1 = i\tilde{\omega}_1$ , where  $\tilde{\omega}_1 = \sqrt{\frac{\bar{\beta}_1 - \hat{\lambda}^2}{\bar{\alpha}_1}}$  is real. Substituting in Eq. (10) reduces to

$$\bar{k}_1 \tilde{\omega}_1 \cot \tilde{\omega}_1 \gamma_1 + Bi_A = 0 \tag{12}$$

Therefore, the original function  $f$  given in Eq. (8) may become infinite for those values of  $\hat{\lambda}$  for which Eq. (12) is valid, i.e., for which,  $\tilde{\omega}_1$  is equal to a root of the transcendental equation  $g_1(x) = \bar{k}_1 x \cot(\gamma_1 x) + Bi_A$ . As an example, Fig. 3 plots  $g_1(x)$  for  $\bar{k}_1 = 0.75$ ;  $\gamma_1 = 0.6$ ;  $Bi_A = 0.5$ , and illustrates the infinite number of roots of this equation. Denoting these roots by  $\mu_{1,n}$ , this condition may be written mathematically as

$$\hat{\lambda}^2 = \bar{\beta}_1 - \bar{\alpha}_1 \mu_{1,n}^2 \tag{13}$$

Note that  $\mu_{1,n}$  is an increasing series of numbers, and therefore, only a finite number of  $\hat{\lambda}$  may satisfy Eq. (13). As  $\mu_{1,n}$  increases, the right hand side in Eq. (13) eventually becomes negative, at which point, there is no further possibility of a  $\hat{\lambda}$  to satisfy Eq. (13), given that  $\hat{\lambda}$  is real. Therefore, the total number of imaginary eigenvalues that exist because of the denominator of the first term in Eq. (8) becoming zero may be written as  $n_1$ , where  $n_1$  is the smallest non-negative integer for which  $\bar{\beta}_1 - \bar{\alpha}_1 \mu_{1,n_1+1}^2 < 0$ . Since  $\bar{\beta}_1$  is finite and  $\mu_{1,n}$  is a series of increasing numbers, therefore, this condition will eventually be satisfied by a large enough  $n_1$ , and therefore, the number of imaginary eigenvalues in this problem may be large but never infinite. This is in sharp contrast with the infinitely many real eigenvalues of the problem, which can be proved as follows: the denominators in the eigenequation along the real axis, Eq. (7) will attain a value of zero an infinite number of times, due to the appearance of the  $x \cot x$  function.

This, along with the continuous nature of the eigenfunction implies that the eigenfunction must cross the  $x$ -axis an infinite number of times, and, therefore, must have an infinite number of real eigenvalues. Note that real eigenvalues are not of particular interest for the present analysis, since it is the imaginary eigenvalues that cause divergence of the temperature field at large times.

Note that when  $\bar{\beta}_1$  is negative, or positive but small, then Eq. (13) may be satisfied even when  $n_1 = 0$ , indicating that no imaginary eigenvalues are admitted.

A similar analysis may be carried out for the second term. It may be shown that the number of imaginary eigenvalues due to the second term becoming infinite is given by  $n_2$ , where  $n_2$  is the smallest non-negative integer for which  $\bar{\beta}_2 - \mu_{2,n_2+1}^2 < 0$ , where  $\mu_{2,n}$  are roots of the transcendental equation  $g_2(x) = x \cot((1 - \gamma_1)x) + Bi_B$ . Fig. 3 plots  $g_2(x)$  for  $\gamma_1 = 0.6$ ;  $Bi_B = 0.5$  and shows, similar to  $g_1(x)$ , an infinite number of roots  $\mu_{2,n}$ .

Therefore, the total number of imaginary eigenvalues is given by  $n_1 + n_2 + \delta$ , where  $\delta = 1$  if  $f(0) < 0$  and  $\delta = 0$  otherwise.

Based on the derivation above, a limiting condition for the absence of imaginary eigenvalues, i.e. stability of the system, is that  $f(0) > 0$  (i.e.,  $\delta = 0$ ), and  $\bar{\beta}_1 - \bar{\alpha}_1 \mu_{1,1}^2 < 0$  and  $\bar{\beta}_2 - \mu_{2,1}^2 < 0$ . Under these conditions, one may expect the temperature distribution to not diverge at large times. This set of conditions is an improvement over past work [7], in which, the condition for stability only considered the sign of  $f(0)$ , and therefore, for example, a certain set of parameters could have been predicted to result in stability, when, in fact, it is not.

#### 4. Special case – single layer body

Imaginary eigenvalues similar to the one shown for a two-layer body presented in Section 3 may also exist in the special case of a single layer body. The results derived in Section 3 for a two-layer body may be reduced to those for a homogeneous single layer body by choosing the thermal properties of the two layers to be the same ( $\bar{k}_1 = \bar{\alpha}_1 = 1$ ) and the generation coefficients to the same ( $\bar{\beta}_1 = \bar{\beta}_2 = \bar{\beta}$ ). In such a case, the general eigenequation for the problem, given by Eq. (7) reduces to

$$f(\lambda) = \frac{-\omega + Bi_A \cot(\omega \gamma_1)}{\omega \cot(\omega \gamma_1) + Bi_A} + \frac{-\omega + Bi_B \cot(\omega(1 - \gamma_1))}{\omega \cot(\omega(1 - \gamma_1)) + Bi_B} = 0 \tag{14}$$

where  $\omega = \sqrt{\lambda^2 + \bar{\beta}}$ . It can be shown that the roots of Eq. (14) are the same as the roots of the following equation:

$$f(\lambda) = Bi_A + \omega \frac{-\omega + Bi_B \cot \omega}{\omega \cot \omega + Bi_B} = 0 \tag{15}$$

which indeed matches exactly with the eigenequation derived from first principles for a homogeneous single-layer body.

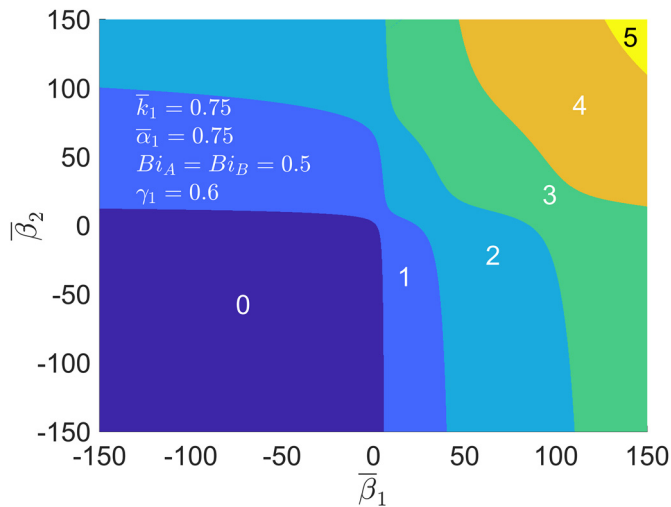


Fig. 4. Colorplot showing the distribution of number of imaginary eigenvalues of the two-layer problem in the  $\beta_1$ - $\beta_2$  space.

Using analysis methods similar to those used in the previous section for a two-layer body, the number of imaginary eigenvalues for this special case may be written as  $n + \delta$ , where  $\delta = 1$  if

$$f(0) = -\sqrt{\beta} \frac{\sqrt{\beta} - Bi_B \cot \sqrt{\beta}}{\sqrt{\beta} \cot \sqrt{\beta} + Bi_B} + Bi_A < 0 \quad (16)$$

and  $\delta = 0$  otherwise. Further,  $n$  is the smallest non-negative integer, for which,  $\beta - \mu_{n+1}^2 < 0$ , where  $\mu_n$  are roots of the transcendental equation  $g(x) = x \cot x + Bi_B$ .

Note that divergence analysis for a homogeneous cylinder with convective boundary condition on the outer surface has been presented in the past, leading to the derivation of a non-dimensional number that governs stability [13]. However, results in this paper were specific to the cylindrical geometry and did not analyze the number of eigenvalues, as has been done here.

### 5. Discussion

It is of interest to determine the regimes in which one or more imaginary eigenvalues arise. Fig. 4 presents a colormap showing the distribution of number of imaginary eigenvalues in the  $\beta_1$ - $\beta_2$  space. Other problem parameters are  $k_1 = 0.75$ ;  $\alpha_1 = 0.75$ ;  $Bi_A = Bi_B = 0.5$ ;  $\gamma_1 = 0.6$ . As expected, the theoretical model presented in Section 2 predicts zero imaginary eigenvalues in the third quadrant ( $\beta_1 < 0$ ,  $\beta_2 < 0$ ). In this region, both reaction terms represent

consumption, and therefore, no imaginary eigenvalues leading to runaway are expected. Fig. 4 shows that no imaginary eigenvalues are expected even for small positive values of either  $\beta_1$  or  $\beta_2$ , provided the other coefficient is strongly negative. This represents a scenario where a small amount of generation in one of the layers is sufficiently counter-balanced by strong consumption in the other layer as well as heat removal from the boundaries, resulting in no runaway. In all quadrants other than the third, the number of imaginary eigenvalues increases as the magnitude of the positive reaction coefficients increases. This is expected because the larger the positive reaction coefficients, the more strongly temperature-dependent the reaction is, resulting in greater heat generation as temperature increases, which is the root cause of divergence and thermal runaway. Note that each additional imaginary eigenvalue is of greater magnitude than previous ones, and therefore, makes the system more and more strongly diverging. This has practical implications for Li-ion batteries, where thermal runaway propagation from one cell to the other may depend on how strong the thermal runaway is [15].

The effect of key problem parameters on the number of imaginary eigenvalues is discussed in Fig. 5. Fig. 5(a) presents the minimum value of reaction coefficient in the second layer that results in one or two imaginary eigenvalues as a function of the Biot number, assumed to be the same on both ends. Reaction coefficient in the first layer is held constant. This plots shows that as Biot number increases, it is possible to tolerate a greater value of  $\beta_2$  before imaginary eigenvalues appear. The minimum value of  $\beta_2$  increases slowly when  $Bi$  is small, and then rapidly because of the increased impact of boundary cooling when  $Bi$  is reasonably large. A saturation effect is observed at large  $Bi$ , when the boundary condition becomes close to isothermal. The limiting value of minimum  $\beta_2$  at small  $Bi$  is positive but small (around 0.1 in this case), indicating that when the boundaries are heavily insulated, even a small reaction coefficient is capable of causing an imaginary eigenvalue, and therefore, thermal runaway. As expected, the minimum value of  $\beta_2$  needed for two imaginary eigenvalues is always larger than for one. Fig. 5(b) plots the minimum value of  $\beta_2$  needed for one or two imaginary eigenvalues as functions of  $\beta_1$ , while other parameters, including the Biot number are fixed. When  $\beta_1$  is negative, the minimum value of  $\beta_2$  is largely insensitive to  $\beta_1$ , which is because a negative value of  $\beta_1$  does not contribute towards imaginary eigenvalues, and in this regime, there is only a small increase in the minimum value of  $\beta_2$  as  $\beta_1$  changes. On the other hand, when  $\beta_1$  becomes positive, it also begins to contribute towards thermal runaway, and therefore, the minimum value of  $\beta_2$  decreases more sharply. Consistent with the theoretical results in Section 2, and similar to Fig. 5(a), it is found that the minimum  $\beta_2$  needed for two imaginary eigenvalues is always greater than that for one.

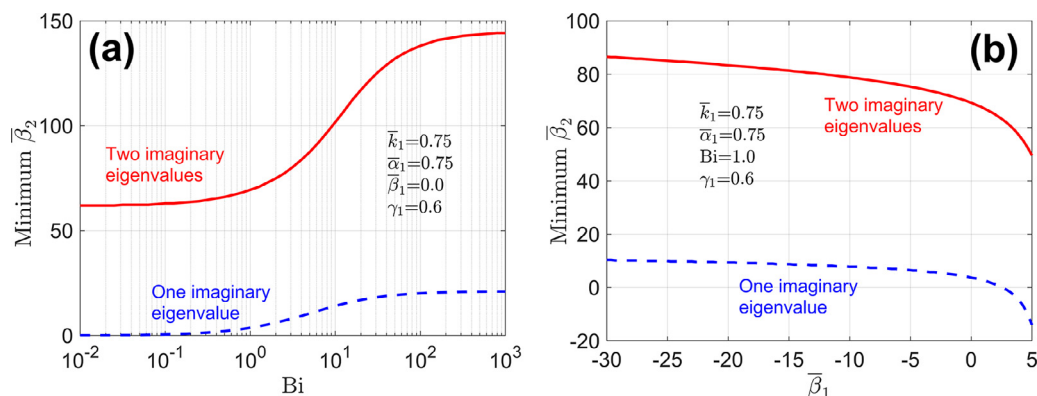
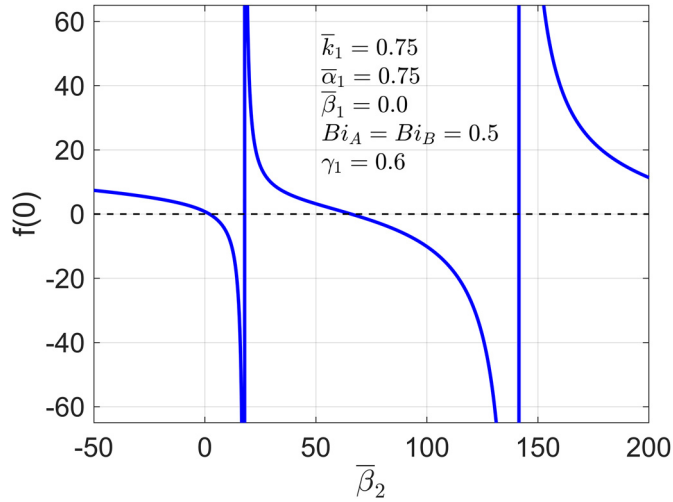


Fig. 5. Effect of key problem parameters on the number of imaginary eigenvalues: Minimum value of  $\beta_2$  that results in one or two imaginary eigenvalues as a function of (a)  $Bi$ , (b)  $\beta_1$ . Other parameters are  $k_1 = 0.75$ ;  $\alpha_1 = 0.75$ ;  $\gamma_1 = 0.6$ . In addition, in (a),  $\beta_1 = 0$ , and in (b),  $Bi = 1$ . The same Biot number is assumed on both ends.



**Fig. 6.** Plot of the initial value of the eigenequation,  $f(0)$  as a function of  $\bar{\beta}_2$ . One imaginary eigenvalue is admitted in regions where  $f(0) < 0$ . Other problem parameters are  $k_1 = 0.75$ ;  $\bar{\alpha}_1 = 0.75$ ;  $Bi_A = Bi_B = 0.5$ ;  $\gamma_1 = 0.6$ ;  $\bar{\beta}_1 = 0$ .

Finally, the sign of  $f(0)$  is of interest, since it determines the existence of one imaginary eigenvalue. As discussed in Section 2, a negative value of  $f(0)$  predicts the existence of at least one imaginary eigenvalue. In order to investigate this further, Fig. 6 plots  $f(0)$  as a function of  $\bar{\beta}_2$ , while other parameters are held constant ( $k_1 = 0.75$ ;  $\bar{\alpha}_1 = 0.75$ ;  $Bi_A = Bi_B = 0.5$ ;  $\gamma_1 = 0.6$ ;  $\bar{\beta}_1 = 0$ ). For negative values of  $\bar{\beta}_2$ ,  $f(0)$  is positive, which is consistent with zero imaginary eigenvalues in that regime. As  $\bar{\beta}_2$  becomes positive and increases,  $f(0)$  becomes negative, which corresponds to the appearance of an imaginary eigenvalue.  $f(0)$  is a discontinuous function, and quite interestingly, at even larger values of  $\bar{\beta}_2$ ,  $f(0)$  becomes positive again. This does not indicate, however, that there are zero imaginary eigenvalues in this regime, because, as discussed in Section 2, imaginary eigenvalues may also appear due to the value of the eigenequation becoming infinite. This is responsible, in this case, for one or more imaginary eigenvalues to appear, despite the positive value of  $f(0)$ .

## 6. Conclusion

Thermal runaway is a well-known problem in engineering systems such as Li-ion cells. Theoretical analysis of such problems is important because thermal runaway experiments are costly and cumbersome to carry out. A good understanding of the nature of eigenvalues in such problems is needed, particularly whether the problem admits imaginary eigenvalues, and if so, how many. This is important not only for theoretical interest, but also for practical computation, since accurate temperature computation requires that all eigenvalues, whether real or imaginary, be accounted for. The key contribution of the present work is in the closed-form expression derived for the number of imaginary eigenvalues for any given set of parameters. This helps understand the fundamental nature of thermal runaway, and can potentially be used to design thermal systems capable of preventing thermal runaway. Results derived here are also applicable to equivalent mass transfer problems in reacting systems.

## Declaration of Competing Interest

All authors hereby declare that they do not have conflicts of interest as described by Elsevier's policies (<http://www.elsevier.com/conflictsofinterest>).

## CRediT authorship contribution statement

**Girish Krishnan:** Conceptualization, Methodology, Validation, Investigation, Data curation, Writing – original draft, Writing – review & editing. **Ankur Jain:** Conceptualization, Methodology, Formal analysis, Validation, Investigation, Data curation, Supervision, Project administration, Writing – original draft, Writing – review & editing.

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## Appendix A: Proof that Eq. (6) is an exact solution of Eq. (1)

Eqs. (1) and (6) are reproduced below:

$$\frac{\partial \theta_m}{\partial \tau} = \bar{\alpha}_m \frac{\partial^2 \theta_m}{\partial \xi^2} + \bar{\beta}_m \theta_m \quad (m = 1, 2, 3 \dots M) \quad (1)$$

$$\theta_m(\xi, \tau) = \sum_{n=1}^{\infty} c_n [A_{m,n} \cos(\omega_{m,n}\xi) + B_{m,n} \sin(\omega_{m,n}\xi)] \exp(-\lambda_n^2 \tau) \quad (m = 1, 2 \dots M) \quad (6)$$

By inserting Eq. (6) in each term of Eq. (1), it can be shown that

$$\frac{\partial \theta_m}{\partial \tau} = \sum_{n=1}^{\infty} -\lambda_n^2 c_n [A_{m,n} \cos(\omega_{m,n}\xi) + B_{m,n} \sin(\omega_{m,n}\xi)] \exp(-\lambda_n^2 \tau) \quad (A.1)$$

$$\bar{\alpha}_m \frac{\partial^2 \theta_m}{\partial \xi^2} = \sum_{n=1}^{\infty} -\bar{\alpha}_m \omega_{m,n}^2 c_n [A_{m,n} \cos(\omega_{m,n}\xi) + B_{m,n} \sin(\omega_{m,n}\xi)] \times \exp(-\lambda_n^2 \tau) \quad (A.2)$$

$$\bar{\beta}_m \theta_m = \sum_{n=1}^{\infty} \bar{\beta}_m c_n [A_{m,n} \cos(\omega_{m,n}\xi) + B_{m,n} \sin(\omega_{m,n}\xi)] \exp(-\lambda_n^2 \tau) \quad (A.3)$$

Adding Eqs. (A.2) and (A.3) results in

$$\bar{\alpha}_m \frac{\partial^2 \theta_m}{\partial \xi^2} + \bar{\beta}_m \theta_m = \sum_{n=1}^{\infty} (\bar{\beta}_m - \bar{\alpha}_m \omega_{m,n}^2) c_n \times [A_{m,n} \cos(\omega_{m,n}\xi) + B_{m,n} \sin(\omega_{m,n}\xi)] \exp(-\lambda_n^2 \tau) \quad (A.4)$$

Since  $\omega_{m,n} = \sqrt{\frac{\lambda_n^2 + \bar{\beta}_m}{\bar{\alpha}_m}}$ , therefore Eq. (A.4) may be re-written as

$$\bar{\alpha}_m \frac{\partial^2 \theta_m}{\partial \xi^2} + \bar{\beta}_m \theta_m = \sum_{n=1}^{\infty} -\lambda_n^2 c_n \times [A_{m,n} \cos(\omega_{m,n}\xi) + B_{m,n} \sin(\omega_{m,n}\xi)] \exp(-\lambda_n^2 \tau) \quad (A.5)$$

Comparing Eqs. (A.1) and (A.5), it can be concluded that

$$\frac{\partial \theta_m}{\partial \tau} = \bar{\alpha}_m \frac{\partial^2 \theta_m}{\partial \xi^2} + \bar{\beta}_m \theta_m \quad (A.6)$$

which is the same as Eq. (1). Therefore, Eq. (6) satisfies Eq. (1) exactly. Note that there are no approximations in the analysis presented above.

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