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Effective Thermal Conductivity Prediction of Foods Using Composition and Temperature Data

James K. Carson, Jianfeng Wang, Mike F. North, Donald J. Cleland



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1 **EFFECTIVE THERMAL CONDUCTIVITY PREDICTION OF**  
2 **FOODS USING COMPOSITION AND TEMPERATURE**  
3 **DATA**

4  
5 James K. Carson<sup>1\*</sup>, Jianfeng WANG<sup>2</sup>, Mike F. NORTH<sup>3</sup>, Donald J. CLELAND<sup>4</sup>

6 <sup>1</sup>University of Waikato, Private Bag 3105, Hamilton, New Zealand

7 <sup>2</sup>Skope Industries Limited, Christchurch, New Zealand

8 <sup>3</sup>Taranaki Bio Extracts, P. O. Box 172, Hawera, New Zealand

9 <sup>4</sup>Massey University, Private Bag 11222, Palmerston North, New Zealand

10 \*corresponding author: [j.carson@waikato.ac.nz](mailto:j.carson@waikato.ac.nz)

11  
12 **ABSTRACT**

13 Thermal conductivity data are important for food process modelling and design.

14 Where reliable thermal conductivity data are not available, they need to be predicted.

15 The most accurate ‘first approximation’ methodology for predicting the isotropic

16 thermal conductivity of foods based only on data for composition, initial freezing

17 temperature and temperature dependent thermal conductivity of the major food

18 components was sought. A key feature of the methodology was that no experimental

19 measurements were to be required. A multi-step prediction procedure employing the

20 Parallel, Levy and Effective Medium Theory models sequentially for the components

21 other than ice and air, ice and then air respectively is recommended. It was found to

22 provide the most accurate predictions over the range of foods considered (both frozen

23 and unfrozen, porous and non-porous). The Co-Continuous model applied in a single

24 step also provided prediction accuracy within  $\pm 20\%$  (on average), except for the

25 porous frozen foods considered. For greater accuracy more rigorous modelling

26 approaches based on knowledge of the foods structure would be required.

27 **Keywords:** thermal conductivity prediction, foods

28

## 29 **1. Introduction**

30 Thermal property data are needed for modelling and design of food processing  
31 operations. Datta (2007) argued that the implementation of advanced thermal  
32 processing models of food is now limited more by the accuracy and availability of  
33 input parameters (which includes thermal conductivity) than by computing power or  
34 modelling expertise.

35

36 A large number of thermal conductivity data may be found in the literature (Houska et  
37 al., 1994; Houska et al. 1997; Rahman 2009, ASHRAE, 2010) and online (e.g.  
38 *Nelfood.com*, Nesvadba et al., 2004) but most of these data are for minimally  
39 processed foods. In the event that thermal property data for the food of interest are not  
40 available, to predict them with similar precision to thermal conductivity  
41 measurement using relatively simple thermal conductivity models would be highly  
42 desirable.

43 The literature describes a large number of thermal conductivity models (Murakami  
44 and Okos, 1989; Maroulis et al., 2002; Carson et al. 2006; Carson 2006, Wang et al.  
45 2006; Rahman, 2009; ASHRAE, 2010). Many of them are simply empirical data-  
46 reduction models, and hence have a limited range of applicability. A large number  
47 have theoretical bases, although many of them include one or more parameters the  
48 values of which must be determined empirically, and these often perform well in

49 model validation exercises (e.g. Murakami and Okos, 1989; Hamdami et al., 2003).  
50 However, if the numerical value of the empirical parameter is an unknown, these  
51 models are of little use if the user intends to perform a prediction without  
52 performing any measurements), particularly if very little is known about the  
53 microstructure of the food. The aim of this paper was to determine the most accurate  
54 model/method for obtaining a first approximation (preferably to within  $\pm 20\%$ ) of the  
55 thermal conductivity of any isotropic food product, by referring just to its composition  
56 data, initial freezing temperature (if applicable) and the temperature of the food,  
57 without the need to perform any measurements.

## 58 **2. Thermal Conductivity Prediction for Foods**

59 The thermal conductivity of food products depends on three basic factors:  
60 composition, processing conditions, and structure (Rahman, 2009). Foods may be  
61 considered as mixtures of the following major components: water, protein, fat,  
62 carbohydrate and ash (i.e. non-combustible solids such as minerals etc). Some foods  
63 may contain a significant volume fraction of ice and/or air (porosity). Temperature is  
64 the most critical processing condition in solid and liquid phases although  
65 pressure can be significant too e.g. high pressure processing (HPP). The  
66 temperature-dependent thermal conductivities of the major food components were  
67 measured by Choi and Okos (1986) and have been reproduced in a number of other  
68 sources (e.g. Rahman, 2009; ASHRAE, 2010). In general terms, the thermal  
69 conductivities of protein, fat, carbohydrates and ash are similar; about three times  
70 lower than that of water, nine times lower than that of ice and ten times higher than  
71 that of air.

72

73 It is the dependence of the thermal conductivity of the food on structure that is  
74 accounted for by the thermal conductivity model. In this study only models which  
75 are functions of the composition of the food and thermal conductivities of the major  
76 food components only, and do not involve any parameters which must be measured  
77 experimentally.

78 For first approximations, one of two approaches may be employed:

- 79 1) Predict the thermal conductivity of the food of concern in a single step by  
80 using a single model equation
- 81 2) Use an algorithm consisting of a number of steps in which more than one  
82 model may be used to predict the thermal conductivity

#### 84 *2.1 Single-Step Approach*

85 Such an approach is desirable because of its simplicity and relative ease of  
86 implementation. In addition to the requirement that they must only require the  
87 volumetric fractions and thermal conductivities of the components as inputs,  
88 suitable models for first approximations will need to be able to be applied to multi-  
89 component mixtures and they should treat each component equally, and hence require  
90 no knowledge of the food structure.

91  
92 The simplest thermal conductivity models that meet the Single-step criteria are,  
93 respectively, the arithmetic, harmonic and geometric weighted means of the thermal  
94 conductivities of the components of the food, where the weighting coefficients being  
95 provided by the volumetric fractions of the food:

96

97 Parallel Model (Rahman, 2009):  $k_e = \sum_i k_i v_i$  (1)

98 Series Model (Rahman, 2009):  $k_e = \frac{1}{\sum_i \frac{v_i}{k_i}}$  (2)

99 Geometric Model (Rahman, 2009):  $k_e = \prod_i k_i^{v_i}$  (3)

100

101 The Series and Parallel models physically match structures where the components are  
 102 in layers perpendicular or parallel to the heat flow direction respectively. The  
 103 geometric model represents no particular physical structure but it is  
 104 mathematically simple. The Series and Parallel models respectively represent the  
 106 theoretical lower and upper bounds of the thermal conductivity of mixtures, provided  
 107 thermal conduction is the only transport mechanism involved (Carson, 2005). It is  
 180 therefore unlikely that they will provide the most accurate predictions; however, since  
 109 they provide limits it is useful to consider their predictions in any modelling exercise.  
 110 The predicted values by the Geometric model always lie between those predicted by  
 111 the Series and Parallel models.

112

113 Two other models which meet the single-step criteria are the well-known Effective  
 114 Medium Theory model (EMT) (Landauer, 1952):

115 
$$\sum_i v_i \frac{k_e - k_i}{2k_i + k_e} = 0$$
 (4)

116 and Wang's Co-continuous model (CC) (Wang et al., 2008):

117 
$$k_e = \frac{\sum_i \frac{v_i}{k_i}}{2} \left( \sqrt{1 + \frac{8 \sum_i k_i v_i}{\sum_i \frac{v_i}{k_i}}} - 1 \right)$$
 (5)

118 The EMT model represents the physical structure where all of the components are  
119 randomly dispersed with each other (co-dispersed) i.e. no component necessarily  
120 represents a continuous phase. The Co-Continuous (CC) model represents a  
121 physical structure where all of the components are continuous but intertwined and  
122 none is dispersed. Figure 1 shows plots of these five models (Eqs. 1 to 5) for a food  
123 with two components in which the ratio of thermal conductivities of the components  
124 ( $k_1/k_2$ ) is 20.

125

126 The well-known Maxwell-Eucken model (described below, Eqs. 8 and 9) represents  
127 the physical structure where a component is dispersed in another one which is  
128 continuous. The above single-step criteria rule out the Maxwell-Eucken model for use  
129 in a single step, since it requires the designation of a continuous, and a dispersed  
130 phase, and is only capable of handling two components at a time. The Maxwell-  
131 Eucken model is, however, suitable for use in a multi-step approach since it does not  
132 contain any empirical parameters.

### 133 *2.2 Multi-Step Approach*

134 While the single-step, single model approach offers simplicity, there is the potential  
135 for greater accuracy from the same input data using a multi-step method, since more  
136 than one structural model may be employed. Also, components in foods seldom exist  
137 in a single well-defined micro-structure. Multi-step thermal conductivity prediction  
138 methods have been proposed and implemented previously (e.g. Maroulis et al., 2002;  
139 Carson, 2006; Cogné et al., 2003); however, only the method proposed by Wang et al.  
140 (2010) does not employ models containing parameters which typically must be

141 determined by experimental measurement, so a similar procedure will be used in this  
142 study, as outlined below.

143 Figure 2 shows plots of the predictions of the Series and Parallel models for four  
144 different thermal conductivity ratios ( $k_1/k_2$ ). The difference between the Series and  
145 Parallel models increases with increasing in  $k_1/k_2$ , and, since this region contains all  
146 the possible effective thermal conductivities (provided  $k_i$  and  $v_i$  are accurate, and only  
147 conduction is involved), it follows that the uncertainty involved in thermal  
148 conductivity prediction also increases as  $k_1/k_2$  increases. Based on the ratio of the  
149 maximum and minimum thermal conductivity components, the problem of thermal  
150 conductivity prediction for foods can be divided into four classes (Carson et al., 2006):

151

- 152 I. Unfrozen, non-porous foods ( $k_{water}/k_{solids} \approx 3$ )
- 153 II. Frozen, non-porous foods ( $k_{ice}/k_{solids} \approx 12$ )
- 154 III. Unfrozen, porous foods ( $k_{water}/k_{air} \approx 25$ )
- 155 IV. Frozen, porous foods ( $k_{ice}/k_{air} \approx 100$ )

156

157 Class I foods with the lowest maximum thermal conductivity ratio are the simplest  
158 foods to predict thermal properties for since the uncertainty involved is relatively low,  
159 as indicated by the small region bounded by the Series and Parallel models (Fig. 2a).

160 In this case, most thermal conductivity models commonly found in the food  
161 engineering and refrigeration literature will provide predictions of sufficient accuracy.

162 However, food Classes II, III and IV provide greater challenges to thermal  
163 conductivity prediction since the thermal conductivity of ice is an order of magnitude  
164 higher than the thermal conductivities of the other components, and the thermal

165 conductivity of air is an order of magnitude lower (Figs. 2b – 2d). Further, for frozen  
 166 or porous foods the location (structure) of the ice or air component may be definitive.  
 167 The approach recommended by Wang et al. (2010) is to break the problem down and  
 168 deal with the different food components sequentially, i.e. Class II foods are  
 169 considered as being a mixture of a Class I food and ice, Class III foods are considered  
 170 as being a mixture of a Class I food and air, and Class IV foods are considered as  
 171 being a mixture of Class II foods and air. This approach is schematically illustrated in  
 172 Figure 3.

173 Wang et al. (2010) recommended that thermal conductivity model for Class I food  
 174 components (as indicated in Figure 3) should be the Parallel model, since it is the  
 175 simplest model yet provides sufficient accuracy and allows for any number of  
 176 components. This approach was also adopted for this study.

177  
 178 Wang et al. (2009) and Wang et al. (2010) recommended that for Class II foods the  
 179 presence of ice should be accounted for using Levy's model (Levy 1981):

$$180 \quad k_{II} = k_{Levy} = k_{ice} \frac{2k_{ice} + k_1 - 2(k_{ice} - k_1)F}{2k_{ice} + k_1 + (k_{ice} - k_1)F} \quad (6)$$

$$181 \quad F = \frac{2/G - 1 + 2(1 - v_{ice}) - \sqrt{[2/G - 1 + 2(1 - v_{ice})]^2 - 8(1 - v_{ice})/G}}{2} \quad (7)$$

$$182 \quad G = \frac{(k_{ice} - k_1)^2}{(k_{ice} + k_1)^2 + k_{ice}k_1/2} \quad (8)$$

183

184 The Levy model physically represents the structure where the two components are  
 185 mixed in a combination of one dispersed in the other which is continuous and vice

186 versa in proportions such that the composite conductivity is the same (i.e. a mixture of  
187 the two versions of the Maxwell-Eucken model as shown in Figure 4).

188

189 Class III and IV foods require consideration of porosity in addition to ice, water and  
190 other components. The effect of porosity on thermal conductivity is complicated by  
191 the widely differing structures that porous foods may have, e.g. air may be dispersed  
192 as bubbles in continuous matrix, or it may form a continuous phase, as is the case with  
193 particulate foods, or in some cases it may exist in both dispersed and continuous  
194 phases. For a given thermal conductivity of the so-called condensed phase (i.e. the  
195 phase containing the solid and liquid components), the thermal conductivity of food  
196 will be significantly higher if the air forms a dispersed phase rather than a continuous  
197 phase (Carson et al., 2005). This is best illustrated by the Maxwell-Eucken model,  
198 which assumes a structure of one phase sparsely dispersed within another. If air forms  
199 the dispersed phase then the Maxwell-Eucken model as the following form (“ME1”):

$$201 \quad k_{ME1} = k_I \frac{2k_{II} + k_a - 2(k_{II} - k_a)v_a}{2k_{II} + k_a + (k_{II} - k_a)v_a} \quad (9)$$

202 If air forms the continuous phase then it has the following form (“ME2”):

$$203 \quad k_{ME2} = k_a \frac{2k_a + k_I - 2(k_a - k_I)(1 - v_a)}{2k_a + k_I + (k_a - k_I)(1 - v_a)} \quad (10)$$

204 Figure 4 shows plots of the two forms of the Maxwell-Eucken model, and it is clear  
205 that the form in which air is the dispersed phase (sponge/foam-like materials) predicts  
206 significantly higher thermal conductivities, than when air is the continuous phase  
207 (particulate materials), other than for very low or very high porosities. The EMT, CC  
208 and levy models are also shown in Figure 4. They each traverse the space between  
209 ME1 and ME2 in different ways due to the structures they represent.

210

211 The objective of this paper was to recommend a procedure in which nothing,  
 212 including the nature of the air distribution, is assumed to be known about the structure  
 213 of the food; however, some general inferences may be drawn about the air-  
 214 distribution from the porosity of the food. Specifically, if the porosity is ‘low’ (e.g. <  
 215 0.3) then it is reasonable to assume (in general) that the air is dispersed as bubbles.  
 216 Likewise, if the porosity is ‘high’ (e.g. > 0.7) then it is reasonable to assume (in  
 217 general) that air forms a continuous phase, and the food is most probably in  
 218 particulate form. In the mid-porosity range both particulates (air continuous) and  
 219 sponge/foam structures (air dispersed) are possible. Ideally, a model accounting for  
 220 porosity would provide similar predictions to the Maxwell-Eucken model with air as  
 221 the dispersed phase for low porosities, similar predictions to the Maxwell-Eucken  
 222 model with air as the continuous phase for high porosities, and predictions which are  
 223 mid-range between the two forms of the Maxwell-Eucken model for mid-range  
 224 porosities. Figure 4 shows that the EMT model fulfils these requirements adequately  
 225 for first approximations (see also Carson et al., 2005), and therefore it is  
 226 recommended for use in the multi-step procedure, to account for porosity, i.e.  
 227 Class III and IV food. The EMT model for the multi-step process is:

$$228 \quad k_{\text{III}} = \frac{(3v_a - 1)k_a + [3(1 - v_a) - 1]k_{\text{I}} + \sqrt{\{(3v_a - 1)k_a + [3(1 - v_a) - 1]k_{\text{I}}\}^2 + 8k_{\text{I}}k_a}}{4} \quad (11)$$

$$229 \quad k_{\text{IV}} = \frac{(3v_a - 1)k_a + [3(1 - v_a) - 1]k_{\text{II}} + \sqrt{\{(3v_a - 1)k_a + [3(1 - v_a) - 1]k_{\text{II}}\}^2 + 8k_{\text{II}}k_a}}{4} \quad (12)$$

230 Eqs. 11 and 12 are simply the two-component forms of Eq. (4) rearranged to be  
 231 explicit in terms of the effective thermal conductivity.  
 232

## 233 2.3 Composition data and ice fractions

234 When using a multi-step method for Class II, III, or IV foods, it is important to bear in  
 235 mind that intermediate volume fractions must be used at the intermediate stages. For  
 236 example, if a Class II (frozen, non-porous) food is being modelled, then the first stage  
 237 is to determine the conductivity of the ‘non-ice’ phase, using the Parallel model. In  
 238 this case the volume fractions employed must be the volume fractions for the non-ice  
 239 phase, e.g. for protein:

$$240 \quad v_p = \frac{v_p}{v_w + v_p + v_f + v_c + v_{ash}} = \frac{v_p}{1 - v_{ice}} \quad (13)$$

241 and similarly for the other components. This may be implemented most conveniently  
 242 by using the following form of the Parallel model, rather than Eq. (1):

$$243 \quad k_I = \frac{k_w v_w + k_p v_p + k_f v_f + k_c v_c + k_{ash} v_{ash}}{v_w + v_p + v_f + v_c + v_{ash}} \quad (14)$$

244 For a complete worked example of this method, refer to Wang et al. (2010).

245

246 It is also important to recognise that composition data for foods are usually available  
 247 on a mass basis, and yet the thermal conductivity models employ volume fractions,  
 248 since thermal conductivity is a volumetric property. The conversion between mass  
 249 and volume fractions for liquid and solid components may be modelled by the  
 250 following relationship (Choi and Okos, 1986; Rahman, 2009):

$$251 \quad v_i = x_i \frac{\rho_{cond}}{\rho_i} \quad (15)$$

252 where:

$$\rho_{cond} = \frac{1}{\sum_i \frac{x_i}{\rho_i}} \quad (16)$$

253 If the food is porous and the porosity (i.e. the volume fraction of air,  $v_a$ ) has not been  
 254 measured it may be estimated from the apparent (bulk) density ( $\rho_e$ ) (Choi and Okos,  
 255 1986; Rahman, 2009):

$$256 \quad \frac{1 - v_a}{\rho_e} = \sum_i \frac{x_i}{\rho_i} \quad (17)$$

257 Density data for the major food components as functions of temperature may be found  
 258 in the same sources as the thermal conductivity data (i.e. Choi and Okos, 1986;  
 259 Rahman, 2009; ASHRAE, 2010). Rahman (2009) discusses other models for  
 260 predicting porosity of foods from composition and density data; however, Eqs. (15) to  
 261 (17) were deemed to be sufficiently accurate for this exercise.

262  
 263 The thermal conductivity of frozen foods is strongly dependent on the ice fraction,  
 264 which in turn is strongly dependent on temperature, and therefore an ice fraction  
 265 model is required for thermal conductivity prediction. There are several ice  
 266 fraction models in the literature (Pham, 1987; Fikiin, 1998; Boonsupthip and  
 267 Heldman, 2007; Rahman 2009). Many of these require calculations of mole fractions,  
 268 which in turn requires estimation of molar masses for the macromolecules (proteins  
 269 and complex carbohydrates). Many contain empirical parameters, and most require  
 270 knowledge of the amount of bound or un-freezable water. The empirical model  
 271 proposed by Tchigeov based only on total water content, system temperature, and  
 272 initial freezing temperature ( $T_F$ ) has been found to work well for  $-45^\circ\text{C} < T < T_F$   
 273 and  $-2 < T_F < -0.4^\circ\text{C}$  (Fikiin, 1998, Pham, 2014):

$$274 \quad x_{ice} = \frac{1.105x_{w,total}}{\left[ 1 + \frac{0.7318}{\ln(T_F - T + 1)} \right]} \quad (18)$$

275 For initial freezing temperatures below  $-2^\circ\text{C}$  a more general relationship may be used:

$$x_{ice} = (x_{w,total} - x_b) \left( 1 - \frac{T_F}{T} \right) \quad (19)$$

Note that in both Eq. (18) and (19) temperature is in degrees Celsius rather than Kelvin. If the fraction of bound water ( $x_b$ ) is unknown it may be related to the composition of the food. For example, Pham (1987) recommended that for meat products

$$x_b = 0.4x_p \quad (20)$$

In this study, Tchigeov's method (Eq. 18) was used for foods with an initial freezing temperature above  $-2$  °C, and Pham's method based on protein composition (Eq. 20) was used for foods with initial freezing temperatures below  $-2$  °C. No simple but sufficiently accurate ice fraction prediction method without empirical parameters was found in the literature. However, unlike the thermal conductivity models that contain empirical parameters which are specific to the food in question and typically need to be determined from a thermal conductivity measurement, the empirical parameters in Eq. (18) and (Eq. 20) apply generally and do not need to be determined from an ice fraction measurement, provided, in the case of Eq. (18) that the initial freezing temperature is in the specified range, and, in the case of Eqs. (19) and (20), the food contains a non-zero protein content. Therefore their use is consistent with the aim of predicting thermal conductivity without experimental measurements being required.

Initial freezing temperature data are available in the literature for some foods, and some predictive models have also been proposed (Boonsupthip and Heldman, 2007; Rahman, 2009). In the absence of any measured data, Fikiin (2014) recommends the use of  $-1.0$  °C for first approximations. In this study, measured initial freezing temperature data were used for all foods considered.

300

301 The amount of unfrozen water in frozen foods is simply the difference between the ice  
302 fraction and the total water content:

$$303 \quad x_w = x_{w,total} - x_{ice} \quad (21)$$

304

### 305 **3. Comparison of Single Step and Multi-step Predictions with Measured Data**

306 The predictions from each of the five single-step models (Eqs. 1 – 5) along with the  
307 multistep procedures (as illustrated in Fig. 3) have been compared to measured  
308 thermal conductivity data from the literature for the four different Classes of foods.

309 The difference is defined as:

$$310 \quad \delta = \frac{|k_{exp} - k_{mod}|}{k_{exp}} \times 100\% \quad (22)$$

311 Only measured thermal conductivity data where the measurement methodology was  
312 proven accurate and the composition of the food (including porosity) and, in the case  
313 of frozen foods, initial freezing temperature were available were considered for this  
314 assessment exercise.

315

#### 316 *3.1 Class I Foods*

317 Figure 5 shows plots of thermal conductivity predictions from Eqs. (1) to (5) as a  
318 function of combined solids content for a range of Class 1 foods at 20 °C (data from  
319 Willix et al., 1998). Table 1 summarises the average differences ( $\delta$ ) between the  
320 model predictions and experimental data for each of the different foods. All the model  
321 predictions are within  $\pm 20\%$ , which is quite acceptable for a first approximation.  
322 Hence the decision to employ the Parallel model in the multi-step procedure is  
323 justified, since it is the simplest model, and for the foods considered actually

324 produced the lowest average difference.

325

### 326 *3.2 Class II foods*

327 Table 2 shows the differences between the model predictions and experimental data  
328 for the same selection of foods (data from Willix et al., 1998); however, this time the  
329 temperature is  $-20^{\circ}\text{C}$ . The Parallel model no longer provides sufficient accuracy;  
330 however, the Co-continuous and Geometric models, as well as the multi-step  
331 procedure (i.e. using the Parallel model for the thermal conductivity of the non-ice  
332 components, followed by Levy's model to account for the ice fraction) all provide  
333 predictions within, on average,  $\pm 20\%$ .

334

### 335 *3.3 Class III foods*

336 Suitable data for testing the predictions for porous non-frozen foods were difficult to  
337 obtain, since very often only minimal composition data are provided. In particular,  
338 bulk or apparent density or porosity data is often not available in the literature. Many  
339 of the data for which composition and temperature data were supplied were highly  
330 questionable, since they lay outside the Series and Parallel model bounds. Table 3  
331 shows the differences between the model predictions and experimental data for four  
332 different Class III foods where all of the following data were provided: porosity (or  
333 bulk density), moisture content, measurement temperature. Where these data were not  
334 supplied, the solids contents were assumed based on typical compositions for the  
335 particular food in question (Rahman, 2009). Table 3 shows that the multi-step  
336 procedure (i.e. using the Parallel model for the thermal conductivity of the non-ice  
337 components, followed by the EMT model to account for porosity) and the single-step

338 CC and EMT equations on average provide predictions of sufficient accuracy for first  
339 approximations, with the multi-step procedure providing the greatest accuracy.  
340 However, differences from individual measurements were sometimes greater than  
341 20%, which highlights the greater uncertainty involved in thermal conductivity  
342 prediction once porosity is introduced.

343

#### 344 *3.4 Class IV foods*

345 The number of examples of Class IV foods is relatively small, and the group is mainly  
346 comprised of frozen desserts. Of these, ice cream is probably the most widely studied  
347 in the food engineering literature, and the data from Cogné et al. (2003) were used  
348 since all the necessary data (composition, temperature, initial freezing temperature)  
349 were available. Table 4 shows the differences between the model predictions and  
350 experimental data at two different temperatures ( $-15\text{ }^{\circ}\text{C}$ , and  $-30\text{ }^{\circ}\text{C}$ ). In this instance  
351 the multi-step procedure (i.e. using the Parallel model for the thermal conductivity of  
352 the non-ice components, followed by Levy's model to account for the ice fraction,  
353 and the EMT model to account for porosity) has the clear advantage over the single-  
354 step models, and none of the single-step procedures provided predictions which are  
355 accurate to within  $\pm 30\%$ .

356 Note that while the average difference between the thermal conductivities predicted  
357 by the standard Multi-step procedure is within  $\pm 20\%$ , individual values are  
358 considerably higher (up to 50%). The difference between the predictions and the data  
359 increases as the porosity increases. This may be explained by the fact that the air  
360 remains dispersed as discrete bubbles even though the porosity of ice cream increases  
361 well beyond 0.3, and therefore the Maxwell-Eucken model with air as the dispersed

358 phase is the more suitable model of the porous structure than the EMT model, which  
359 assumes that air begins to form a continuous phase as the porosity increases. The final  
360 column in Table 4 shows the difference between experimental data and model  
361 predictions when the porosity is accounted for by the Maxwell Eucken model with air  
362 is the dispersed phase, rather than the EMT model. As expected the predictions are  
363 more accurate with the average difference being almost half of the average difference  
364 when the standard multi-step model using the EMT model is employed. Figure 6  
365 serves to further illustrate this point.

366

#### 368 **4. Discussion**

369 The results of the prediction comparison exercises show that for foods containing  
370 porosity, both frozen and unfrozen, the multi-step thermal conductivity prediction  
371 procedure proved to be the most accurate. The multi-step procedure also has the  
372 advantage over the single-step procedure in that while it can be employed without any  
373 knowledge of the structure of the food, there is scope for knowledge of the structure  
374 of the food to be incorporated into the method (as was illustrated in Section 3.4).  
375 Other than for the Class IV foods, the single-step Co-Continuous model provided, on  
376 average, prediction accuracies within the 'first approximation' range of  $\pm 20\%$  and is  
377 simpler to implement than the multi-step method.

378

379 On balance, the authors recommend that the multi-step procedure be used since it  
380 provided the greatest prediction accuracy over the range of foods considered, it has  
381 the scope for improving prediction accuracy by allowing for equations at each stage  
382 of the procedure to be changed if structural information about the food is known, and

383 yet it can also be used with reasonable confidence in the form presented here without  
384 any knowledge of the structure of the food.

385

## 386 **5. Conclusion**

387 Using only composition and initial freezing temperature data and knowledge of the  
388 food's temperature, a multi-step thermal conductivity prediction procedure provided  
389 the most accurate thermal conductivity predictions for the range of foods considered.

390 However, the single-step Co-Continuous model also provided predictions within  
391  $\pm 20\%$  other than for food containing both ice and air voids. On balance, however, the  
392 multi-step procedure is recommended for general use, since it provided the most  
393 accurate predictions over the widest range of foods, and also because there is scope  
394 for enhancements to be made within its framework, unlike the single-step method. It  
395 is emphasised that this methodology is intended for first approximations based on the  
396 minimum of input data, rather than as a rigorous modelling framework.

## 397 **Acknowledgments**

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490

## 491 NOMENCLATURE

492

- 493  $F$  intermediate variable (Eq. 7)
- 494  $G$  intermediate variable (Eq. 8)
- 495  $k$  thermal conductivity ( $\text{W m}^{-1} \text{K}^{-1}$ )
- 496  $T$  temperature ( $^{\circ}\text{C}$ )
- 497  $v$  volume fraction
- 498  $x$  mass fraction
- 499
- 500  $\delta$  difference between experimental value and model prediction
- 501  $\rho$  density ( $\text{kg m}^{-3}$ )
- 502  $v$  intermediate volume fractions

503

## 504 Subscripts

- 505 1 property of component 1

|     |                |   |
|-----|----------------|---|
| 506 | 2              | property of component 2                                       |
| 507 |                |   |
| 508 | <i>a</i>       | property of air   |
| 509 | <i>ash</i>     | property of ash   |
| 510 | <i>b</i>       | property of bound water                                       |
| 511 | <i>c</i>       | property of carbohydrate                                      |
| 512 | <i>cond</i>    | property of condensed (i.e. solid/liquid) phase               |
| 513 | <i>e</i>       | effective property  |
| 514 | <i>exp</i>     | experimental property   |
| 515 | <i>f</i>       | property of fat   |
| 516 | <i>F</i>       | initial freezing property                                     |
| 517 | <i>i</i>       | <i>i</i> th component   |
| 518 | I, II, III, IV | property relating the class of food as defined in Section 2.2 |
| 519 | <i>ice</i>     | property of ice   |
| 520 | <i>mod</i>     | property predicted by a model                                 |
| 521 | <i>p</i>       | property of protein   |
| 522 | <i>w</i>       | property of water   |

### 523 **Figure and Table Captions**

524 Table 1: Comparison of the differences between predicted and experimental thermal  
525 conductivity values for unfrozen, non-porous (Class I) foods at 20°C  
526 (experimental data from Willix, et al., 1998).

527 Table 2: Comparison of the differences between predicted and experimental thermal  
528 conductivity values for frozen, non-porous (Class II) foods at -20°C  
529 (experimental data from Willix, et al., 1998).

530 Table 3: Comparison of the differences between predicted and experimental thermal  
531 conductivity values for unfrozen, porous (Class III) foods [experimental data  
532 Sources: a - Baik et al. (1999), b - Rahman (2009), c - Houska et al. (1997), d -  
533 Houska et al. (1994), e - Carson et al. (2004), f - Muramatsu et al., (2008), g –  
534 Carson (2014), h - Carson and Kemp (2014)]

535 Table 4: Comparison of the differences between predicted and experimental thermal  
536 conductivity values for ice cream (Class IV food) at  $-15^{\circ}\text{C}$  and  $-20^{\circ}\text{C}$   
537 (experimental data from Cogné, et al., 2003)

538

539 Figure 1: Plots of the Series, Parallel, Geometric, EMT and Co-continuous models for a  
540 binary mixture in which  $k_1/k_2 = 20$

541 Figure 2 Plots of Series and Parallel models: 2a)  $k_1/k_2 = 3$ , 2b)  $k_1/k_2 = 12$ , 2c)  $k_1/k_2 = 25$ ,  
542 2d)  $k_1/k_2 = 100$ .

543 Figure 3: Schematic representation of the sequential approach for predicting the thermal  
544 conductivity of foods

545 Figure 4: Plots of the Maxwell-Eucken model with air as the dispersed phase (“ME1”),  
546 air as the continuous phase (“ME2”) plus the EMT, Co-Continuous (CC) and  
547 Levy models

548 Figure 5: Plots of the thermal conductivity predictions of Series, Parallel, Geometric,  
549 EMT and Co-Continuous models with experimental data for unfrozen, non-  
550 porous (Class I) foods

551 Figure 6: Plots of the thermal conductivity predictions of Series, Parallel, Geometric,  
552 EMT and Co-continuous models, standard Multi-step prediction method, and  
553 modified Multi-step prediction method with experimental data for ice cream  
554 (Class IV food)

|                    | $\delta$   |             |            |               |            |
|--------------------|------------|-------------|------------|---------------|------------|
|                    | Parallel   | Series      | Geometric  | Co-continuous | EMT        |
| Lean beef          | 0.2        | 11.6        | 1.3        | 0.3           | 2.0        |
| Beef mince         | 13.1       | 9.0         | 6.1        | 9.0           | 12.6       |
| Boneless chicken   | 5.4        | 6.8         | 4.1        | 5.1           | 8.2        |
| Pork Sausage meat  | 11.2       | 11.6        | 3.3        | 6.2           | 8.9        |
| Trim Pork Mince    | 13.8       | 4.2         | 9.6        | 11.4          | 15.2       |
| Veal mince         | 10.1       | 6.8         | 7.1        | 9.1           | 10.9       |
| Venison            | 5.2        | 6.5         | 4.3        | 5.3           | 7.9        |
| Lemon Fish fillets | 0.2        | 12.6        | 2.7        | 1.8           | 2.6        |
| Snapper fillets    | 5.0        | 5.7         | 4.2        | 5.0           | 7.7        |
| Tarakihi fillets   | 5.5        | 5.9         | 4.3        | 5.2           | 7.8        |
| Cheddar cheese     | 0.0        | 21.3        | 9.9        | 4.8           | 6.9        |
| Edam Cheese        | 0.9        | 21.5        | 9.6        | 4.6           | 5.1        |
| Mozzarella cheese  | 0.2        | 20.8        | 8.2        | 3.7           | 4.6        |
| <b>Average</b>     | <b>5.5</b> | <b>11.1</b> | <b>5.7</b> | <b>5.5</b>    | <b>7.7</b> |

Table 1: Comparison of the differences between predicted and experimental thermal conductivity values for unfrozen, non-porous (Class I) foods at 20°C (experimental data from Willix, et al., 1998).

|                    | $\delta$    |             |             |               |             |             |
|--------------------|-------------|-------------|-------------|---------------|-------------|-------------|
|                    | Parallel    | Series      | Geometric   | Co-continuous | EMT         | Multi-step  |
| Lean beef          | 29.4        | 59.7        | 11.0        | 16.0          | 13.3        | 3.0         |
| Beef mince         | 87.7        | 49.6        | 10.0        | 14.6          | 48.9        | 32.3        |
| Boneless chicken   | 33.8        | 58.5        | 8.2         | 13.4          | 17.1        | 6.4         |
| Pork Sausage meat  | 48.8        | 57.7        | 13.4        | 7.0           | 15.0        | 5.3         |
| Trim Pork Mince    | 94.1        | 43.0        | 25.5        | 23.0          | 64.2        | 47.5        |
| Veal mince         | 71.1        | 51.3        | 7.5         | 7.0           | 42.3        | 27.2        |
| Venison            | 39.5        | 56.2        | 3.7         | 9.3           | 22.3        | 11.3        |
| Lemon Fish fillets | 42.2        | 55.4        | 0.9         | 7.5           | 25.5        | 14.4        |
| Snapper fillets    | 36.7        | 54.3        | 2.1         | 8.8           | 21.9        | 11.8        |
| Tarakihi fillets   | 37.6        | 54.4        | 2.1         | 8.5           | 22.3        | 12.0        |
| Cheddar cheese     | 22.7        | 58.2        | 37.3        | 17.5          | 29.1        | 20.1        |
| Edam Cheese        | 13.8        | 63.1        | 42.3        | 24.9          | 33.0        | 25.9        |
| Mozzarella cheese  | 33.4        | 62.0        | 33.9        | 16.5          | 18.1        | 15.3        |
| <b>Average</b>     | <b>45.4</b> | <b>55.6</b> | <b>15.2</b> | <b>13.4</b>   | <b>28.7</b> | <b>17.9</b> |

Table 2: Comparison of the differences between predicted and experimental thermal conductivity values for frozen, non-porous (Class II) foods at  $-20^{\circ}\text{C}$  (experimental data from Willix, et al., 1998).

|                                 | $\delta$    |             |             |             |               |             |             |
|---------------------------------|-------------|-------------|-------------|-------------|---------------|-------------|-------------|
|                                 | $v_a$ range | Parallel    | Series      | Geometric   | Co-continuous | EMT         | Multi-step  |
| Cup cake <sup>a</sup>           | 0.29 - 0.83 | 32.1        | 59.4        | 34.4        | 15.0          | 27.6        | 24.6        |
| Defatted soy flour <sup>b</sup> | 0.54 - 0.72 | 59.7        | 43.4        | 13.2        | 11.1          | 5.9         | 7.8         |
| Dried beef <sup>c</sup>         | 0.67 - 0.73 | 25.4        | 46.8        | 26.6        | 8.1           | 18.5        | 17.5        |
| Milk powder <sup>d</sup>        | 0.40 - 0.43 | 17.8        | 64.5        | 43.5        | 28.2          | 19.8        | 15.1        |
| Model food <sup>e</sup>         | 0.04 - 0.65 | 10.8        | 75.4        | 41.0        | 39.3          | 19.2        | 19.2        |
| Rice <sup>f</sup>               | 0.36 - 0.41 | 22.4        | 62.3        | 28.9        | 20.9          | 13.4        | 2.7         |
| Sponge Cake <sup>g</sup>        | 0.45 - 0.61 | 40.0        | 64.7        | 35.0        | 16.7          | 18.9        | 10.5        |
| Sucrose powder <sup>h</sup>     | 0.44 - 0.51 | 34.8        | 48.4        | 8.2         | 5.1           | 1.2         | 3.1         |
| <b>Average</b>                  |             | <b>28.0</b> | <b>58.6</b> | <b>31.3</b> | <b>20.4</b>   | <b>17.4</b> | <b>14.5</b> |

Sources: a - Baik et al. (1999), b - Rahman (2009), c - Houska et al. (1997), d - Houska et al. (1994), e - Carson et al. (2004), f - Muramatsu et al., (2008), g - Carson (2014), h - Carson and Kemp (2014)

Table 3: Comparison of the differences between predicted and experimental thermal

conductivity values for unfrozen, porous (Class III) foods [experimental data Sources: a - Baik et al. (1999), b - Rahman (2009), c - Houska et al. (1997), d - Houska et al. (1994), e - Carson et al. (2004), f - Muramatsu et al., (2008), g - Carson (2014), h - Carson and Kemp (2014)]

| Porosity       | $\delta$    |             |             |               |             |             |             |
|----------------|-------------|-------------|-------------|---------------|-------------|-------------|-------------|
|                | Parallel    | Series      | Geometric   | Co-continuous | EMT         | Multi-step  |             |
|                |             |             |             |               |             | Standard    | Max-Euck    |
| -15 °C         |             |             |             |               |             |             |             |
| 0.13           | 51.4        | 84.9        | 33.4        | 39.4          | 8.4         | 4.5         | 5.8         |
| 0.23           | 62.3        | 88.2        | 44.6        | 43.8          | 2.4         | 3.8         | 8.7         |
| 0.33           | 70.0        | 89.6        | 54.2        | 45.5          | 9.8         | 2.1         | 9.4         |
| 0.41           | 73.2        | 90.1        | 60.8        | 46.2          | 23.8        | 11.3        | 8.2         |
| 0.46           | 72.1        | 90.3        | 64.8        | 46.9          | 34.6        | 19.9        | 5.7         |
| 0.6            | 102.6       | 88.1        | 67.1        | 36.2          | 54.2        | 38.3        | 18.9        |
| 0.67           | 105.5       | 86.8        | 69.1        | 32.8          | 64.0        | 53.9        | 18.4        |
| <b>Average</b> | <b>76.7</b> | <b>88.3</b> | <b>56.3</b> | <b>41.5</b>   | <b>28.2</b> | <b>19.1</b> | <b>10.7</b> |
| -30 °C         |             |             |             |               |             |             |             |
| 0.13           | 60.4        | 86.4        | 32.8        | 40.3          | 15.8        | 7.0         | 8.5         |
| 0.23           | 63.5        | 89.9        | 47.5        | 47.4          | 3.8         | 1.1         | 6.0         |
| 0.33           | 75.5        | 90.9        | 56.1        | 47.8          | 7.0         | 2.5         | 9.3         |
| 0.41           | 73.7        | 91.6        | 63.9        | 49.9          | 24.8        | 14.4        | 4.9         |
| 0.46           | 77.7        | 91.5        | 66.8        | 49.1          | 34.5        | 20.7        | 5.4         |
| 0.6            | 115.6       | 89.2        | 68.6        | 36.9          | 55.8        | 38.6        | 22.2        |
| 0.67           | 104.6       | 88.8        | 72.6        | 37.7          | 68.5        | 58.4        | 13.7        |
| <b>Average</b> | <b>81.6</b> | <b>89.7</b> | <b>58.3</b> | <b>44.2</b>   | <b>30.0</b> | <b>20.4</b> | <b>10.0</b> |

Table 4: Comparison of the differences between predicted and experimental thermal conductivity values for ice cream (Class IV food) at -15°C and -20°C (experimental data from Cogné, et al., 2003)

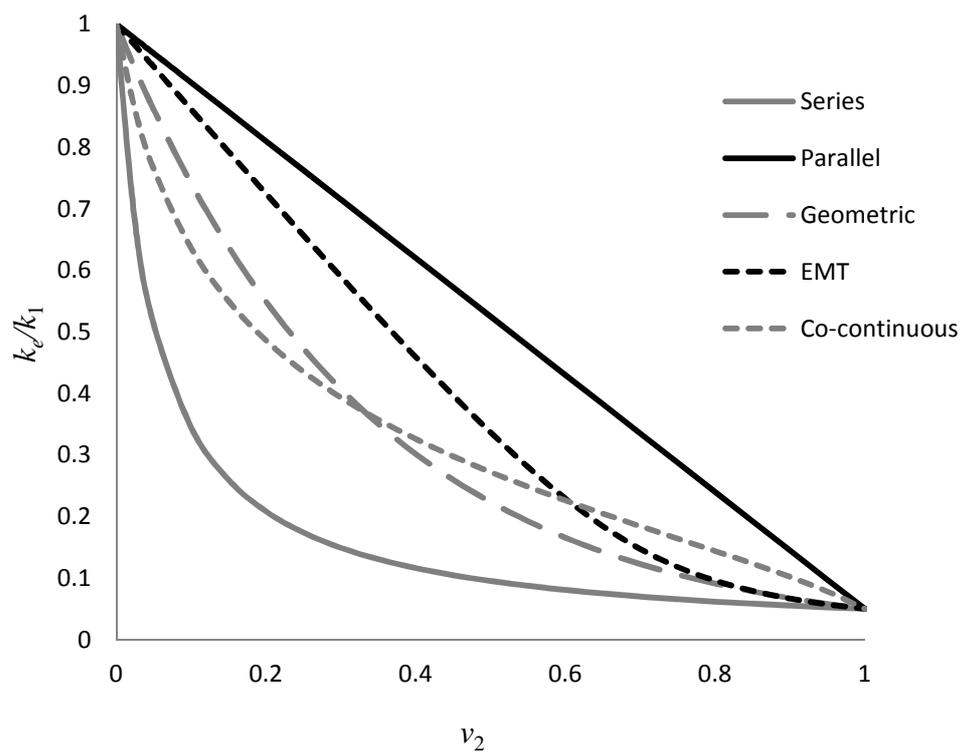


Figure 1: Plots of the Series, Parallel, Geometric, EMT and Co-continuous models for a binary mixture in which  $k_1/k_2 = 20$

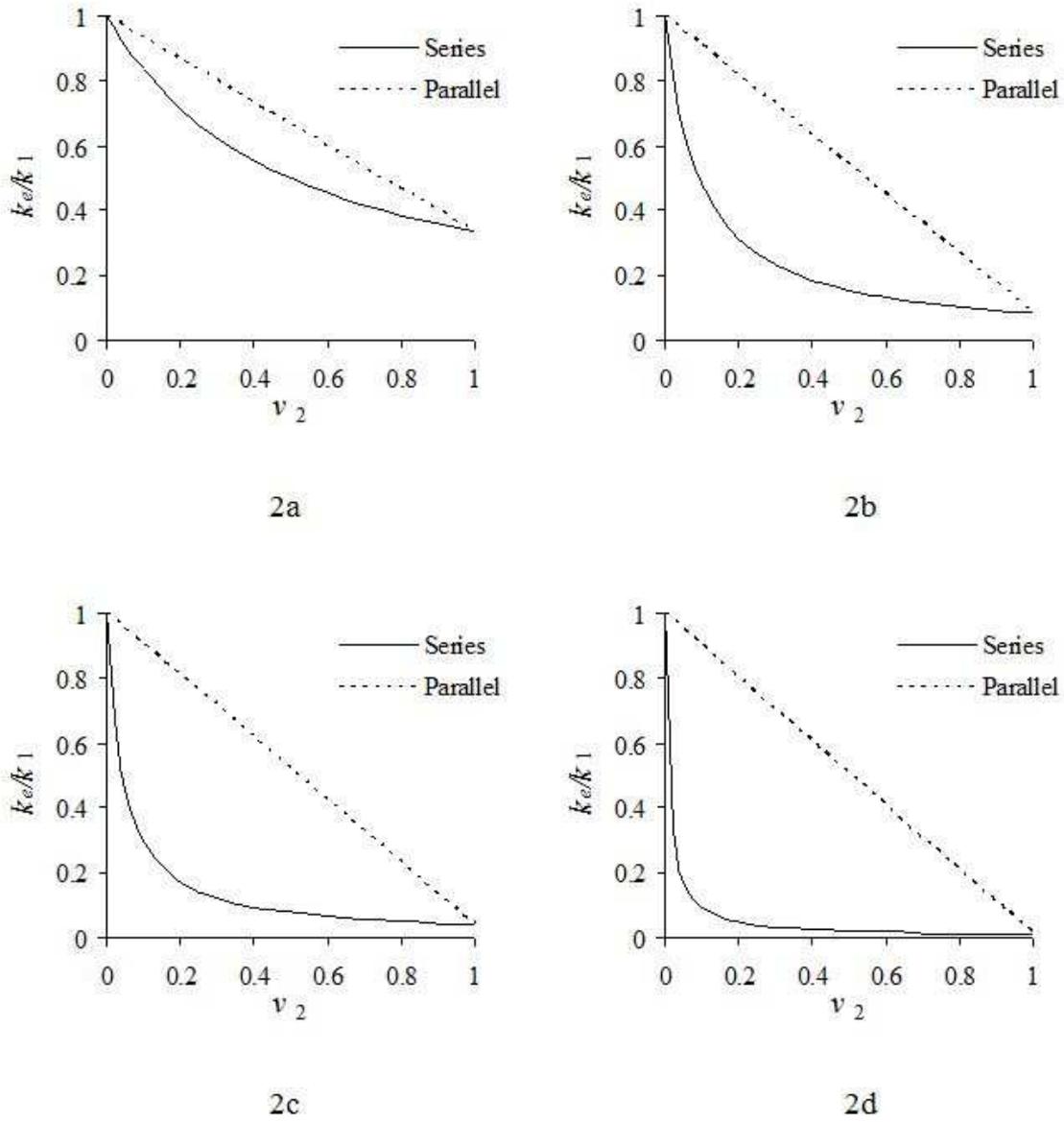


Figure 2 Plots of Series and Parallel models: 2a)  $k_1/k_2 = 3$ , 2b)  $k_1/k_2 = 12$ , 2c)  $k_1/k_2 = 25$ , 2d)  $k_1/k_2 = 100$ .

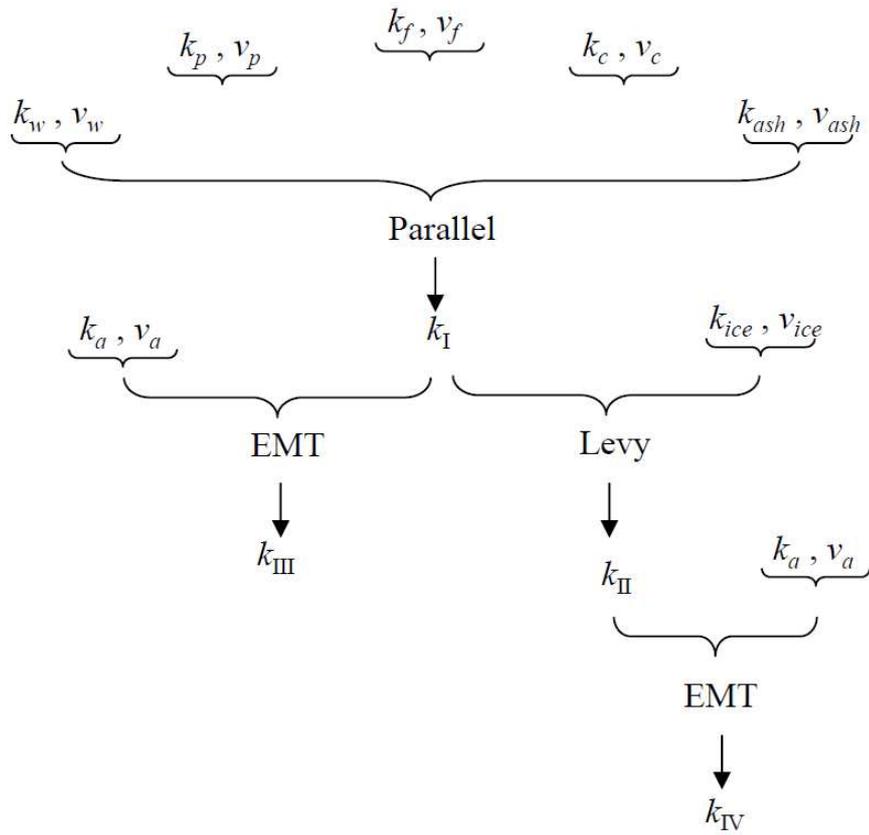


Figure 3: Schematic representation of the sequential approach for predicting the thermal conductivity of foods

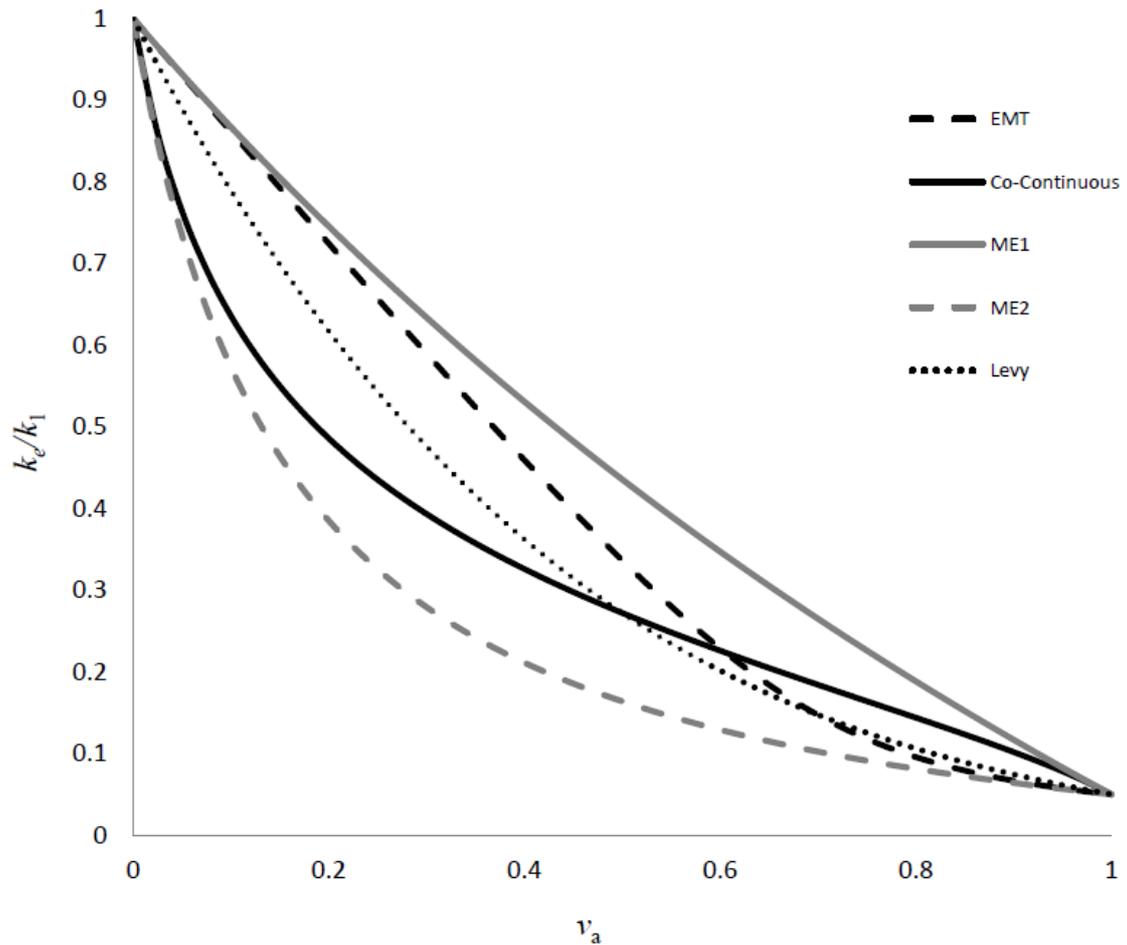


Figure 4: Plots of the Maxwell-Eucken model with air as the dispersed phase (“ME1”), air as the continuous phase (“ME2”) plus the EMT, Co-Continuous (CC) and Levy models

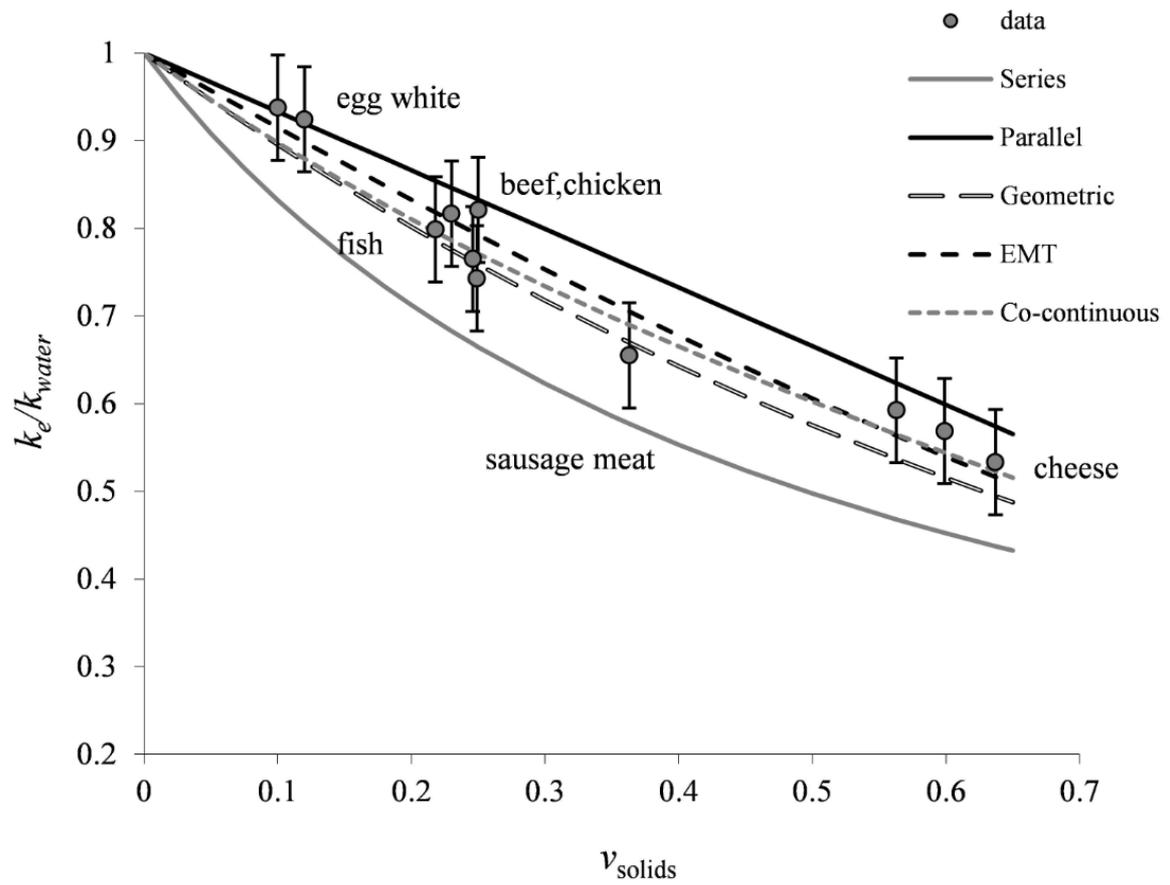


Figure 5: Plots of the thermal conductivity predictions of Series, Parallel, Geometric, EMT and Co-Continuous models with experimental data for unfrozen, non-porous (Class I) foods

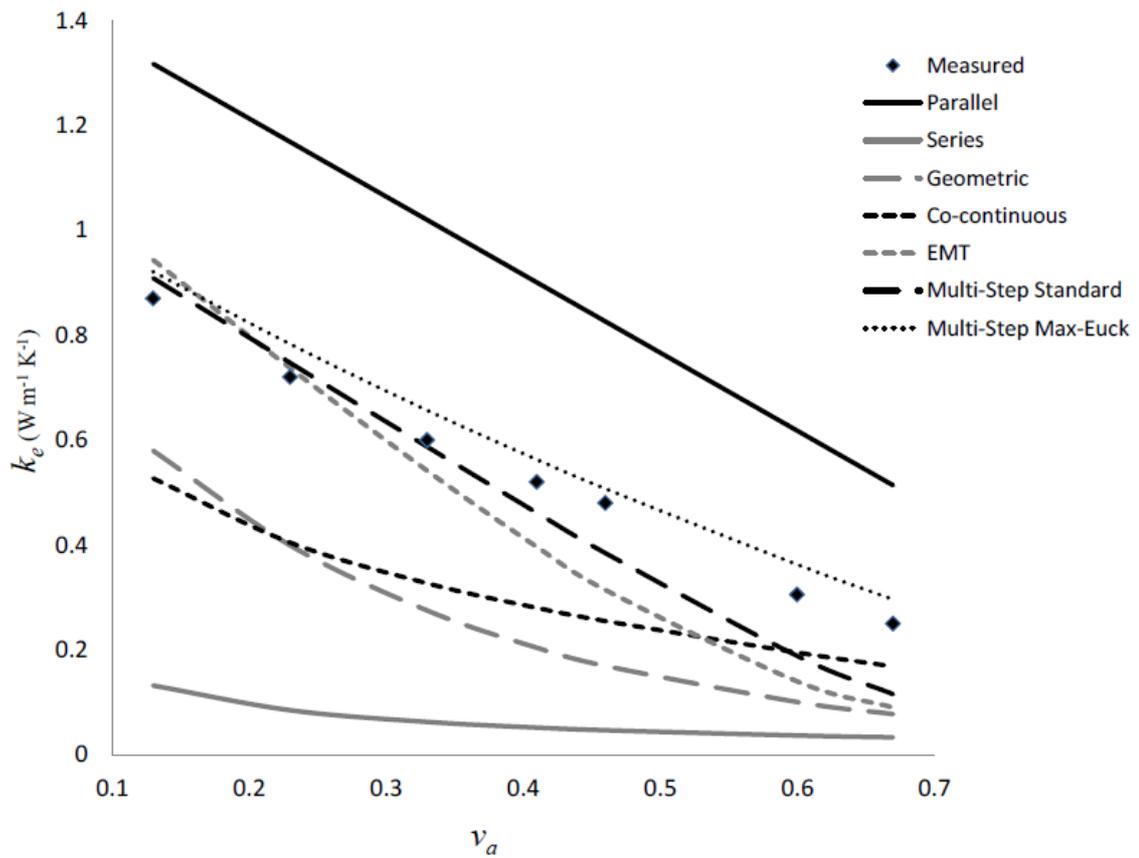


Figure 6: Plots of the thermal conductivity predictions of Series, Parallel, Geometric, EMT and Co-continuous models, standard Multi-step prediction method, and modified Multi-step prediction method with experimental data for ice cream (Class IV food)

- Different methods for predicting thermal conductivity of foods solely from composition and temperature data were compared against measured data
- Multi-step procedure involving sequential application of Parallel, Levy and Maxwell-Eucken model provided most accurate predictions on average
- Other than for frozen, porous foods, the Co-continuous models also provided predictions within  $\pm 20\%$  on average

ACCEPTED MANUSCRIPT