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Hydrothermal carbonization of primary sewage sludge and synthetic faeces: Effect of reaction temperature and time on filterability

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1 **Hydrothermal Carbonisation of Primary Sewage Sludge and Synthetic**
2 **Faeces: Effect of Reaction Temperature and Time on Filterability**

3
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13
14 **ABSTRACT**

15 The effect of reaction temperature and time on the filterability of slurries of primary sewage
16 sludge (PSS) and synthetic faeces (SF) following hydrothermal carbonisation (HTC) was
17 investigated and optimised using response surface methodology (RSM). Filterability was
18 shown to improve as the treatment temperature and reaction time at which the solids were
19 carbonised was increased. The best filtration results were achieved at the highest temperature
20 (200°C) and longest treatment time (240 min) employed here. The specific cake resistance to
21 filtration of the carbonised solids was found to vary between 5.43×10^{12} and 2.05×10^{10} m
22 kg^{-1} for cold filtration of PSS, 1.11×10^{12} and 3.49×10^{10} m kg^{-1} for cold filtration of SF, and
23 3.01×10^{12} and 3.86×10^{10} m kg^{-1} for hot filtration of SF, and decreased with increasing
24 reaction temperature and time for carbonisation. There was no significant difference in the
25 specific resistance of cold and hot filtration for SF. The RSM models employed here were
26 found to yield predictions that were close to the experimental results obtained and should
27 therefore prove useful in designing and optimizing HTC filtration systems for generating
28 solids for a wide variety of end uses.

29
30 **Keywords:** Sludge Treatment, Dewaterability; Filtration; Hydrochar; Specific Cake
31 Resistance.

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Nomenclature

- A cross sectional area (m)
 C_w solid concentration by weight fraction (w/w)
 c dry mass of solids per unit volume (kgm^{-3})
M mass of slurry filtered (g)
Q volumetric rate of flow (m^3)
 R_m medium resistance (m^{-1})
s mass fraction of solids in slurry (g)
 s_f solids in filtrate (% wt.)
T reaction temperature ($^{\circ}\text{C}$)
t filtration time (s)
 t_R reaction or treatment time (min)
V volume of filtrate (m^3)

Greek letters

- α specific cake resistance (mkg^{-1})
 ΔP pressure drop (Pa)
 μ liquid viscosity (Pa s)

Abbreviations

- CCRD central composite rotatable design
HTC hydrothermal carbonisation
PSS primary sewage sludge
RSM response surface methodology
SF synthetic faeces

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74

75 **INTRODUCTION**

76

77 The treatment and disposal of sewage sludge presents a number of environmental challenges
78 [1–3]. Current practice is either to dispose of treated sludges to agricultural land [4], or either
79 to incinerate [5] or landfill [6] them. Hydrothermal carbonisation (HTC) is a promising
80 method of processing sewage sludge that results in the formation of a stable and sanitised
81 product [7]. HTC does not require a dry feedstock, which makes it especially suitable for
82 naturally wet biomass [8,9]. The process is self-contained and hence emission of greenhouse
83 gases (GHG) is minimal. Indeed its implementation could actually result in GHG mitigation
84 if the carbonized solids are used as soil conditioner [7,10]. The carbonaceous solid produced
85 by HTC is commonly referred to as ‘hydrochar’, and in addition to this product an aqueous
86 fraction is formed along with a small quantity of gas. Carbonisation is typically achieved at
87 moderate temperatures (180–250°C) under saturated pressures [9,11]. The hydrochar, which
88 is the main product, has H/C and O/C ratios similar to that of coal and a heating value
89 comparable to low rank coal [12] thus enabling it to be used as fuel. The hydrochar is a
90 carbon-rich compound that is slow to oxidise, and it can be added to soil to sequester carbon
91 that would otherwise be released into the atmosphere as GHG. Moreover, it has been shown
92 to improve soil fertility [8]. Other options for the solids produced include biofuel production
93 by transesterification [13], and gasification for syngas production [14]. These options require
94 different ratios of O/C for optimal operation, giving rise to different reaction temperatures
95 and time requirements during HTC. After processing, the carbonised slurry needs to be
96 dewatered to separate the hydrochar from the liquid product irrespective of its final use.

97

98 Previous work on biomass carbonisation [9,11,15] has tended to focus on the chemistry
99 and mechanisms of the HTC process, and how it can be used to produce various solid
100 products for carbon sequestration, as an energy source or for other purposes [16–19].

101 However, efficient recovery of the hydrochar has hitherto not received attention, and hence, a
102 more comprehensive study into HTC-slurry dewaterability is required in order to identify the
103 optimum conditions and to determine the filtration properties of the dewatered products.

104 Such data would facilitate proper design and scale-up of filtration systems which may operate
at different conditions of temperature and time, depending on the required solids to be

105 produced. Yukseler et al. [20] proposed a model for filterability of sludge, but this is of
106 limited usefulness in this context as the slurry used was not thermally treated, and its
107 filtration characteristics would be markedly different from that of thermally treated sludges.
108 Ramke et al. [21] studied the dewatering properties of various organic wastes (municipal
109 waste, agricultural residues, etc.) following HTC. However, they did not optimise the process
110 and therefore it is difficult to identify the conditions for best filterability. Finally, Yanagida et
111 al. [22] undertook studies to predict the viscosity of sewage sludge following hydrothermal
112 carbonisation over a range of conditions. However, their study did not extend to the
113 dewatering of the end product.

114 The objectives of the work described here were to investigate the effects of HTC process
115 temperature and treatment times on filterability of HTC-slurry, as well as the interaction
116 effects among these operating conditions. Also, the influence of hot filtration on filterability
117 was investigated for primary sewage sludge as well as a standard faecal simulant. The
118 operating conditions for achieving the optimise filterability was examined using response
119 surface methodology (RSM) models.

120

121 **MATERIALS AND METHODOLOGY**

122 **Primary Sewage Sludge and Faecal Simulant**

123 Primary sewage sludge (PSS), was collected from Wanlip Sewage Treatment Works
124 (Leicestershire, UK). PSS primarily comprises faecal matter removed by settlement and
125 typically contained 4.3% (wt.) solids as received. A synthetic faeces (SF) was prepared using
126 the formulation proposed by Wignarajah et al. [23]. The solids comprised cellulose (37.5%),
127 yeast (37.5%), peanut oil (20%), KCl (4%), $\text{Ca}(\text{H}_2\text{PO}_4)_2$ (1%) (all purchased from Sigma–
128 Aldrich, UK), and tap water – which constituted 90% (wt.) of the SF. The materials were
129 uniformly mixed with water to form a suspension with 10% solids. These were prepared
130 immediately before conducting the carbonisation tests.

131

132 **Hydrothermal Carbonisation**

133 About 150 ml of PSS, as received, was subjected to HTC using a 250 mL stainless steel
134 batch reactor (BS1506-845B, UK) immersed in an oil bath (B7 Phoenix II, Thermo
135 Scientific, UK) containing Shell Thermia oil B (Shell, UK). The PSS was periodically stirred
136 manually to prevent settling prior to carbonisation. HTCs were performed at reaction
137 temperatures of 140, 160, 180, and 200°C. HTC at 140°C was carried out for 4 h; 160°C for
138 1, 2 and 4 h; 180°C for 0.5, 1, 2 and 4 h; and 200°C for 0.25, 0.5, 1, 2 and 4 h. Further details
139 of the HTC process may be found in a previous study [24], which reported the reaction
140 kinetics and equilibrium solids mass remaining. The temperature range used here was
141 selected so as to minimise energy requirement for the HTC system in order that the process
142 would remain economical for developing countries. HTC of SF was performed at the same
143 reaction temperatures and treatment times using a pilot scale semi-batch HTC reactor suitable
144 for cold and hot filtration tests. Hot filtration tests were not possible for the batch PSS reactor
145 system.

146

147 **Filterability**

148 *Cold Filtration of PSS*

149 After HTC had been completed, the reactor was cooled to about 25°C and the gaseous
150 phase vented. The carbonised slurry (about 150 ml) was transferred to a clear filtration cell
151 with a total volume of 300 ml as represented in Figure 1 (a), and connected to a vacuum
152 pump. Filtration was conducted using a metal microporous filter medium, with 10 µm slotted
153 pores and nominal thickness of 0.06 mm, manufactured by Micropore Technologies Ltd.
154 (Hatton, Derbyshire, UK). The vacuum was applied by means of a vacuum pump (CAPEX
155 L2C, Charles Austen Pumps Ltd, UK) and the filtrate was collected in a measuring cylinder
156 placed on an electronic balance (TP2KV, OHAUS Corporation, USA). The weight of the

157 filtrate with respect to time was recorded using a PC customised Labview software. In all the
158 experiments, the vacuum pressure was set to 58.6 kPa using a needle valve. The filter
159 medium was cleaned after each experiment with a detergent using a sonic bath, prior to reuse
160 in further tests.

161 162 *Cold and hot Filtration of SF*

163 After HTC, the reactor was allowed to cool down to 25°C or flashed to 100°C for cold
164 and hot filtration, respectively. Valve-1 was opened to transport the carbonised slurry (about
165 450 ml) to the filtration cell (internal diameter of 55 mm and total volume of 535 ml)
166 connected to the reactor as shown in Figure 1 (b). The pressure in the cell and reactor was
167 adjusted to obtain a filtration pressure between 61.0–78.7 kPa. Filtration was started
168 immediately by opening Valve-2 and the weight of the filtrate with respect to time was
169 recorded. The material was filtered using the same filter medium, electronic balance and PC
170 customised with Labview software as explained in cold filtration of PSS.

171 172 **Analysis of Total Solids in Filtrate**

173 Total solids leached into the filtrate during HTC and filtration were analysed according
174 to EPA method, 1684, by heating about 25 g of each sample in an oven at $103 \pm 2^\circ\text{C}$ for 1 h
175 to dry. Total solids in weight percent, was calculated as the percentage weight difference
176 before and after oven drying [25]. All analyses were performed in triplicate.

177 178 179 **Analysis of Total Organic Carbon in Filtrate**

180 Total organic carbon (TOC) in the filtrate was analysed using a TOC analyser (DC-190,
181 Rosemount Dohrmann, USA), according to Standard Methods 5310 B – High Temperature
182 Combustion Method [26]. The analysis was carried out in two ways: first, the filtrates were
183 directly analysed for TOC. In the second test, the filtrates were further filtered using filter

184 paper (Whatman Grade 2) to obtain suspended and colloidal/dissolved solids. The samples
185 were diluted to a ratio of 1:10, and a 50 μL aliquot injected in to the instrument chamber to
186 analyse total and inorganic carbon. The TOC was obtained by subtraction of inorganic carbon
187 (if any) from the total carbon. Triplicate measurements were made, and the average and
188 standard deviations calculated.

189 **Experimental Design**

191 Response surface methodology (RSM) using a central composite rotatable design
192 (CCRD) was used to study the influence of the two variables (reaction temperature, and
193 retention time) and their interaction on the filterability of HTC-slurry. Design Expert 9.0.1
194 software (Statease, Minneapolis, MN) for the CCRD and statistical analysis of variance
195 (ANOVA), which resulted in 13 tests (4 factorial points, 4 star points, and 5 central points).
196 Each variable was set at 5 levels: $-\alpha$, -1 , 0 , 1 and α (where $\alpha = 1.414$).

197 **RESULTS AND DISCUSSION**

199 **Filtrate Volume**

200 Figures 2 and 3 show that, for both SF and PSS, the amount of liquid recovered
201 following HTC increased as the reaction temperature and treatment time were increased.
202 Higher volumes of filtrate were obtained at shorter filtration times from slurries carbonised at
203 higher temperatures for longer retention times. For instance, about 125 ml of filtrate was
204 obtained in 1 min from cold filtration of PSS slurry carbonised at 200°C for 240 min,
205 compared with about 17 ml obtained in 1 min for HTC at 200°C for 15 min (see Figure 3-d).
206 Hot filtration of SF was faster than cold filtration of SF. For example, within 10 s of hot
207 filtration, 170 and 196 ml of filtrate were obtained for slurries from HTC at 180°C and 200°C
208 both for 240 min respectively, compared with about 70 ml obtained for cold filtration in both
209 cases. Due to differences in size of the HTC reactors used here for filtration of PSS, 150 ml

210 of slurry (containing about 145 ml of liquid) was transferred to the filtration cell, whereas 450
 211 ml of slurry (with about 428 ml of liquid) was transferred to the filtration cell for SF (cold
 212 and hot) filtration. For SF, in order to achieve a minimum filtration pressure that would
 213 enable measurement of the filtrate volume per time, the HTC reactor was flashed: i.e. the
 214 over-pressure required to prevent the water boiling during the HTC process was rapidly
 215 released, resulting in a process fluids temperature of approximately 100°C being rapidly
 216 achieved.

217 The linear equation represented by Eq. (4) gives the t/V versus V plots presented in
 218 Figure 4. The plots illustrate typical sets of data obtained from a constant pressure filtration
 219 method. For slurries from HTC at 200°C, good linearity was obtained for PSS and SF (cold
 220 filtered, and hot filtered) at all treatment times (15–240 min). At lower temperatures and also
 221 shorter treatment times, the slope of the t/V versus V plots increased rapidly (as shown in
 222 Figure 4-a, -b, and -c). This occurred when the filtration behavior was poor, due to poorly
 223 filtering solids and/or filter medium blockage.

224
 225 **Filtration Resistance**

226 During filtration a porous cake builds up on the porous filter medium and further
 227 filtration takes place at the top surface of that cake. By applying Darcy's law
 228 the pressure drop across the filter medium is:

229
$$\Delta P = \left(\frac{\mu R}{A}\right) \left(\frac{dV}{dt}\right) \quad (3)$$

230 Addition of the medium and filter cake pressure drop provides the classical linear
 231 equation for constant pressure filtration:

232
$$\frac{t}{V} = \frac{\mu \alpha c}{2A^2 \Delta P} (V) + \frac{\mu R_m}{A \Delta P} \quad (4)$$

233 R_m and α , are the resistance of the filter medium and filter cake, and may be evaluated from
 234 the intercept and slope of the t/V vs. V plot, respectively.

235 Cake concentration in terms of weight fraction is obtained by dividing the mass of dry
236 cake by the mass of wet cake.

237 Results from the linear equation represented by Eq. (4), which is the conventional
238 method for determining specific cake resistance to filtration, from constant pressure filtration
239 are presented in Figure 4. Theoretically, the plot of t/V versus V gives a straight line (see
240 Figure 4) with

$$241 \quad \text{slope} = \frac{\mu\alpha c}{2A^2(\Delta P)} \quad (5)$$

242 and

$$243 \quad \text{intercept} = \frac{\mu R_m}{A(\Delta P)} \quad (6)$$

244 For cold filtration of PSS and SF, the viscosity of water at 25°C was used, whilst that at
245 100°C was used for hot filtration of SF in both Eqs. (5) and (6). The dry cake mass per unit
246 volume of filtrate at the end of the filtration, c was calculated from knowledge of the filtrate
247 volume (V) and the mass fraction of solids in the slurry (s) and mass of slurry filtered (M):

$$248 \quad c = \frac{sM}{V} \quad (7)$$

249 Specific cake resistance is a measure of filterability or dewaterability; the lower the
250 specific resistance, the greater the dewaterability of a slurry [27]. As shown in Figure 5,
251 values of specific cake resistance to filtration decreased as HTC temperature and reaction
252 time increased. For PSS, filterability is greater for slurry from HTC at 200°C for 240 min
253 treatment time ($\alpha = 2.05 \times 10^{10} \text{ m kg}^{-1}$), though the slurry from 15 min treatment time filtered
254 fairly well ($\alpha = 7.62 \times 10^{11} \text{ m kg}^{-1}$). The filterability of PSS slurry from HTC at 180°C
255 ranged from 3.32×10^{11} – $1.55 \times 10^{11} \text{ m kg}^{-1}$ for treatment times between 120–240 min
256 respectively, but no filter cake was formed between 30–60 min treatment times. Except for
257 HTC at 160°C and 240 min ($\alpha = 5.43 \times 10^{12} \text{ m kg}^{-1}$), cake filtration of PSS slurries from HTC
258 at lower temperatures were not possible, as no filter cakes were formed. As a result, values

259 of specific cake resistance in these cases were not included in Figure 5 (a). Specific cake
260 resistance to filtration of raw sludge is reported to vary from $1.0\text{--}2.9 \times 10^{14} \text{ m kg}^{-1}$ [28].
261 Values of specific cake resistance to filtration exceeding $1.0 \times 10^{12} \text{ m kg}^{-1}$ indicate poor
262 filterability [29]. The poor filterability of slurries from HTC at lower temperatures indicated
263 that the slurries were not well carbonised. In a previous study using identical feedstocks it
264 was shown that conversion of solids to hydrochar is less favoured at lower temperatures and
265 also for shorter retention times [24]. The carbonised cake has a fibrous porous structure with
266 increased porosity. Previous studies indicate that hydrothermal treatment affects the structure
267 of the carbonised solids, and the effect depends on the treatment temperature [30,31]; even
268 though none of the studies relate this to filterability of the end product. The disruption of the
269 colloidal structure and increase in porosity of the carbonised cake accounted for the greater
270 filterability especially at higher temperatures.

271 Specific cake resistance for cold and hot-filtered SF are comparable, especially for slurry
272 from HTC at 200°C ; with values ranging from $8.14\text{--}3.49 \times 10^{10} \text{ m kg}^{-1}$ and $3.08\text{--}3.86 \times 10^{10}$
273 m kg^{-1} for cold and hot filtration respectively, between 15–240 min treatment times. The
274 viscosity of the liquid affects the rate at which the filtrate permeates through the filter and the
275 cake. As the temperature increased, the viscosity of the liquid fraction decreases, and
276 consequently the overall resistance to filtration of hot-filtered SF slurry should be lower than
277 that of cold-filtered SF. However, there was no significant difference between hot and cold
278 filtration of SF on the resulting specific resistance; although hot filtration was observed to be
279 faster as would be expected by a lower viscosity filtrate. Specific cake resistance to filtration
280 of hot-filtered SF slurries from HTC at 160°C and treatment times between 60–120 min were
281 between $1.58 \times 10^{12}\text{--}3.01 \times 10^{12} \text{ m kg}^{-1}$; similar to that obtained for cold-filtered SF
282 feedstock ($1.32 \times 10^{12} \text{ m kg}^{-1}$), the calculation for specific resistance takes in to account
283 different liquid viscosities.

284 Resistance of the filter medium was higher at lower HTC temperatures and also higher
285 temperatures and shorter treatment times; and decreased as HTC temperature and time
286 increased (Figure 6). There was higher resistance of the filter medium during cold filtration
287 of PSS slurry than cold filtered SF. This may be due to differences in the characteristics of
288 the two feedstock materials. Resistance of the filter medium for HTC at 200°C and 15–240
289 min treatment times ranged between 3.33×10^{10} – $2.78 \times 10^{10} \text{ m}^{-1}$, 5.65×10^9 – $9.91 \times 10^9 \text{ m}^{-1}$,
290 1.47×10^{10} – $0.39 \times 10^{10} \text{ m}^{-1}$ for cold filtration of PSS, cold, and hot filtration of SF
291 respectively. The higher medium resistance of hot-filtered SF slurry than that of cold-filtered
292 SF slurry may have been caused by fouling of the filter surface with the hazy liquid of the
293 former containing more solids in the filtrate which is explained in the subsequent section.

294

295 **Solids Leached into Filtrate**

296 Generally, very small amounts of solids were found in the filtrate since the filtration was
297 conducted on an open-slotted filter medium. However, filtration of slurry carbonised at
298 higher HTC temperature and longer treatment time resulted in less solids leached into the
299 filtrate, with decreasing solids as HTC temperature and time increased (see Table 1). More
300 solids were found in the filtrate when the SF was hot-filtered than when it was cold-filtered.
301 Total solids in the filtrate ranged between 1.92–1.68 % (wt.) and 2.83–1.98 % (wt.) for cold
302 and hot-filtration of SF between HTC temperatures of 160–200°C, respectively. In effect,
303 filtrate from hot-filtered SF was hazy, because it contain more colloidal soluble components
304 compared with that from cold-filtered SF slurry. This would result in the higher specific cake
305 resistance to filtration obtained from hot filtration of SF slurry than cold filtered slurry;
306 especially at HTC temperatures between 160°C and 180°C (see Figure 5-b and -c). As
307 explained in Section 3.2, specific cake resistance to filtration for slurries from HTC at 160°C
308 and 60 – 120 min treatment times (3.01×10^{12} – $1.58 \times 10^{12} \text{ m kg}^{-1}$) were similar to that
309 obtained for SF feedstock ($1.32 \times 10^{12} \text{ m kg}^{-1}$).

310

311 **Total Organic Carbon in Filtrate**

312 Table 1 shows the effect of treatment temperature and time on total organic carbon
313 (TOC) in the filtrate following cold filtration of carbonised PSS slurries. As explained in the
314 TOC analysis, the test was performed in two ways: first, the filtrates were directly analysed
315 following filtration with the 10 μm slotted filter medium. In the second test, the filtrates were
316 further filtered using Whatman filter paper. The TOC from the first tests appeared to vary
317 randomly. This may be due to residual colloidal carbon that may have passed through the
318 slotted filter and was detected by the TOC analyzer. A micro needle was used to take filtrate
319 samples for analysis; any form of colloidal particles taken up by the needle that is analysed
320 would cause significantly fluctuating results. TOC determined after the filtrates were further
321 filtered (second tests) was more consistent and increased systematically as the treatment
322 temperature and time were increased (see Table 1). The subsequent filtration ensured
323 complete removal of suspended and colloidal solids in the filtrates; hence TOC reported here
324 is solely that of dissolved solids. The increase in TOC as the treatment temperature and time
325 increased is due to increasing dissolved carbon into the filtrate as the process severity
326 increased.

327

328 **Cake Concentration**

329 Figure 7 shows the effect of reaction temperature and time on cake concentration by
330 weight for cold filtered PSS, and cold and hot filtered SF. Cake concentration was highest at
331 higher reaction temperatures and longer treatment times, and increased as the temperature
332 and time increased. Hot filtration of SF produced the driest cake (i.e. highest cake
333 concentration) with values between 35–58 % (w/w), followed by cold filtration of SF (26–45
334 % w/w), and that for cold filtration of PSS varying between 14–27 % (w/w). For PSS,
335 filtration was not possible for slurries carbonised at the lower reaction temperatures (140–
336 160°C, except that for 160°C and 240 min treatment times), and 180°C at shorter treatment
337 times (30 and 60 min) as no cake was formed during the filtration on the slotted 10 μm filter.
338 A high cake solids content is desired to stabilise the hydrochar (i.e. carbonised solids) for
339 storage or transportation. Although filtrate from hot filtration of SF contains more solid

340 particles, depending on process objectives hot filtration would be the optimum option,
341 particularly if the filtrate is to be reused or digested, e.g. by anaerobic means, as this contains
342 more dissolved organic components.

343 344 **ANOVA, Modelling and Optimisation of Filterability**

345 *Model Fitting and ANOVA*

346 RSM models developed using Design-Expert 9.0.1 software, and ignoring insignificant
347 terms, provide the constituent equations given in Eqs. (8–21) in terms of coded variables,
348 which are presented in Table 2 for filtrate volume, specific cake resistance of filter cake,
349 resistance on filter medium, cake volume concentration and solids leached into filtrate.

350 From the results of the analysis of variance (ANOVA) (see Tables S1–S3) for filtrate
351 volume, regression analysis of the experimental design showed that the linear model terms (T
352 and t_R) were highly significant ($P < 0.05$) for cold filtration of PSS and hot filtration of SF;
353 whilst the linear model term (T), interactive model term ($T t_R$), and quadratic term (T^2) were
354 significant for cold filtration of SF (other terms did not show significant effect). Specific cake
355 resistance for cold filtration of PSS was strongly influenced by the linear model term (T)
356 (other terms did not show significant effect); whilst cold of SF was strongly influenced by the
357 linear model term (T), and interactive term ($T t_R$); although hot filtration of SF was highly
358 affected by linear model terms (T and t_R), and interactive term ($T t_R$).

359 The resistance provided by the medium for cold filtration of PSS was significantly
360 affected by the linear model terms (T and t_R), whilst that of hot-filtered SF has temperature
361 as the only significant term as presented in Eq. (15). A model equation is not presented in the
362 case of cold filtration of SF as there are no significant model terms. For cake concentration
363 from cold filtration of PSS, there were no significant model terms and therefore no model
364 equation is presented here. As explained in Section 3.5, cake concentration of PSS was not
365 possible at lower treatment temperatures, and thus the model was developed taking into

366 consideration only data for HTC at 160°C and 240 min treatment time and at higher HTC
367 temperatures. Cake concentrations of cold- and hot-filtered SF were affected by linear model
368 terms (T and t_R). A linear model (Eq. (19)) was developed for total solids leached into filtrate
369 during cold filtration of PSS, which was affected by the linear model terms (T and t_R).
370 Similarly, solids in filtrate for cold and hot filtration of SF were only influenced by the linear
371 model term (t_R). The temperature term did not show a significant effect.

372 The significant *P*-values ($P < 0.05$) showed that the models were suitable and reliable,
373 with the exception of medium resistance and cake concentration for cold filtration of PSS, as
374 well as solids in the filtrate for cold and hot filtration of SF that have insignificant model
375 terms ($P > 0.05$). The models developed in this study are useful in specifying the effect of
376 each significant term and their interaction on the target variables, and thus the results provide
377 information useful for filtration of HTC-slurry from biomass.

378 379 *Optimisation and Validation*

380 The optimum HTC operating conditions for greater filterability from the RSM models
381 and results of validation experiment for specific cake resistance to filtration, resistance of
382 medium, cake concentration and solids in filtrate are illustrated in Figure 8. The models were
383 validated under the same optimal conditions to check their accuracy. For comparison, other
384 validated results at lower operating conditions are also presented in Figure 8. The minimum
385 specific cake resistance to filtration of $1.70 \times 10^{10} \text{ m kg}^{-1}$, $5.04 \times 10^{10} \text{ m kg}^{-1}$, and 3.02×10^{10}
386 $\text{m k}^{-1}\text{g}$ were predicted at the highest temperature (200°C) with the longest treatment time
387 (240 min) for cold filtration of PSS, cold filtration of SF, and hot filtration of SF,
388 respectively. Experimental results at the same optimal conditions were $2.05 \times 10^{10-1}$, $3.29 \times$
389 10^{10} , and $3.86 \times 10^{10} \text{ m kg}^{-1}$ for cold filtration of PSS, cold filtration of SF, and hot filtration
390 of SF, respectively. For cold and hot filtration of SF, experimental results obtained at 200°C
391 and shorter treatment time (60 min) were closer to the predicted values; with errors of 26%

392 and 7% than that for 240 min treatment time having errors of 35% and 28% respectively. The
393 experimental results were close to the predicted results and confirmed the validity of the
394 model. Based on the model, the lowest medium resistance of 1.20×10^{11} , 1.18×10^{10} , and
395 $0.54 \times 10^{10} \text{ m}^{-1}$ were at 200°C and 240 min treatment time, with the accuracy of the model
396 strongly confirmed for cold and hot filtration of SF (error between 16–27%).

397 The highest cake concentrations of 0.29, 0.44, and 0.54 (w/w) were predicted at 200°C
398 and 240 min for cold-filtered PSS, cold-filtered SF, and hot-filtered SF respectively; which
399 were confirmed by the validation experiments with differences of between 2–7%. Similarly,
400 solids in filtrate of 1.41%, 1.53%, and 2.00% were predicted at 200°C and 240 min. The
401 difference between the experimental results and predicted results were less than 1%, which
402 makes the predicted results validated by the actual values.

403 When designing industrial processes with filtration stages, i.e. scaling up, it is common
404 to apply a constituent equation relating the specific resistance to filtration with some
405 operating condition, such as operating pressure [32]. In the case of filtration of material from
406 the hydrothermal carbonisation process the significant operating condition is temperature,
407 and Table 2 provides the constitutive equations that can be used to relate the specific
408 resistance to this parameter. Table 2 provides similar constitutive equations for the other
409 parameters required for industrial design: filter medium resistance and cake concentration.
410 Using these three constituent equations it is possible to predict the performance of any type of
411 industrial operating mode for filtration: constant pressure, constant rate, or variable pressure
412 and rate; provided that the other system dependent parameters are also supplied: filter area
413 and operating pressure (as well as the viscosity of water at the appropriate temperature). The
414 equation used for the prediction depends on the filtration operating mode, but in every case it
415 is possible to model the filtrate volume with time using these input parameters, together with
416 the temperature dependent values obtained by RSM as shown in Table 2. Thus it is possible

417 to model the influence of residence time and treatment temperature on the overall filtration
418 performance of the system; e.g. it is possible to determine the filter area required to achieve a
419 given filtrate volume in a required time. Such information would be required if designing a
420 process where the filtration stage needs to be completed before the addition of the next batch
421 of carbonised slurry is filtered in a semi-batch system.

422 423 **CONCLUSIONS**

424 Results from the predicted RSM models and experimental data showed that the higher
425 the reaction temperature and the longer the treatment time, the greater was the carbonised
426 slurry's filterability. Specific cake resistance to filtration decreased as reaction temperature
427 and time increased. Dewatering the HTC-slurry whilst hot resulted in higher cake
428 concentrations, although filterability was not concomitantly improved. Filterability of HTC-
429 slurry was shown to be highly influenced by reaction temperature and treatment time.
430 Predictions were generally close to the validation results, which indicates that the models
431 based on the RSM data are applicable for prediction and optimisation of HTC-slurry
432 dewaterability. For example, in a process under design investigation there could be a desire to
433 operate at a lower temperature to provide solids suitable for treatment to form bioethanol,
434 whereas higher temperatures will provide greater quantities of carbonised solids more
435 suitable for gasification and syngas production during further processing. This study provides
436 quantitative information on the expected filterability of solids from the HTC process at
437 temperatures between 140 and 200°C. It can also be concluded that the synthetic faecal
438 sludge does provide similar filtration resistance to the primary sewage sludge. Hence,
439 processes can be justifiably developed using the simulant for preliminary testing.

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448 sludge.

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450 **Supplementary Data**

451 Details of results of the analysis of variance (ANOVA) for filtrate volume, specific cake
452 resistance, resistance of filter medium, cake concentration and solids in filtrate.

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549 **Table 1**
550 Filtration properties of carbonised primary sewage sludge and synthetic faeces slurries.
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552 **Table 2**
553 RSM model equations in terms of coded variables.
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Table 1 – Filtration properties of carbonised primary sewage sludge and synthetic faeces slurries.

HTC Parameters		Cold-filtered PSS			Cold- filtered SF	Hot-filtered SF
Temp. (°C)	Time (min)	TOC ^a (g/L)	TOC ^b (g/L)	Solids in filtrate (% wt.)	Solids in filtrate (% wt.)	Solids in filtrate (% wt.)
140	240	7.61 ± 0.22	4.87 ± 0.06	1.47 ± 0.27	1.94 ± 0.004	1.64 ± 0.01
160	60	9.22 ± 0.07	5.37 ± 0.30	1.90 ± 0.08	1.92 ± 0.26	2.83 ± 0.26
160	120	9.10 ± 0.09	6.63 ± 0.41	1.84 ± 0.02	1.77 ± 0.01	2.63 ± 0.14
160	240	8.36 ± 0.35	9.49 ± 0.73	1.62 ± 0.02	1.61 ± 0.004	2.27 ± 0.09
180	30	9.03 ± 0.11	4.99 ± 0.11	1.90 ± 0.03	1.88 ± 0.01	2.22 ± 0.01
180	60	9.37 ± 0.06	6.47 ± 0.15	1.85 ± 0.15	1.86 ± 0.01	2.15 ± 0.002
180	120	9.24 ± 0.04	7.30 ± 0.13	1.82 ± 0.11	1.76 ± 0.02	2.04 ± 0.68
180	240	9.09 ± 0.08	9.99 ± 0.07	1.65 ± 0.18	1.66 ± 0.01	1.98 ± 0.004
200	15	9.50 ± 0.05	7.67 ± 0.10	1.88 ± 0.14	2.36 ± 0.03	2.64 ± 0.03
200	30	9.06 ± 0.16	7.73 ± 0.10	1.78 ± 0.10	1.74 ± 0.18	2.16 ± 0.01
200	60	8.43 ± 0.30	8.76 ± 0.11	1.72 ± 0.04	1.68 ± 0.002	2.07 ± 0.004
200	120	8.13 ± 0.28	10.10 ± 0.07	1.47 ± 0.05	1.66 ± 0.01	2.00 ± 0.21
200	240	8.57 ± 0.02	13.68 ± 1.12	1.42 ± 0.18	1.52 ± 0.03	1.96 ± 0.001

^a TOC analysed directly after filtration test using 10 µm slotted filter medium. ^b TOC analysed after further filtration with a Whatman filter paper.

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Table 2 – RSM model equations in terms of coded variables

Parameter	Equation	
<i>Filtrate (m³)</i>		
Cold-filtered PSS	$V = 6.53 \times 10^{-5} + 7.42 \times 10^{-5}T + 2.54 \times 10^{-5}t_R$	(8)
Cold-filtered SF	$V = 3.28 \times 10^{-4} + 1.64 \times 10^{-4}T - 9.05 \times 10^{-5}Tt_R - 1.93 \times 10^{-4}T^2$	(9)
Hot-filtered SF	$V = 2.19 \times 10^{-4} + 1.29 \times 10^{-4}T + 5.03 \times 10^{-5}t_R$	(10)
<i>Specific cake resistance (m kg⁻¹)</i>		
Cold-filtered PSS	$\alpha = 1.21 \times 10^{12} - 1.37 \times 10^{12}T$	(11)
Cold-filtered SF	$\alpha = 1.20 \times 10^{11} - 1.59 \times 10^{11}T + 1.73 \times 10^{11}t_R - 3.09 \times 10^{11}Tt_R$	(12)
Hot-filtered SF	$\alpha = 9.78 \times 10^{11} - 1.14 \times 10^{12}T - 8.06 \times 10^{11}t_R + 9.01 \times 10^{11}Tt_R$	(13)
<i>Resistance of medium (m⁻¹)</i>		
Cold-filtered PSS	$R_m = 9.49 \times 10^{11} - 9.15 \times 10^{11}T - 9.79 \times 10^{11}t_R$	(14)
Hot-filtered SF	$R_m = 4.37 \times 10^{11} - 2.01 \times 10^9T$	(15)
<i>Cake concentration (w/w)</i>		
Cold-filtered PSS	$C_w = 0.19 + 0.06T$	(16)
Cold-filtered SF	$C_w = 0.32 + 0.09T + 0.03t_R$	(17)
Ho-filtered SF	$C_w = 0.40 + 0.11T + 0.04t_R$	(18)
<i>Solids in filtrate (% wt.)</i>		
Cold-filtered PSS	$s_f = 1.82 - 0.21 T + 0.20t_R$	(19)
Cold-filtered SF	$s_f = 1.83 - 0.18t_R$	(20)
Hot-filtered SF	$s_f = 2.44 - 0.27t_R$	(21)
T is reaction temperature (°C); t _R is reaction time (min); C _w is cake weight concentration (w/w); and s _f is solids in filtrate (% wt.).		

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610 **Figure 1** Constant pressure filtration equipment for: (a) cold filtration of PSS; (b) cold and
611 hot filtration of SF.

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613 160°C: (a) cold filtration of PSS; (b) cold filtration of SF; (c) hot filtration of SF.

614 **Figure 3** Effect of reaction temperature and time on filtrate volume: (a) cold filtration of
615 PSS; (b) cold filtration of SF; (c) hot filtration of SF, for HTC at 180°C (a, b, c); (d) cold
616 filtration of PSS; (e) cold filtration of SF; (e) hot filtration of SF, for HTC at 200°C (d, e, f).

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619 at 180°C (a, b, c); (d) cold filtration of PSS; (e) cold filtration of SF; (f) hot filtration of SF,
620 for HTC at 200°C (d, e, f).

621 **Figure 5** Contour plot showing the effect of reaction temperature and time on specific cake
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623 SF.

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625 filter medium: (a) cold filtration of PSS; (b) cold filtration of SF; (c) hot filtration of SF.

626 **Figure 7** Contour plot showing the effect of reaction temperature and time on cake
627 concentration: (a) cold filtration of PSS; (b) cold filtration of SF; (c) hot filtration of SF.

628 **Figure 8** Plots of observed values from experiment and predicted optimised values from the
629 models: (a) specific cake resistance (b) resistance of medium; (c) cake concentration; and (d)
630 solids in filtrate.

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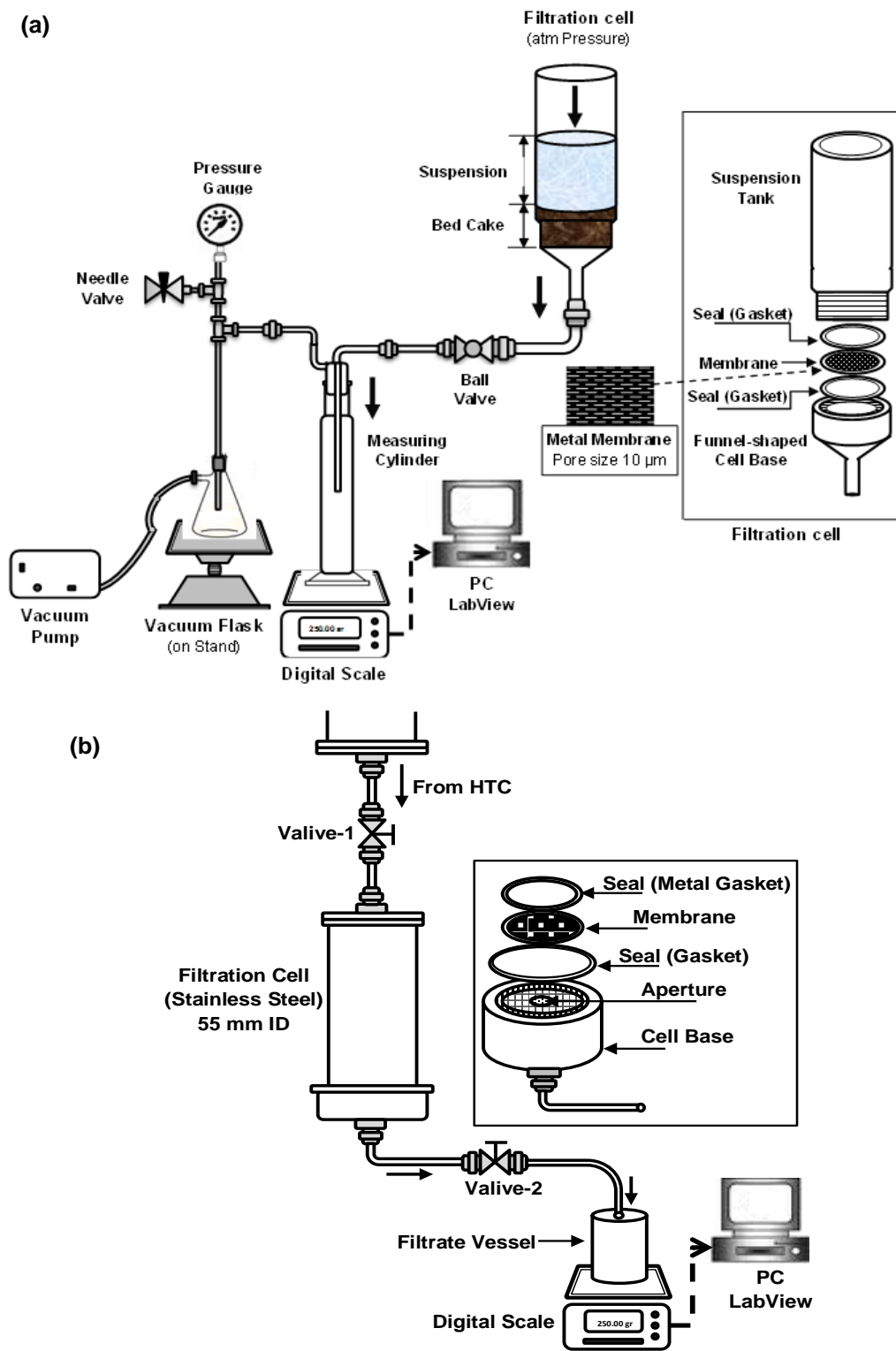
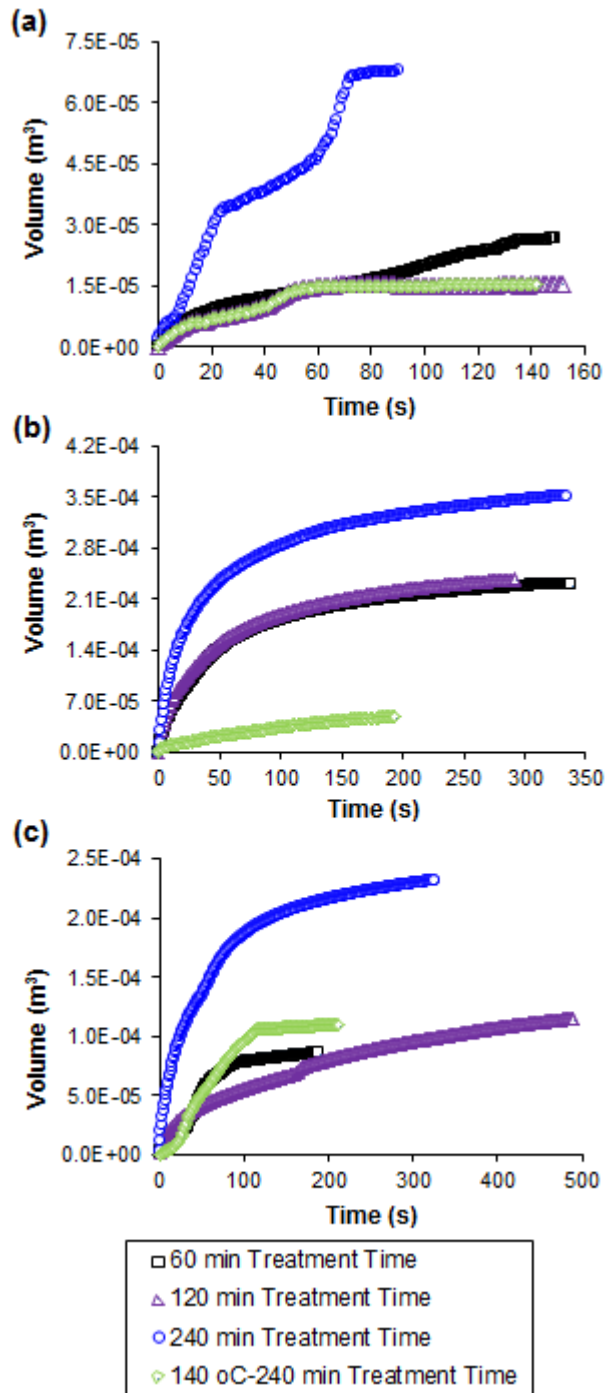


Figure 1 – Constant pressure filtration equipment for: (a) cold filtration of PSS; (b) cold and hot filtration of SF.

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Figure 2 – Effect of reaction temperature and time on filtrate volume for HTC at 140°C and 160°C: (a) cold filtration of PSS; (b) cold filtration of SF; (c) hot filtration of SF.

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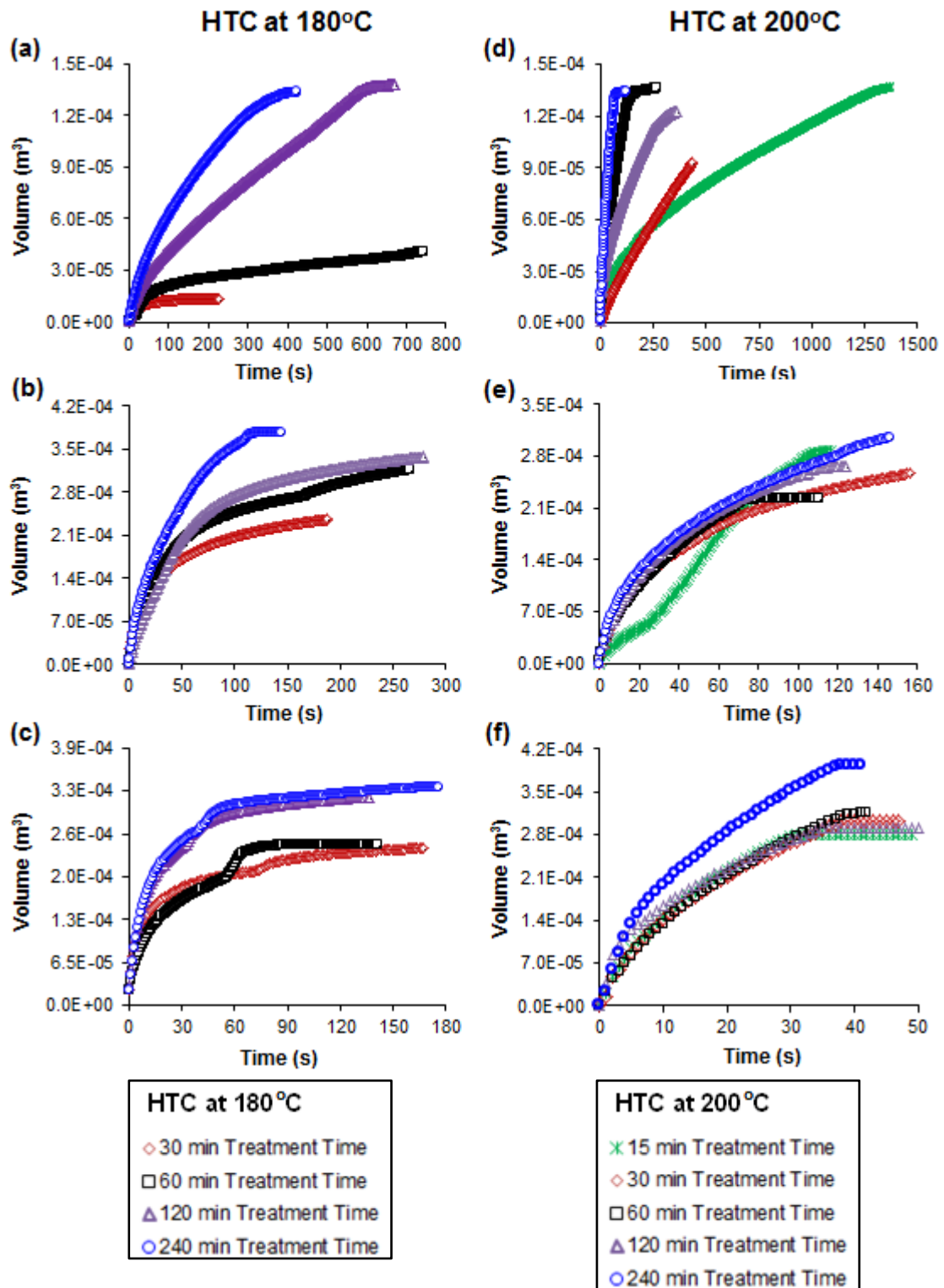
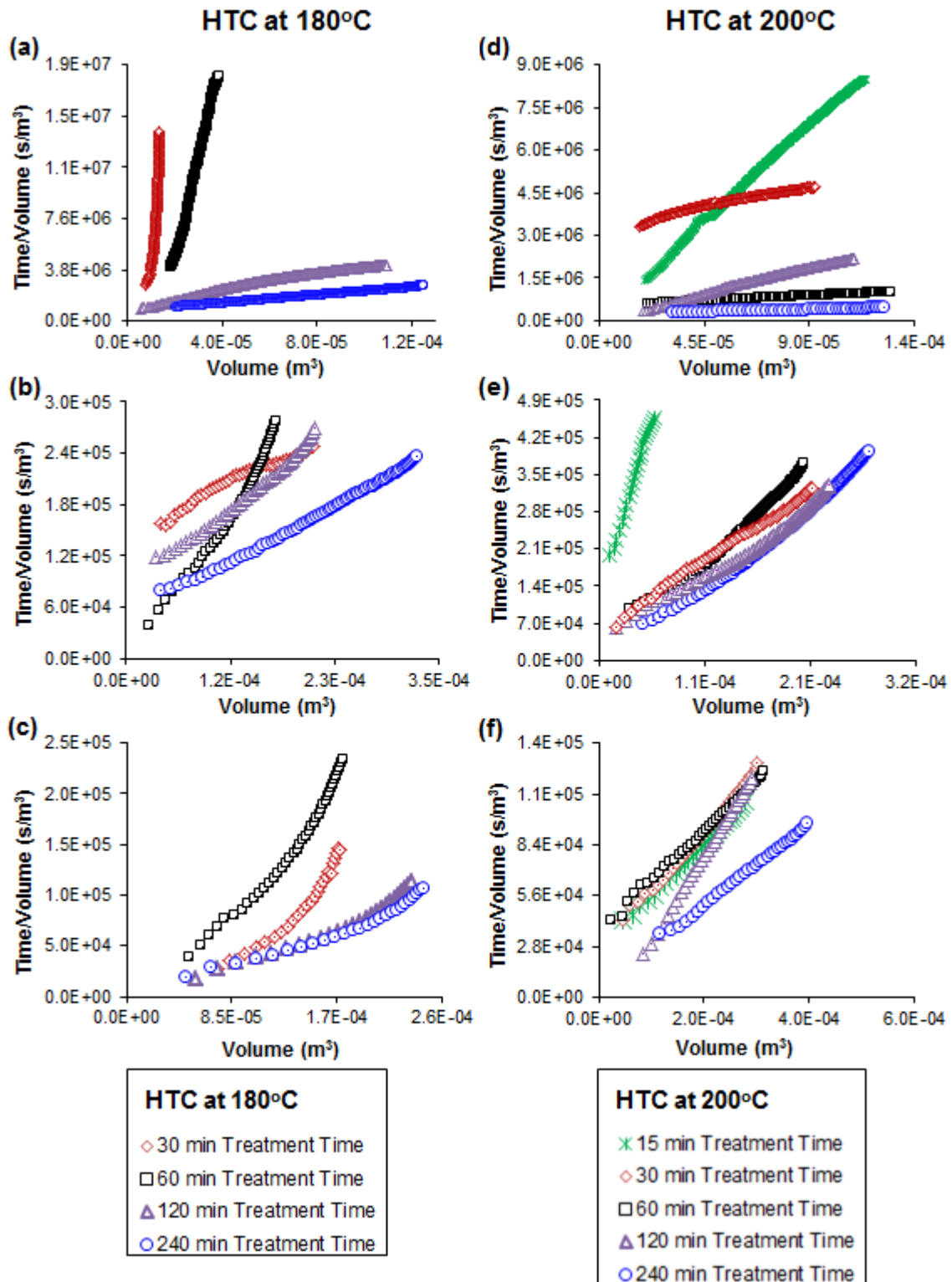


Figure 3 – Effect of reaction temperature and time on filtrate volume: (a), (d) cold filtration of PSS; (b), (e) cold filtration of SF; (c), (f) hot filtration of SF.

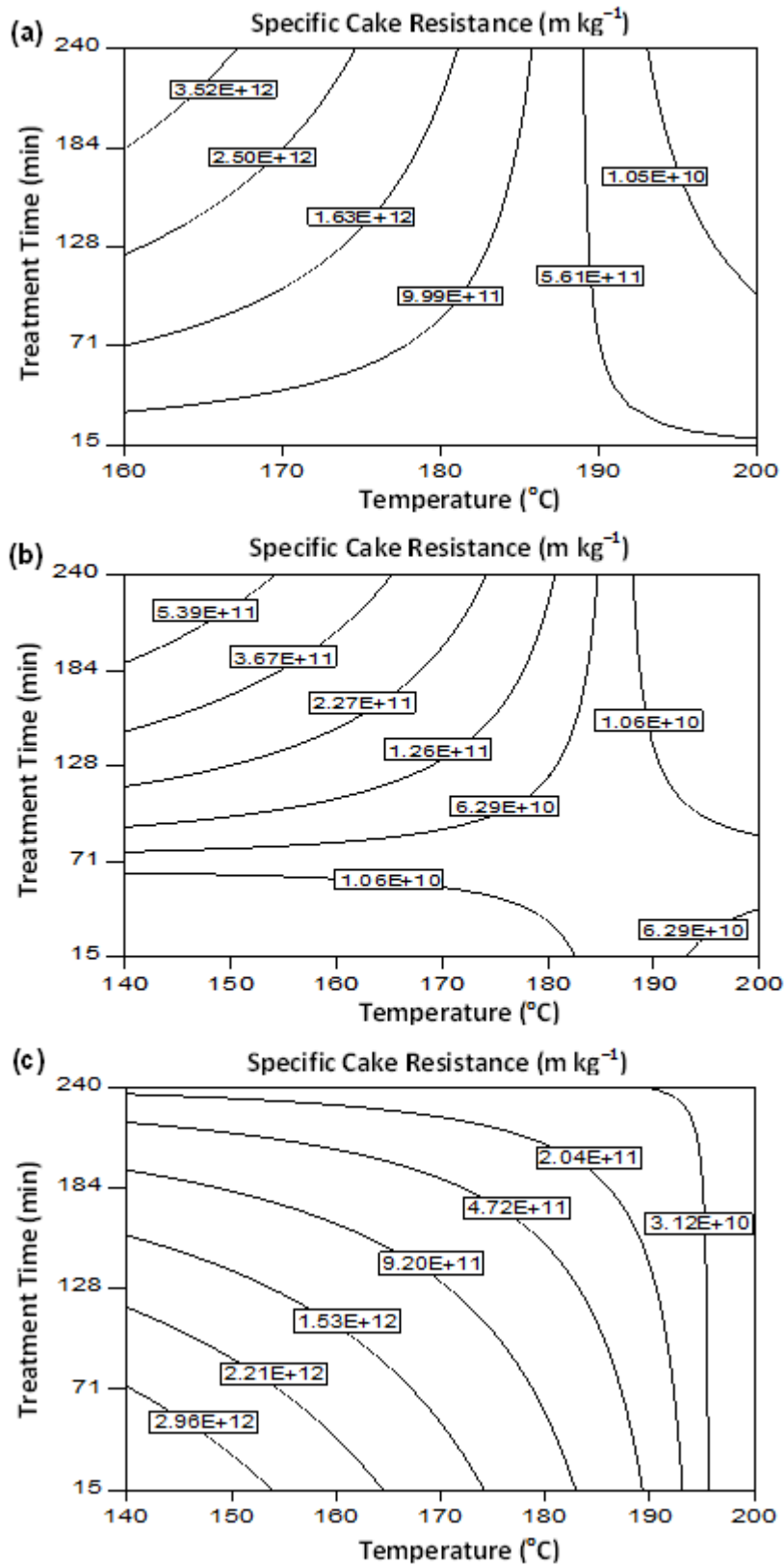


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708 **Figure 4** – Effect of reaction temperature and time on filtrate volume analysis by parabolic

709 rate law: (a), (d) cold filtration of PSS; (b), (e) cold filtration of SF; (c), (f) hot filtration of

710 SF.

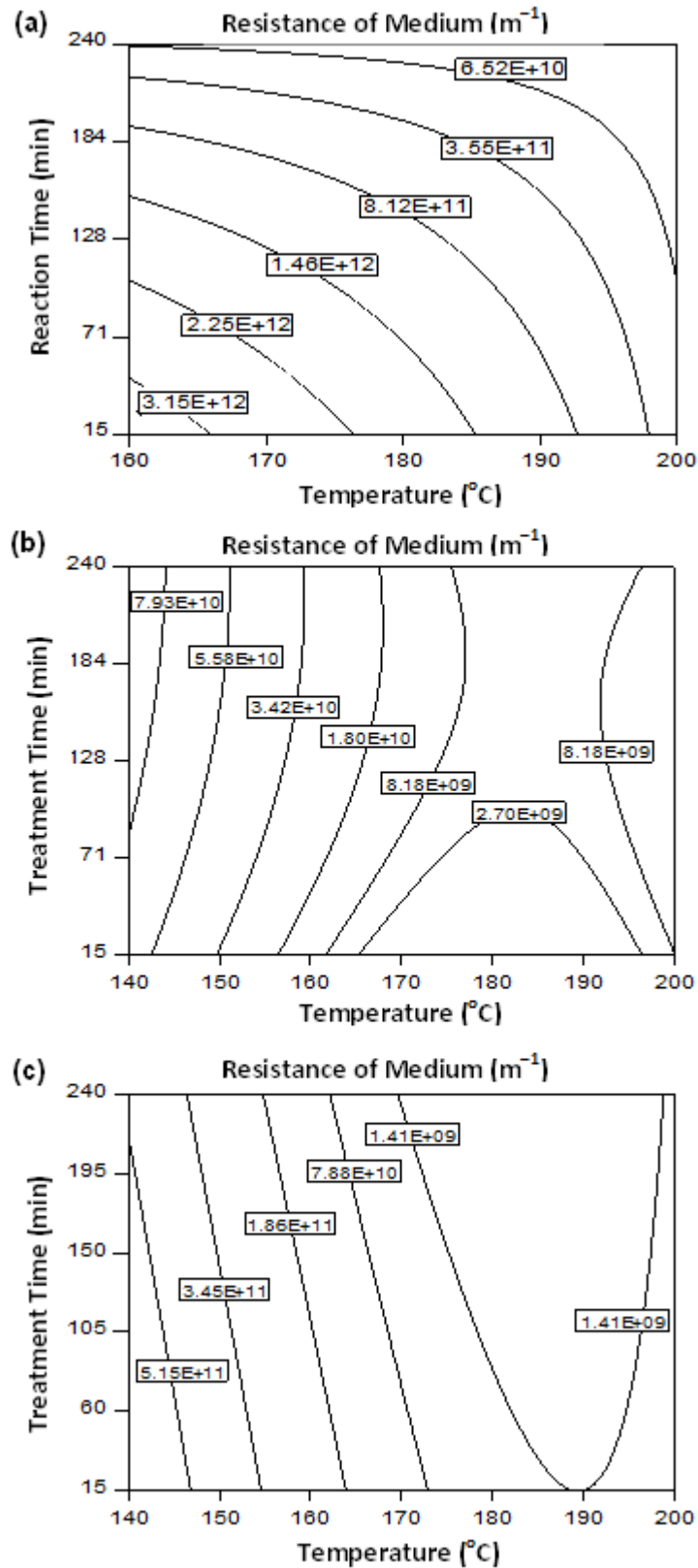


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712 **Figure 5** – Contour plot showing the effect of reaction temperature and time on specific cake

713 resistance to filtration: (a) cold filtration of PSS; (b) cold filtration of SF; (c) hot filtration of

714 SF.

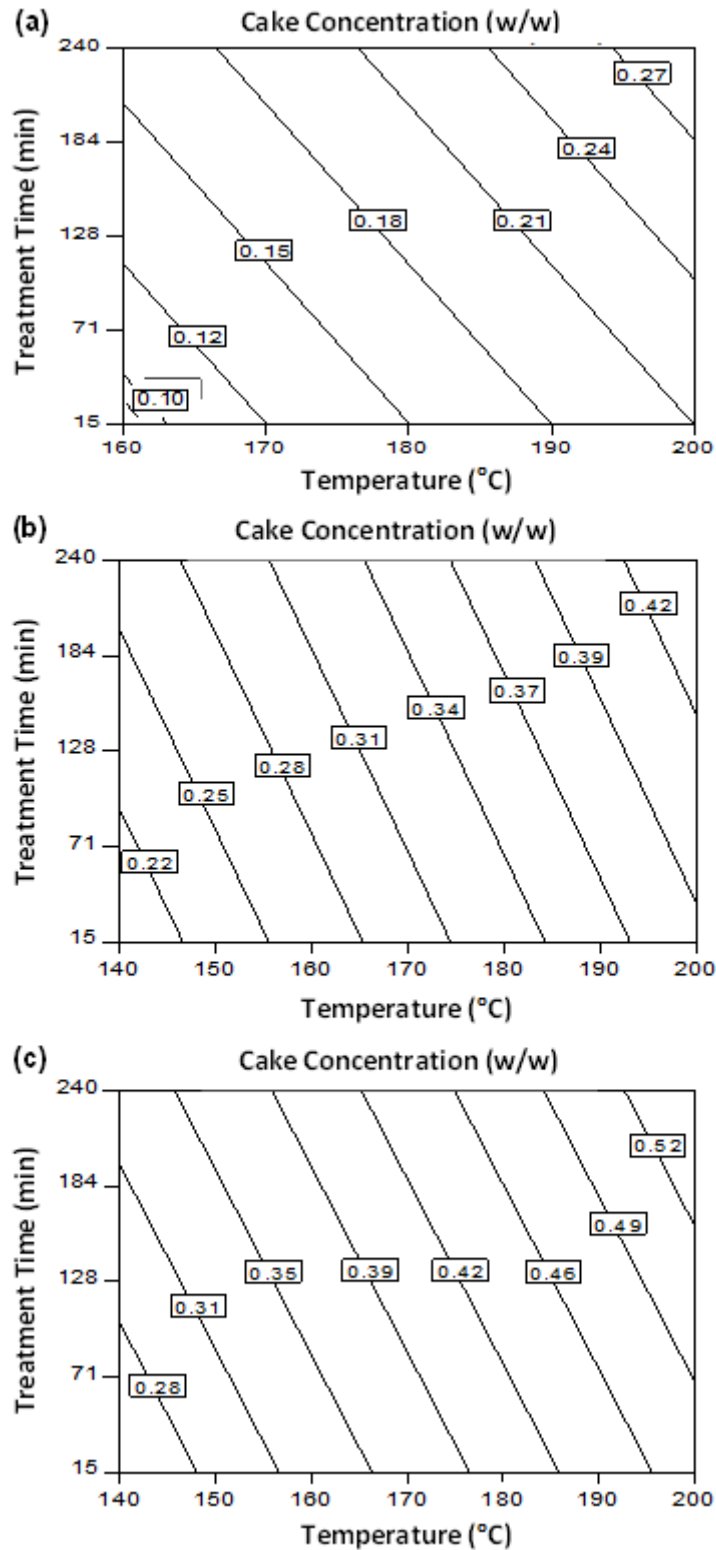


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716 **Figure 6** – Contour plot showing the effect of reaction temperature and time on resistance of

717 filter medium: (a) cold filtration of PSS; (b) cold filtration of SF; (c) hot filtration of SF.

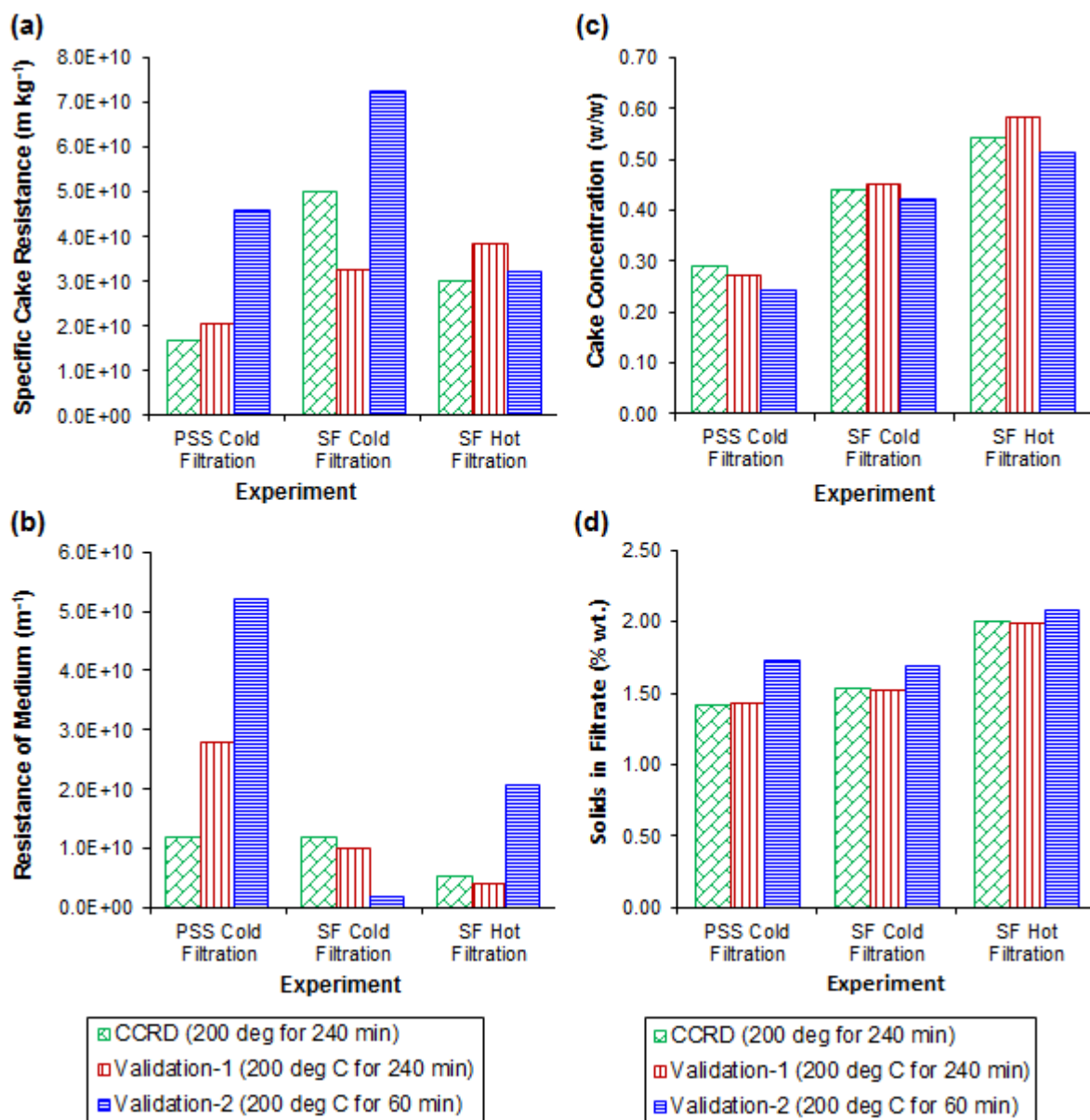
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720 **Figure 7** – Contour plot showing the effect of reaction temperature and time on cake
 721 concentration: (a) cold filtration of PSS; (b) cold filtration of SF; (c) hot filtration of SF.

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726 **Figure 8** – Plots of observed values from experiment and predicted optimised values from the
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 728 solids in filtrate.

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