

Image-based Monitoring for Early Detection of Fouling in Crystallisation Processes

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Abstract

Fouling or encrustation is a significant problem in continuous crystallisation processes where crystal deposits at surfaces impede heat transfer, increase flow resistance and reduce product quality. This paper proposes an automatic algorithm to detect early stages of fouling using images of vessel surfaces

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from commodity cameras. Statistical analysis of the pixel intensity variation offers the ability to distinguish appearance of crystals in the bulk solution and on the crystalliser walls. This information is used to develop a fouling metric indicator and determine separately induction times for appearance of first crystals at the surfaces and in the bulk. A method to detect process state changes using Bayesian online change point detection is also proposed, where the first change point is used to determine induction time either at the surface or in the bulk, based on real-time online measurements without using any predetermined threshold which usually varies between experiments and depends on data acquisition equipment. This approach can be used for in situ monitoring of early signs of encrustation to allow early warning for corrective actions to be taken when operating continuous crystallisation processes.

Keywords:

Continuous Crystallisation, Fouling, Encrustation, Nucleation, Induction Time, Imaging, Change Point Detection

1. Introduction

Crystallisation is an important unit operation in chemical, pharmaceutical and food industries for isolation and purification of intermediates and final products. Fouling or encrustation in the context of crystallisation is the formation of crystal deposits on equipment surfaces, pipe walls and process analytical probes and occurs as a result of heterogeneous nucleation and/or attachment of crystals and their subsequent growth at solid surfaces [1, 2]. Fouled surfaces impede heat transfer [3], increase flow resistance and decrease product quality [4]. The impact of fouling and encrustation is even

10 greater on continuous crystallisation processes where system blockages re-
11 sult in shutdowns leading to losses of time and revenue. Process conditions
12 that influence fouling in crystallisation processes include solvent composi-
13 tion, supersaturation, pH, temperature, flow velocity and regime, additives
14 or impurities, while interface conditions that affect fouling include surface
15 energy, roughness and topography, number of nucleation sites and aging of
16 the fouling layer [5]. Mechanisms of initiating encrustation have been stud-
17 ied previously, where a crystalline phase was nucleated on solid surfaces at
18 controlled local supersaturation and without influence from particle attach-
19 ment [6].

20 Common methods to mitigate fouling include mechanically altering the
21 surface using methods such as ultrasound or scraping or chemically altering
22 the surface using coatings to reduce the number of nucleation sites and/or
23 remove attached crystals [7]. Increasing the induction time for surface fouling
24 can be achieved by changing the energy and topography of the surface or by
25 changing the flow conditions [8]. The rough surface can stabilising crystal
26 nuclei and promote growth on the surface, while fouling is typically delayed
27 at smoother surfaces.

28 A review of detection methods of fouling in the food industry can be
29 found in previous literature [4]. Common, non-invasive methods to detect
30 fouling are to monitor the hydrodynamic and heat transfer parameters. Hy-
31 drodynamic methods monitor the inlet and outlet pressures and infer the
32 degree of deposits on the walls of a tube. Heat transfer methods monitor
33 heat transfer losses to infer the degree of fouling. Both of these methods
34 have low sensitivity and generally not suitable for early stage monitoring of

35 fouling in continuous crystallisers since significant fouling must occur before
36 temperature or pressure variations are detectable. Other methods include
37 the measurement of the electrical resistance or conductivity, ultrasound and
38 vibration, however all these alter the fouling (i.e. fouled deposits may be
39 broken off and then drift downstream). In the crystallisation domain it is
40 common to use reflectance, turbidity and other spectroscopic methods to
41 monitor processes in situ but none of these are specifically suited to provide
42 direct information about the state of vessel walls and presence or absence
43 of fouling therein. For example, when turbidity probes are used to estimate
44 nucleation induction time [9, 10], the method is unable to distinguish ap-
45 pearance of crystals in the liquid phase from that on vessel walls if and when
46 fouling occurs.

47 Imaging is another technique commonly used to track various proper-
48 ties of multiphase systems containing suspended solid particles and/or bub-
49 bles [11], identify nucleation induction time [12, 13] or crystal growth [14] in
50 industrial settings. A review of recent advances in monitoring and control
51 of crystallisation systems using imaging can be found in [15]. Use of in-situ
52 endoscopy-stroboscopy [16] equipment and multivariate image analysis for
53 process monitoring has been demonstrated. Estimating nucleation induction
54 time in the bulk through Bulk Video Imaging [17] using multivariate image
55 analysis and by converting pixel intensities time series of acoustic signals
56 allows to eliminate stirrer effects through band-stop filtering in batch crys-
57 tallisers. Methods to automatically estimate induction time using Shewhart
58 Charts was shown in [17, 18]. These techniques apply to bulk monitoring only
59 and to our knowledge there is no previously published method for detection

60 of fouling through image analysis.

61 Fouling is an important problem in both batch and continuous crystalli-
62 sation. It is especially significant for continuous crystallisation processes
63 where early detection of fouling can be used to guide relevant corrective ac-
64 tions in order to keep process running. In this work a batch system has been
65 designed which locally mimics the behaviour of a continuous plug-flow type
66 crystallisation platform, in terms of keeping the local temperature and so-
67 lution concentration constant, at least until the appearance of first crystals
68 at walls or in the bulk. The approach proposed here is applicable subject
69 to visual access to the crystalliser walls and uses an off-the-shelf commodity
70 camera, pointing towards an area of interest at solid-liquid interface while
71 looking through a transparent crystalliser wall¹. Statistical analysis of the
72 acquired images allows distinguishing crystal appearance in the bulk solution
73 from fouling on the solid surfaces. Note that the induction times determined
74 here refer to the location where crystals are observed and appearance of crys-
75 tals at that location does not necessarily mean that nucleation occurred there.
76 The aim of the proposed technique is to separately determine induction times
77 for appearance of crystals at solid surfaces and in the bulk automatically and
78 in real-time, by providing a sensitive early warning system for detection of
79 fouling in crystallisation processes.

¹Industrial settings with opaque walls will require appropriate adjustments.

80 **2. Methodology**

81 *2.1. Experimental Setup*

82 The experimental setup used to monitor fouling at glass crystalliser walls
83 under isothermal conditions was a batch system known as a moving fluid
84 oscillatory baffled crystalliser (MFOBC). However, we note that the image-
85 based monitoring and image analysis approach proposed here can be used
86 to analyse data collected from any other experimental setup where real-time
87 images of vessel walls are available, under batch or continuous, isothermal or
88 non-isothermal conditions.

89 The MFOBC has orifice baffles spaced equally throughout the glass pipe
90 which are overlaid with the direction of fluid oscillation [19]. Local mixing
91 is facilitated through Eddie formation due to the interaction of the baffles
92 with the oscillatory flow. The oscillation conditions are defined through fre-
93 quency and amplitude [20]. This system can achieve close to plug flow when
94 operating under continuous conditions, as a continuous oscillatory baffled
95 crystalliser (COBC), at relatively low net flow rates while good heat and
96 mass transfer is radial direction as well as solid suspension is facilitated by
97 oscillatory motions. In a typical COBC setup there are several glass sections
98 operated under different temperatures and fouling is likely to occur in a sec-
99 tion with highest supersaturation. In order to monitor the behaviour in such
100 a section independent from the rest of the COBC setup, a physical batch
101 model locally representing the COBC conditions was constructed by remov-
102 ing the section of interest from the continuous arrangement and mounting
103 it vertically between two other sections providing suitable boundary condi-
104 tions to prevent nucleation outside of the section of interest by keeping the

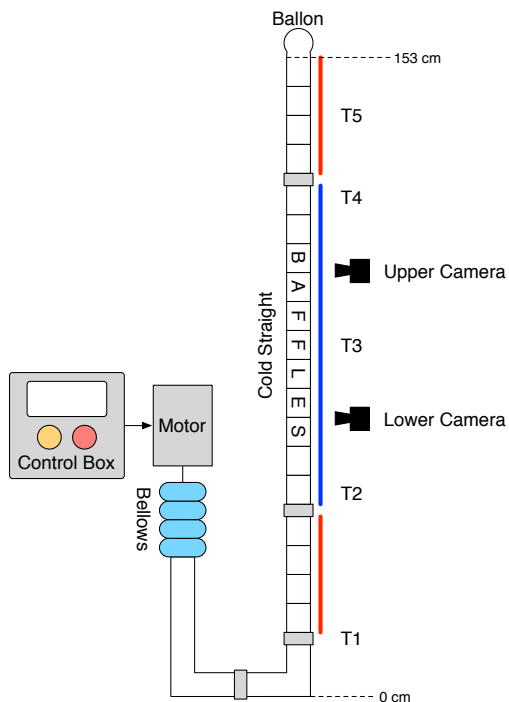


Figure 1: Experimental setup showing positions of temperature measurement, cameras, glass sections and collars which connect the glass components together.

105 temperature of the fluid outside of the section above saturation temperature.
 106 The resulting batch setup can reproduce local temperature, concentration
 107 and flow conditions identical to the relevant section of the COBC under cor-
 108 responding conditions, in the absence of any solid phase until onset of crystal
 109 formation is observed.

110 The MFOBC used here (shown in Figure 1) consists of tubular jacketed
 111 glass sections with orifice baffles spaced equally throughout. Each full glass
 112 sections is made up of 22 individual cells of volume 5 ml and internal diame-
 113 ter 15 mm (DN15). The batch setup contains one half glass straight attached
 114 to bellows and kept at temperature T1, followed by one full straight where

115 desired target temperature is kept at the position of monitoring cameras to
116 achieve required supersaturation, and then another half straight at the top
117 kept at temperature T5, completing the setup. The principle behind this
118 configuration is that while all three regions are interconnected, the tempera-
119 tures are calibrated so that the cameras are always monitoring regions of fixed
120 temperature. The two half straights above and below are kept hot to prevent
121 seeding from above and below, respectively. Temperatures T1 and T2 are
122 monitored during experiments via built-in thermocouples. Temperatures T3,
123 T4 and T5 are recorded for calibration purposes prior to the experiment as
124 multiple thermocouples in the system may affect the fouling process.

125 Two sets of oscillations conditions were used for fouling experiments, high
126 oscillations with frequency and amplitude of 2 Hz and 45 mm, respectively,
127 and low oscillations with frequency and amplitude of 1 Hz and 45 mm, re-
128 spectively. Table 1 gives temperature information pertaining to the two
129 oscillation conditions used for the experiments.

130 L-Glutamic Acid (LGA) was crystallised from water in this setup. The
131 supersaturations of the solutions were calculated from the solubility at 20°C,
132 which has a value of 7.07 g/L of water [21, 22]. Supersaturations 3, 4 and
133 6 were used. Hot solution at 20°C was prepared out with the MFOBC and
134 pumped while hot in to the MFOBC using a peristaltic pump via a port at
135 the base of the equipment. This was filled so that the upper half section was
136 75% full. At this point the oscillation was set and the experiment was run and
137 images were taken. Two Microsoft LifeCam VX-3000 cameras are used and
138 LED torch is used for illumination inside an enclosed environment to ensure
139 constant lighting and to minimise reflections. The jacket temperatures of the

Position on MFOBC	High Oscillation	Low Oscillation
T1 (16.5 cm)	82°C	84.0°C
T2 (54.5 cm)	26.8°C	29.5°C
Lower Camera (66.5 cm)	22.0°C	23.3°C
T3 (85.5 cm)	19.5°C	19.8°C
Upper Camera (88.0 cm)	19.5°C	19.7°C
T4 (110 cm)	23.7°C	24.6°C
T5 (134 cm)	38.4°C	41.5°C

Table 1: Temperature Profiles for High Oscillation Conditions (2 Hz, 45 mm) and Low Oscillation Conditions (1 Hz, 45 mm).

140 glass straights were preset before being filled with hot solutions in order to
141 quickly achieve the desired bulk solution temperatures. A balloon was placed
142 over the top section of the MFOBC to eliminate effects from evaporation.

143 *2.2. Image Preprocessing*

144 The web camera output is a series of JPEG images with resolution of
145 320x240 pixels. Each pixel consists of a tuple of the Red, Green and Blue
146 8-bit colour components. The 8-bit component values allow colour intensities
147 to be captured in the range of 0 and 255, where 0 is no presence and 255
148 is maximum presence of the corresponding component. A pixel with value
149 $(R,G,B) = (255, 255, 255)$ represents a white pixel, while a pixel with value
150 $(R, G, B) = (0, 0, 0)$ represents a black pixel.

151 The sequence of RGB JPEG images is converted to grey scale images us-
152 ing the formula $0.2989R+0.5870G+0.1140B$ which is the well accepted NTSC

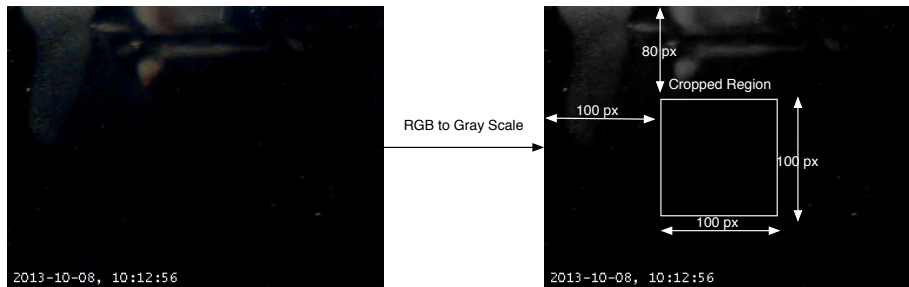


Figure 2: Image pre-processing.

153 standard for luminance. The images are cropped to 100x100 pixels, with off-
 154 set of 80 pixels from the top and 100 pixels from the left (shown in Figure 2),
 155 in order to eliminate reflections from the cylindrical reactor walls. The exact
 156 location of cropping is not important as long as reflections due to the pipe
 157 curvature and the time stamp annotations are eliminated. As a result, the
 158 initial cropped frame has only black pixels (intensity 0) when the crystalliser
 159 has no growth in the bulk or walls. The cropping is performed in all images
 160 acquired and the resulting image sequence consists of a 3 dimensional array
 161 with $N = 100$ rows by $M = 100$ columns and K frames. The instantaneous
 162 intensity of a pixel at location (i, j) at time t is denoted by $I_{ij}(t)$.

163 2.3. Analysis Methodology

164 Often experimentalists capture signal traces (profiles) and estimate in-
 165 duction time offline after the experiments are finished using a first order
 166 polynomial regression near the region of the crystal appearance. This kind of
 167 approach clearly does not allow early detection of crystal appearance and/or
 168 fouling events since an onset of change in relevant measurable quantities
 169 can only be determined after the extent of change becomes very significant.

170 Moreover, choice of regression window can be arbitrary and often cannot be
171 uniquely determined. An online real-time method to identify induction time
172 is usually based on selection of a predetermined, fixed amplitude threshold
173 and when the signal traces exceed that threshold, it is concluded that the
174 induction time has been observed. Although this may be adequate for well
175 characterised setups with known compounds and experimental conditions, it
176 is impractical when applied to new systems or under different conditions or
177 equipment. In order to address this challenge, a Bayesian Online Change
178 Point Detection technique is presented in Section 2.4 for the automatic esti-
179 mation of the induction time independent of setup, compound or conditions
180 and it is agnostic of the signal trace used (i.e. sensor type). Its applicability
181 for a wide range of signal traces is demonstrated throughout this work as it
182 is the method chosen here to identify induction time in either bulk solution,
183 at solid surfaces, or both combined.

184 Typical signal traces used to measure induction times are provided by
185 Process Analytical Technology (PAT) equipment based on spectroscopic,
186 scattering or video/imaging methods. However, such equipment is generally
187 only able to detect induction time for crystal appearance in the bulk solution
188 and in case signals from both bulk solution and vessel walls are recorded, it
189 provides information on combined bulk and wall changes together. Some
190 instruments such as the Mettler Toledo FBRM now provide a fouling index
191 indicator for the probe window however these may not be representative of
192 the encrustation levels experienced on the reactor walls, since probe coating
193 materials are often different from those on the reactor walls hence varying
194 the degree of encrustation. Some previous reports in the literature have em-

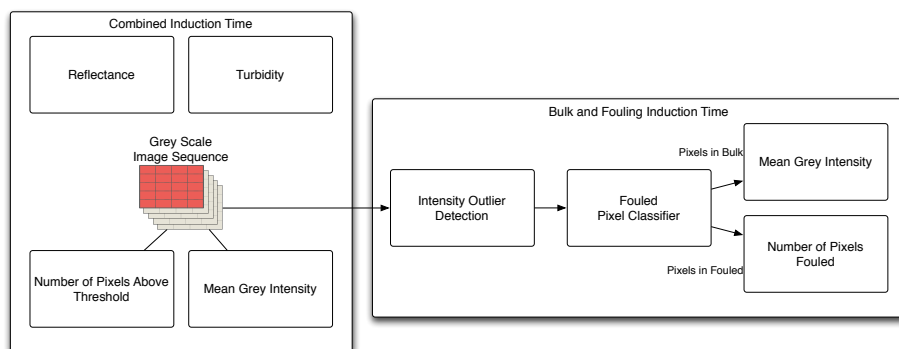


Figure 3: Signals that can be used to estimate combined induction time and the proposed method to separate crystal appearance in the bulk solution and at the solid surfaces.

195 employed imaging systems to estimate induction time [12, 13]. These techniques
 196 are able to report in real-time the induction time using Mean Grey Intensity
 197 as a signal trace and amplitude thresholding as a detection method. Their
 198 performance is similar to the that of PAT equipment such as ATR-IR [13].
 199 Another rudimentary signal trace is to count the number of pixels above an
 200 intensity threshold. All the signal traces that can be used to measure com-
 201 bined induction time are illustrated in Figure 3 and described in detail in
 202 Section 3.

203 Figure 3 shows the proposed approach to detect separately crystal ap-
 204 pearance in the bulk solution and on the vessel walls. The acquired images
 205 are initially passed through an Intensity Outlier Detector which highlights
 206 pixels with higher intensity. These pixels are subsequently processed by the
 207 Fouled Pixel Classifier that infers pixel state as fouled and non-fouled. The
 208 non-fouled pixels (i.e. in the bulk) are subsequently processed to generate
 209 the Mean Grey Intensity signal trace for estimation of the bulk induction
 210 time, while the fouled pixels are counted to provide a fouling index indicator

211 and estimate the fouling induction time. The bulk and fouling induction
212 time estimations are described in detail in Section 4.

213 *2.4. Automatic Change Point Detection*

214 A typical progression profile from turbidity, reflectance or image Mean
215 Grey Intensity start with low amplitude and as crystals appear, the amplitude
216 rises. Induction times can be determined in a post-processing fashion after
217 an experiment is completed or online, through thresholding the amplitude of
218 the signal trace. Although these techniques are effective, they are sensitive
219 to acquisition equipment and experimental conditions with parameter tuning
220 necessary. In order to overcome these limitation, a Bayesian Online Change
221 Point Detection [23] approach is used here, modelling the data as a Poisson
222 process and the rate of exponential prior on the change point interval $\lambda_{gap} =$
223 1000 (i.e. change point distribution) in a similar fashion to the Coal Mine
224 Disaster Data in [23].

225 The top plot in Figure 4 shows the MGI data of an experiment against
226 time. For every new observation (i.e. MGI value), while the parameter η of
227 the Poisson distribution remains the same, there is no occurrence of a change
228 point and the run length increases by 1. When a new observation appears
229 that comes from a Poisson with a different parameter η' , the run time drops
230 indicating a change point. The change point interval has rate λ_{gap} which
231 models probabilistically the occurrence of a change point.

232 At the bottom graph of Figure 4 the intensity plot shows the posterior
233 probability of the current run length $P(r_t|x_{1:t})$ at each time step t using a
234 logarithmic scale. Darker pixels indicate higher probability while white pixels
235 indicate zero probability. For a time step t , a column of pixels of the intensity

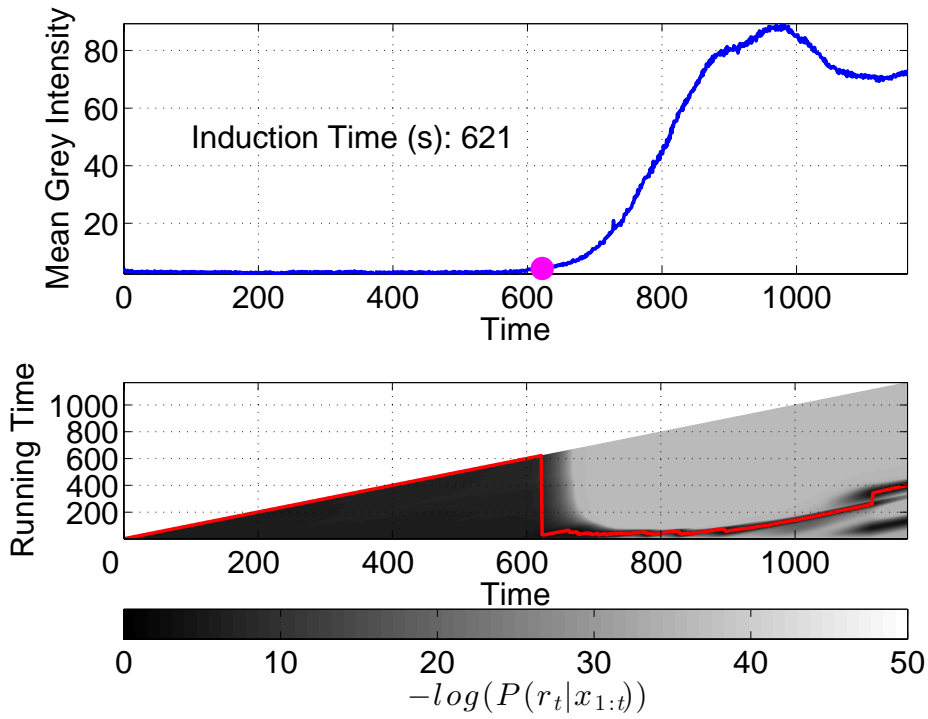


Figure 4: Top: Mean Grey Intensity profile against time. Bottom: The intensity plot shows the probability of the current run time $P(r_t|x_{1:t})$ at each time step (in logarithmic scale). The most probable run time r_t is annotated with the red line.

236 plot illustrate the probability mass function for every run time/length.

237 For example, for $t = 600$, the run time above 600 has zero probability
 238 and hence the $-\log(0) = Inf$ which is shown as white. For run time below
 239 600, the probability is between 0 and 1 which results in a shade of grey. The
 240 highest probability occurs at run time 600 which is the darkest pixel on that
 241 column and a change point has yet to occur. Similarly, for time $t = 1000$,
 242 the maximum occurs at run length 140 (a number of changes points have
 243 already occurred). The most probable run times are annotated with a red
 244 line on the bottom graph of Figure 4.

245 The algorithm finds the negative slopes on the most probable run times
246 (i.e. the points where the run time is reset) and these are the detected change
247 points. The first change point is attributed to the onset of crystal formation.
248 Subsequent change points are caused from breakage of encrusted regions and
249 re-encrustation, but those are not considered in this analysis.

250 **3. Combined Induction Time Estimation**

251 In this section, the Bayesian Online Change Point Detection algorithm
252 is applied in a number of signal traces to estimate combined induction time.
253 The algorithm is applied to turbidity, FBRM and image based signals as
254 shown in Figure 3.

255 As an example, consider a crystallisation experiment for LGA with con-
256 centration 45 g/L in water, oscillation amplitude of 30 mm and frequency
257 1.5 Hz. The measured profiles of the experimental signals are shown in Fig-
258 ure 5. The induction times based on the algorithm presented in Section 2.4
259 using the FBRM and turbidity signal traces are 23.50 and 27.25 minutes
260 respectively.

261 Previous applications of image processing to estimate induction time have
262 proposed an automatic detection from Mean Grey Intensity (MGI) signal
263 traces [12, 13]. These works have identified that the size of the interrogation
264 window affects the induction time estimation. Large windows spatially av-
265 erage a greater area and consequently, might miss the early appearance of
266 crystals. On the counter side, small windows are prone to noise and other
267 effects such as bubbles may lead to false detections.

268 Formally, the mean grey intensity $\bar{I}(t)$ for every frame at time $t \in \{1, 2, \dots, K\}$,

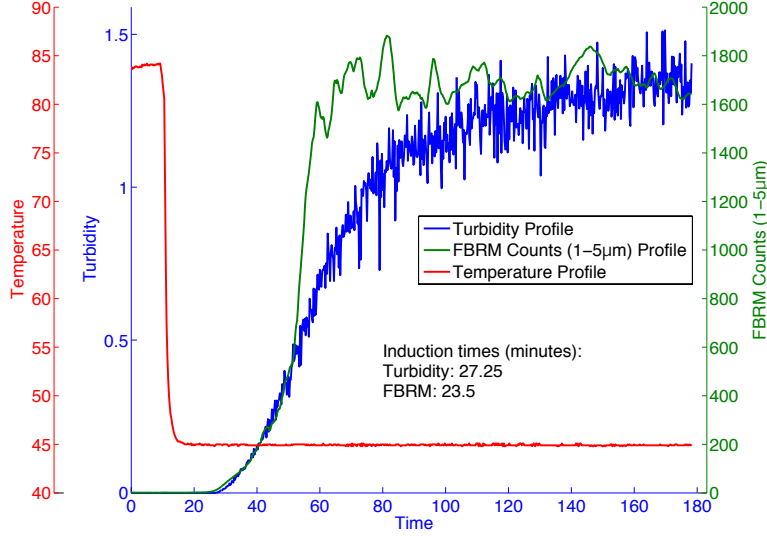


Figure 5: Temperature, Turbidity and FBRM signal traces for crystallisation of LGA in water (experiment with no fouling).

269 aS:

$$\bar{I}(t) = \frac{\sum_{i=1}^N \sum_{j=1}^M I_{ij}(t)}{NM} \quad (1)$$

270 where $I_{ij}(t)$ is the instantaneous grey intensity of the pixel (i, j) at time
 271 t .

272 Often, the limitations of MGI traces is that it dampens the early detec-
 273 tion of crystals that flow through the camera view finder making the method
 274 of estimating induction time through MGI is sensitive to the interrogation
 275 window size. The Mean Grey Intensity curves against time for various in-
 276 terrogation window sizes are shown in Figure 6. The estimated induction
 277 time for all interrogation windows is 25.75 minutes and the effect of the win-

278 dow sizes is not significant. Unlike the work presented in [12] the camera
 279 resolution used here is low and individual crystals can not be detected with
 280 confidence hence some sensitivity is lost. Also, the proposed change point
 281 detection algorithm estimates the induction time in a probabilistic fashion
 282 and even if there are some spikes in the waveform. For example for windows
 283 10x10 px and 20x20 px, the waveforms have spikes before the detected in-
 284 duction point which was most likely caused from bubbles passing through
 285 the window.

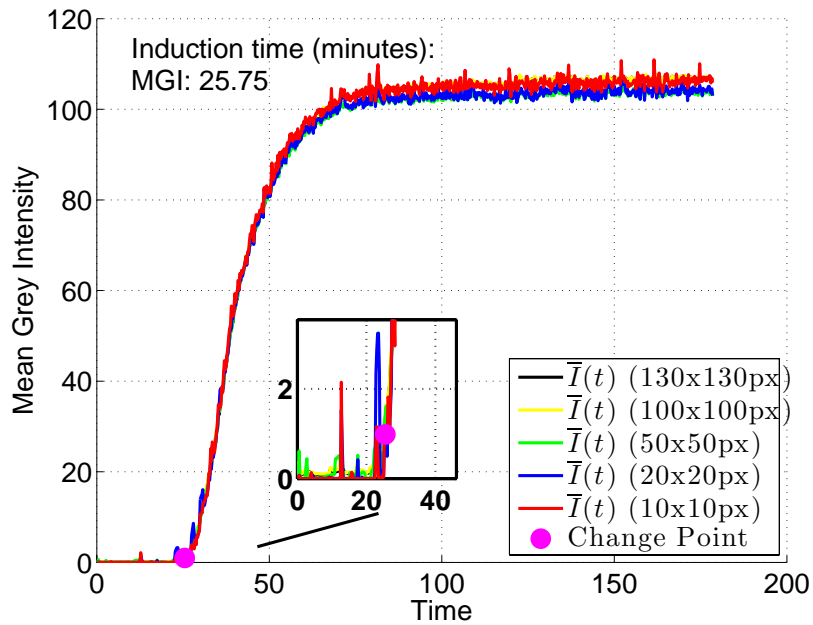


Figure 6: Mean Grey Intensity profile for various interrogation window sizes. LGA in water (experiment with no fouling).

286 Another method which could increase sensitivity compared to MGI is
 287 counting pixels above a threshold. Intuitively, the crystal appearance has
 288 started when the solution in the crystalliser gets cloudy i.e. the intensity of

289 the pixels in the frame start to rise. Frequently experimentalists capture im-
 290 ages at regular intervals to retrospectively verify the experiments. A standard
 291 method to determine process progression is to select an intensity threshold τ
 292 and at every time instance $t \in \{1, 2, \dots, K\}$, count the percentage of pixels
 293 exceeding the threshold². More formally for a threshold $\tau \in \{1, 2, \dots, 255\}$,
 294 the function counting the ratio of pixels above the threshold is:

$$C_\tau(t) = \frac{\sum_{i=1}^N \sum_{j=1}^M u_\tau(I_{ij}(t))}{NM} \quad (2)$$

295 where $u_\tau(x)$ is the step function:

$$u_\tau(x) = \begin{cases} 1 & \text{if } x \geq \tau \\ 0 & \text{if } x < \tau \end{cases} \quad (3)$$

296 Figure 7a shows the percentage of pixels above the threshold τ versus
 297 time. Applying the change point detection method on the threshold signal
 298 traces, indicates that the estimated induction varies significantly with the
 299 threshold value τ . The relation between τ and the estimated induction time
 300 is shown in Figure 7b. The induction time increases almost linearly with
 301 the threshold. Low intensity thresholds provide high sensitivity however,
 302 application of this technique on other datasets with higher image noise, did
 303 not provide a robust estimation. Low thresholds produce to spiky signal
 304 traces, especially during the beginning of the experiment where camera noise
 305 is significant leading to incorrect estimation of induction time. For high

²Note: This threshold is applied on all pixels of every frame in the image sequence, unlike the MGI comparison threshold mentioned previously for detection purposes.

306 thresholds, the crystal appearance must be well developed in order to observe
 307 a waveform rise.

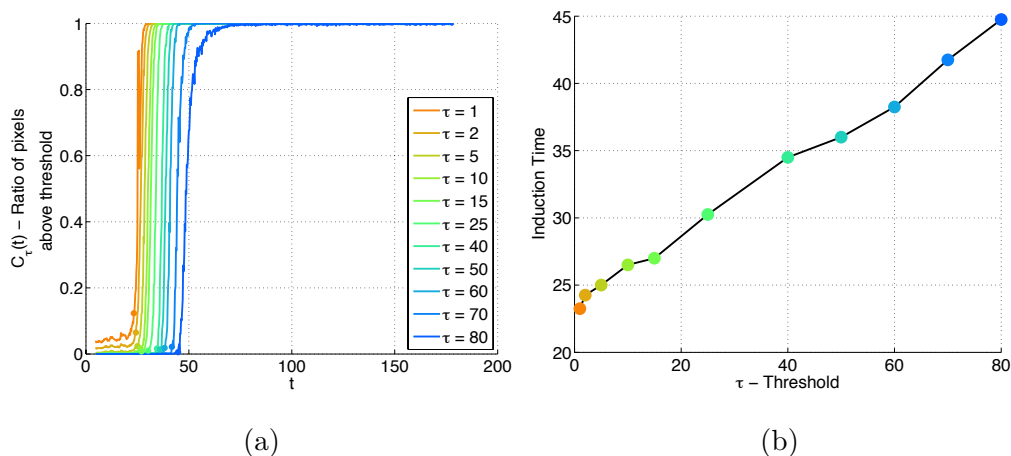


Figure 7: LGA in water (experiment with no fouling): (a) Percentage of pixels above threshold τ versus time for various threshold levels, (b) Induction time for varying threshold levels.

308 The combined induction times obtained from various signal traces when
 309 the Bayesian Online Change Point Detection algorithm is applied are sum-
 310 marised in Table 2. FBRM signal traces are the most sensitive followed by
 311 the MGI and Turbidity.

312 The turbidity probe has no means to detect fouling and hence measure
 313 the combined induction time. The FBRM probe is able to distinguish en-
 314 crustation on the probe through fouling index however, does not capture any
 315 information related to fouling on the crystalliser walls.

316 Estimating induction time through imaging contain information related
 317 to the fouling of the crystalliser walls, however this information is not being
 318 utilised.

Parameter	Induction Time (minutes)
FBRM	23.50
MGI	25.75
Turbidity	27.25
Pixel Thresholding	(23.25-44.75) Varying with threshold τ

Table 2: Comparison of induction time from various signal traces. LGA in water, (experiment with no fouling).

319 4. Fouling and Bulk Induction Time

320 Images from a commodity web camera pointing towards the crystalliser
321 walls does not only contain information about crystal appearance in the bulk
322 but also the crystalliser walls. This section presents a method to separate
323 fouling and bulk induction time through statistical analysis of the acquired
324 images as summarised in Figure 3.

325 Crystals moving through the camera view result in variations in pixel
326 intensity. Regions where crystals are present will have higher pixel intensity
327 compared to background regions. Crystals stuck on the crystalliser walls are
328 closer to the camera viewfinder and reflect more light. Crystals in the bulk
329 also reflect light leading to high pixel intensities, however the pixel intensities
330 are not consistently high for a consecutive number of frames. As the crystals
331 move away with the liquid flow intensities drop. The proposed pixel detection
332 algorithm consists of two steps:

- 333 1. Identify pixels with the highest intensity.
- 334 2. Identify pixels which have the highest intensity for a consecutive num-
335 ber of frames. This rule is necessary to avoid false positives where

336 particles larger/brighter than the encrusted region are passing through
 337 the view.

338 To achieve the first step of the process an upper outlier detection method
 339 based on Chebyshev's inequality is used [24]. The inequality provides a bound
 340 on the percentage of data point falling further than k standard deviations
 341 away from the mean. On this occasion the inequality is applied to the pixel
 342 intensities distribution of the frame at time t .

$$P(|I(t) - \bar{I}(t)| \geq k\sigma) \leq \frac{1}{k^2} \quad (4)$$

343 The inequality is used to determine upper and lower Outlier Detection
 344 Value (ODV) limits (ODV_U and ODV_L) and does not make any assumptions
 345 on the underlying data distribution. Pixels with intensity outside the limits,
 346 are classified as outliers. For $k = 5$ this leads to maximum 4% of pixels been
 347 classified as outliers on both directions. The outlier detection limits are given
 348 by:

$$\begin{aligned} ODV_L &= \bar{I}(t) - k\sigma \\ ODV_U &= \bar{I}(t) + k\sigma \end{aligned} \quad (5)$$

349 From these two detection values, only upper outliers (greater than ODV_U)
 350 indicate fouled regions and the following detection function is defined:

$$d_{ij}(t) = \begin{cases} I_{ij} & \text{if } I_{ij} \geq ODV_U \\ -Inf & \text{otherwise} \end{cases} \quad (6)$$

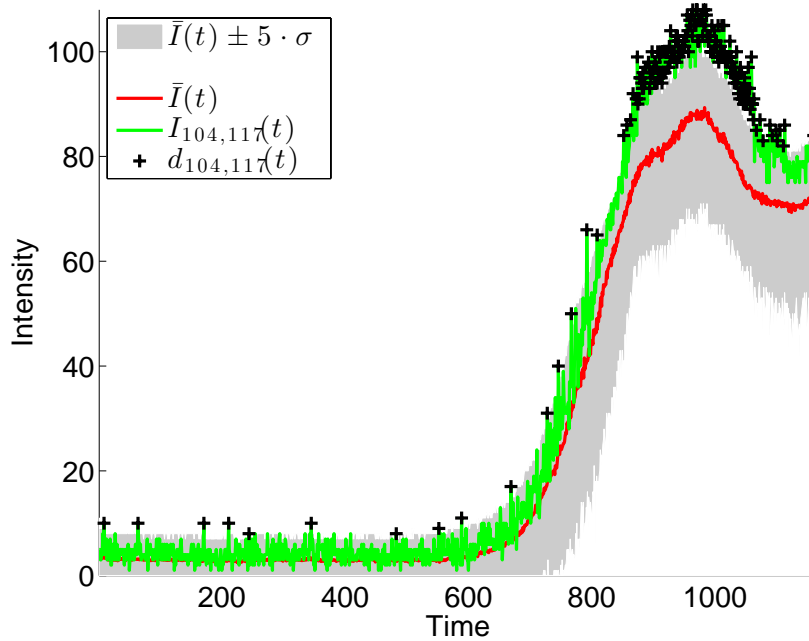


Figure 8: Mean Grey Intensity profile with Chebyshev's outlier detection bounds for $k = 5$. The green line shows the instantaneous intensity of pixel (104,117), while the black crosses indicate the points where the pixel intensity has exceeded the ODV_U ; i.e. the output of the $d_{ij}(t)$ from Equation 6. LGA in water (experiment with fouling).

351 The detection function has value of $-Inf$ for pixels that are not detected
 352 as outliers while the detection function is equal to the corresponding pixel
 353 intensity (I_{ij}).

354 Figure 8 shows in red line the MGI profile curve, while the grey shaded
 355 area shows the intensities that lie between the Outlier Detection Value Lower
 356 (ODV_L) and the Outlier Detection Value Upper (ODV_U) from Eq. 5. The
 357 green line shows the instantaneous pixel intensity of an arbitrary pixel at
 358 location (104,117). Every time pixel intensity exceed the ODV_U , the pixel is
 359 classified as an outlier as in Eq. 6 and is annotated on the graph with black

360 crosses. All pixels that have intensity lower than the upper bound ODV_U
 361 have detection value of $-Inf$. Not all pixels that the detection function
 362 has identified as outliers, are necessarily fouled. The high intensity can be
 363 caused either due to camera noise or objects passing through that pixel re-
 364 gion. However, pixels that the detection function has consistently identify
 365 as outliers; i.e. for N_S consecutive number of frames, are fouled and the
 366 following filtering algorithm is used to detect those:

- 367 1. For time step t , create a set $\mathcal{A}(t)$ with all pixels in the frame

$$\mathcal{A}(t) : \{(i, j)\}, \text{ for all } i \in \{1, \dots, N\} \text{ and } j \in \{1, \dots, M\} \quad (7)$$

- 368 2. Using the set $\mathcal{A}(t)$ estimate the ODV_U required for the outlier detection
 369 function in Eq. 6.
- 370 3. Create a set $\mathcal{F}(t)$ with all the pixels that their d_{ij} is greater than or
 371 equal to 0 for the previous $N_S = 5$ time steps/frames. Essentially,
 372 this steps looks to previous images to determine if the pixels where
 373 consistently an outlier based on the instantaneous detection function.

$$\mathcal{F}(t) : \{(i, j)\}, \text{ where } d_{ij}(t-l) \geq 0, \\ \text{for all } l \in \{1, \dots, N_S\}, i \in \{1, \dots, N\}, j \in \{1, \dots, M\} \quad (8)$$

- 374 4. The number of fouled pixels are equal to cardinality (number of ele-
 375 ments) of the set $\mathcal{F}(t) : |\mathcal{F}(t)|$.
- 376 5. Recompute ODV_U for the set of pixels in the intersection $\mathcal{A}(t) \cap \overline{\mathcal{F}(t)}$
 377 (i.e. exclude pixels that were classed as fouled in the current time t).
- 378 6. Go to step 3.

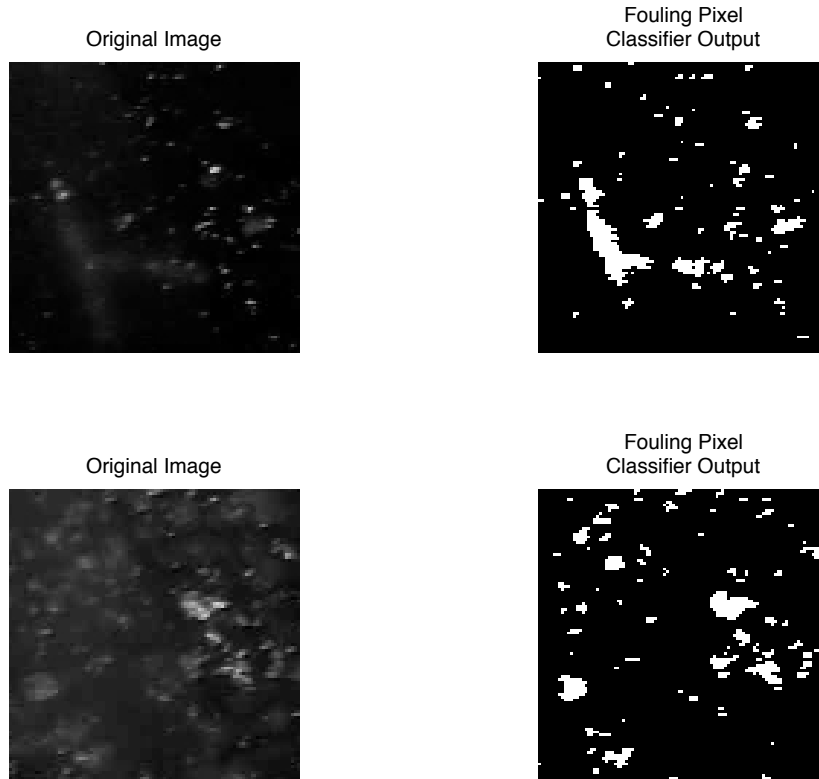


Figure 9: Two examples of original images and outputs of the fouling classifier. LGA in water (experiment with fouling).

379 The output of the classification algorithm can subsequently be used to
 380 visualise regions where fouling has occurred. Figure 9 shows two example
 381 images and their corresponding outputs of the classifier.

382 Figure 10 shows the percentage of pixels classified as fouled, against time
 383 which is obtained as the ratio of the cardinality of the set $\mathcal{F}(t)$ over the total
 384 number of pixels in the frame. The percentage of pixel can be used as a
 385 fouling index indicator at any stage of the experiment. Figure 10 also shows
 386 the combined MGI signal trace i.e. without taking in consideration the pixel

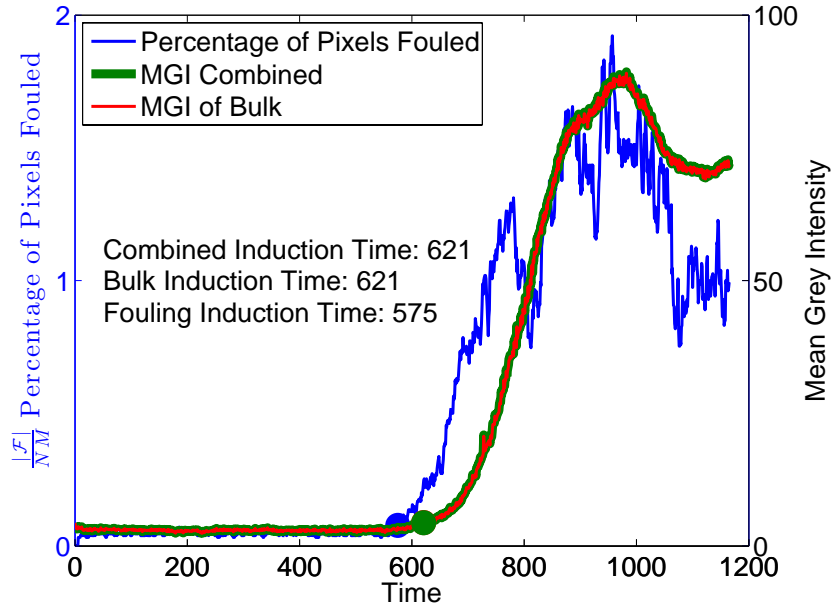


Figure 10: Signal trace profiles for fouling index, bulk MGI and combined MGI. LGA in water (experiment with fouling).

387 class as in Section 3, while the MGI bulk signal trace only considers pixels
 388 that are not fouled (i.e. belonging to the bulk).

389 In this experiment, the combined MGI and Bulk MGI are almost identical
 390 and this is due to a low number of fouled pixels (i.e. only 2% of pixels are
 391 fouled). Using the change point detection to estimate induction times from
 392 these signal traces, fouling appears at 575 seconds, while induction in the
 393 bulk occurs at 621 seconds, with identical time for the combined trace.

394 It is important to highlight that the fouling induction time is detected
 395 before bulk or combined induction times. This is justified when considering
 396 that the fouling signal trace provides higher sensitivity when compared to
 397 MGI traces. The MGI traces inherently damp localised changes until the

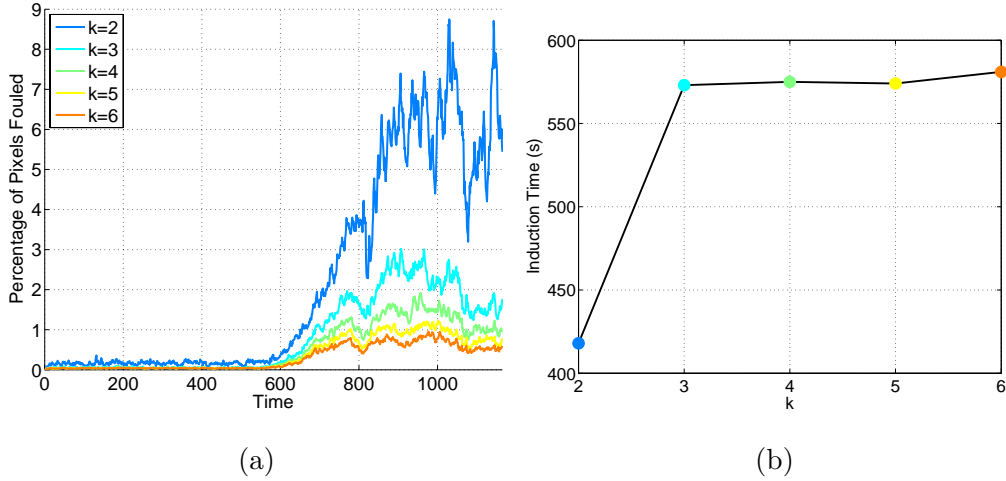


Figure 11: (a) Percentage of fouled pixels versus time for various k , (b) Induction time against k .

398 intensity of a significant number of pixels has increased. This difference in
 399 sensitivity introduces a potential uncertainty of the order of events, e.g., when
 400 induction times on the wall and in the bulk are near each other. The aim in
 401 this work is to provide an early warning system for fouling and this is achieved
 402 through the high sensitivity provided by the fouling signal trace. Measuring
 403 bulk induction time with high sensitivity could be achieved through the use
 404 of other PAT equipment such as reflectance (i.e. FBRM).

405 4.1. Sensitivity of Fouling Classifier Parameters

406 The fouling pixel classifier sensitivity can be adjusted through two pa-
 407 rameters. The outlier detection sensitivity k standard deviations and the
 408 number of consecutive frames N_S a pixel has to remain an outlier, before it
 409 is classified fouled.

410 Figure 11a shows the percentage of pixels against time for various k stan-

411 dard deviations and $N_S = 5$. The maximum percentage of pixels intensities
412 beyond $k = 2$ standard deviations from the mean is 25% hence a greater num-
413 ber of pixels have the potential to be identified from the detection function
414 in Eq. 6. Higher values of k restrict the bound and hence sensitivity of the
415 detection function. For $k = 6$ the maximum percentage of pixels is restricted
416 to 2.77%. The fouling induction time against k is shown in Figure 11b. For
417 $k = 2$ the bound is wide and pixels in the bulk are classified prematurely as
418 fouled. However, as the k increases to 3 and beyond the sensitivity reduces
419 resulting to identical induction times.

420 Similar sensitivity analysis is performed for N_S ; the number of consecutive
421 frames a pixel has to be identified an outlier for a fixed value of $k = 5$. The
422 percentage of fouled pixels against time are shown in Figure 12a. The longer
423 the time period a pixel remains an outlier the more sever the fouling. As
424 expected, the lower the N_S greater chances that the pixel intensity is an
425 outlier and consequently highlighted as fouling. As the N_S increases pixels
426 have to remain outliers for a longer period of time. The fouling induction
427 time against N_S is shown in Figure 12b. It should be noted that although the
428 algorithm is sensitive to the selection of the N_S parameter, the relative change
429 in induction time is less the 5% of the absolute induction time. Inspection
430 of the acquired images indicate that parameters $k = 5$ and $N_S = 5$ are the
431 most suitable to detect fouled pixels regions.

432 5. Conclusions

433 In this article we presented a method to automatically detect induction
434 time through Bayesian Online Change Point Detection in real-time while the

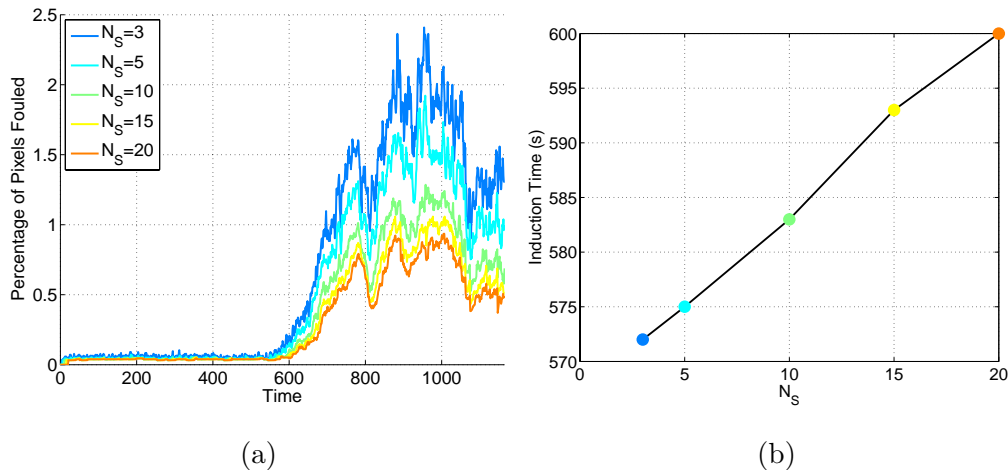


Figure 12: (a) Percentage of fouled pixels versus time for various N_S , (b) Induction time against N_S .

435 experiment is in progress. It is demonstrated that the change point detection
 436 algorithm can estimate induction time for various types signal traces such as
 437 reflectance, turbidity, Mean Grey Intensity and percentage of pixels fouled
 438 without the need to tune model parameters. This approach to estimating
 439 induction time is more robust than thresholding where the estimates can
 440 vary significantly with the threshold selection.

441 We have also developed a novel method for the early detection of fouling
 442 through commodity web cameras. Classification of image pixels correspond-
 443 ing to either bulk solution or fouled surface was achieved through statistical
 444 analysis of pixel intensity time series. The proposed technique is applicable
 445 to industrial settings where visual access to the fouling surface is available.

446 The number of pixels classified as fouled is an indicator of the degree of
 447 fouling at every stage of the experiment. Using the fouling indicator and
 448 applying the automatic change point detection, fouling induction time can

449 be estimated. We note that surface fouling and bulk crystallisation signal
450 traces provide significantly different sensitivity and that may introduce an
451 uncertainty in the order of events when induction times at surface and in the
452 bulk are close to each other.

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