AUTOMATED DROP DETECTION USING IMAGE ANALYSIS FOR ONLINE PARTICLE SIZE MONITORING IN MULTIPHASE SYSTEMS

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ABSTRACT
Image analysis has become a powerful tool for the work with particulate systems, occurring in chemical engineering. A major challenge is still the excessive manual work load which comes with such applications. Additionally manual quantification also generates bias by different observers, as shown in this study. Therefore a full automation of those systems is desirable. A MATLAB® based image recognition algorithm has been implemented to automatically count and measure particles in multiphase systems.

A given image series is pre-filtered to minimize misleading information. The subsequent particle recognition consists of three steps: Pattern recognition by correlating the pre-filtered images with search patterns, pre-selection of plausible drops and the classification of these plausible drops by examining corresponding edges individually. The software employs a normalized cross correlation procedure algorithm. The program has reached hit rates of 95% with an error quotient under 1% and a detection rate of 250 particles per minute depending on the system.

KEYWORDS
particle size distribution, image analysis, online monitoring, Sauter mean diameter, dispersion, automatic particle recognition

1 INTRODUCTION
The competitive pressure in the chemical industry makes it necessary to take measures that enable processes to be drastically improved in order to remain competitive also in the future. (Ruscitti et al. 2008). Product quality control is more complex in particulate than in conventional chemical processes. The key properties of the product are often related to the particle size distribution (PSD) which is influenced by the operating conditions and the history of the process (Zeaiter et al. 2006). Disturbances in operating conditions may irreversibly change the quality of the product. Quantitative real-time measuring is needed to enable feedback control. Monitoring and control of such processes has evoked interest in the use of image-based approaches to estimate product quality in real time and in situ (Zhou et al. 2009).

During the last decades extensive research has been performed to establish and improve technologies which measure particle properties, such as size, shape, composition, velocity, etc. Concerning the interpretation of particle size distributions using different physical principles there is still a considerable lack of understanding (Leschonski 1986).
Various authors found unsatisfying results, analyzing spherical drops in different liquid/liquid systems, using laser optical measurement techniques based on back scattering (Boxall et al. 2010; Greaves et al. 2008; Honkanen et al. 2010; Maaß et al. 2011a). These authors are questioning the reliability of these online probes in general and reaffirming the need to use image analysis (IA) instead as the particle surface unpredictably influences the signals.

A further limitation, according to different authors (Martínez-Bazán et al. 1999; Niknafs et al. 2011; Pacek et al. 1994), is the use of external physical sampling. This never can guarantee that the particle size does not change during measuring. Even for sampling times less than a second, significant measurement errors can occur. In order to get reliable drop size distribution (DSD) measurements the technique needs to be chosen carefully. This work is focused on a MATLAB® based image recognition algorithm, which is able to automatically measure particles robust in different multiphase systems.

The paper is structured as follows. The use of image analysis for sizing fluid particles is shortly reviewed in the section 2 followed by an introduction of the used experimental set-ups in section 3. Image pre-processing and image analysis size measurements are given in sections 4 and 5. The results achieved by that method are compared with manual results in section 6.

2 IMAGE ANALYSIS IN MULTIPHASE SYSTEMS

Although process characterization based on image analysis (IA) can be intensely time consuming, it needs to be applied to almost every dry and wet particulate system. A short summary of published applications in liquid systems is given by Guevara-López et al. (2008) and for dry systems by Andres et al. (1996). Emerging applications show that utilization of image analysis can facilitate the creation of new and sophisticated models for the control of particulate systems (Williams and Jia 2003). In this paper we only focus on the characterization of the size of particles based on IA.

The most extensive available review on this specific field is given by Junker (2006). A detailed description of the technical and historical developments can be found there. This review also shows that the photo-optical in-situ measurement of particle sizes in multiphase systems is already well established. Many applications have been reported in literature with different set-ups (Aakre et al. 2005; Alban et al. 2004; Fantini et al. 1990; Galindo et al. 2005; Hossain et al. 2011; Junker et al. 2007; Kamel et al. 1987; Khalil et al. 2010; Mickler et al. 2011; O’Rourke and MacLoughlin 2005; Roitberg et al. 2006; Torabi et al. 2005), all of which worked well for the applications investigated. They are based on digital, high-speed, high resolution modular camera systems and the images are analyzed with commercial or self developed image analysis software and standard statistical methods.

Junker (2006) gives an organized overview of the applied photographic techniques used in literature. They stated that CCD-cameras are the optimum for the effort / cost ratio. Figure 1 shows an example image gallery from such a standard camera. Pictures are taken at a maximum frequency of 50 frames per second with this technique which is equal to a recording time, also called data acquisition time (DAT) of 1/50 s per image. Already Leschonski (1986) and also Junker (2006) emphasize the necessity of short DAT and additionally a short measurement acquisition time (MAT). The MAT is the DAT plus the time needed to extract the necessary information from a sample image and translate this information into the particle size distribution. Ideally the MAT equals the DAT (Crawley and Malcolmson 2004).
To get statistically reliable data sets 100's or 1000's of particles have to be measured. Particle systems are very different and so are the number of particles on one image (see Figure 1) and therewith the number of frames required for a single measurement. A minimum of 250 drops need to be captured within some seconds to achieve real time DAT. Junker (2006) reports in her studies a variation between 2 to 400 frames. The required number of objects per measurement becomes important for storage reasons, if all images need to be reviewed and therefore saved. This problem should become less and less significant with the ongoing developments of computer hardware.

Usually for particle sizing applications the MAT (5 to 60 min) is much greater than the DAT (Junker 2006). These time ranges are unsuitable for process monitoring and control. The manual evaluation of such images is highly time-consuming and therefore automation of image analysis should be employed to speed up the MAT at least one to two magnitudes. Another disadvantage of manual evaluation was firstly reported by Gwyn et al. (1965). Due to the subjective nature of manual particle counting the measured distributions are human biased. The significant statistical variations mostly occur especially at the "tails" of the distributions. Boxall et al. (2010) also showed the influence of human bias with an average difference of 5.1% in the average drop size for two different analyzers. Automated quantification would avoid bias by different observers.

Simplified image analysis for the discussed multiphase systems can be achieved by using only low dispersed phase fractions in which no overlapping occurs. Several examples for this approach are reported in literature (Khalil et al. 2010; Mickler et al. 2011; Scherze et al. 2005). These successful implementations of image analysis algorithms all fail for highly concentrated (phase fraction larger than 10 to 15 %) dispersions. Additionally no commercial software for the
analysis of such systems, which would be needed for industrial relevant applications is currently available (Brás et al. 2009). More promising are the works of Alban et al. (2004) and Brás et al. (2009).

The image analysis technique employed by Alban et al. (2004) includes several steps of arc and circle centre detection and pattern matching. The number of particles detected in the image depends on the image quality. Detection levels vary from 10 to 90%. Large particles are accurately detected even when they are obstructed by smaller particles. Errors in detection occur especially for very small particle sizes when thick particle peripheries are present. This does not significantly affect the used Sauter mean diameter ($d_{32} = \frac{\sum d_i^3}{\sum d_i^2}$), due to emphasis of large particles in this calculation. Brás et al. (2009) are using a two step approach that automatically identifies the contour of existing drops and classifies them according to their diameter. In the first step, they detect the edges of the drops in the original image by monitoring the values of the image grey values as well as the descending thickness and by creating an output image with those contours. In the second phase, they detect the drops in this contour image, using the Hough transformation (Cohen and Toussaint 1977) to evaluate the circles.

Although both works use automated drop size analysis, both are not capable for process monitoring or control as both have too large MAT. One goal of this work was to decrease the MAT significantly to achieve online monitoring of the particle size distribution in a multiphase system.

3 MATERIAL AND METHODS

The objective of these investigations was to develop an image analysis algorithm, which allows precise automatic particle size measurement in real time. The accuracy and the online capability of the particle size monitoring were tested in various stirred applications.

3.1 Experimental set-up

Figure 2 shows the main features of the used experimental set-up. The images are taken intrusively from inside the vessel by placing an endoscope in front of a CCD camera as a microscope lens (see Figure 2). A strobe flash is guided by a fibre optic cable surrounding the endoscope to ensure sharp pictures even in vicinity of the stirrer (Maaß et al. 2011a).

A broad evaluation is necessary to ensure the reliability of the image algorithm software. Often the software is developed and optimized only according to the application in its founding working group. The major challenge is to develop a software program which can be adapted to many differing multiphase systems. Therefore, four different dispersed phases were used for the validation of the developed software. Deionised water always served as the continuous phase. An overview of the work program is given in Table 1. The dispersed phase fraction $\phi$ was varied from 2 to 45 percent to analyze the influence of the concentration, which influences the optical properties dramatically. Dilute systems like the one shown in Figure 1(a) have almost no particle overlapping and are therefore easier to analyse. However, the more concentrated systems ($\phi > 5\%$) have one advantage. The drops are so dense packed, that all visible particles are at the vicinity of the focal plane of camera lens. Therefore no correction for possible out of focus depths (Kashdan et al. 2003) are necessary.
3.2 Measurement procedure

Every image batch according to the individual system was analyzed twice; the first counting was always carried out manually to build a base for evaluation of the second counting, which was carried out automatically with image analysis software. The time needed for one data point is around 30 min. The influence of human bias on drop size measurements were tested first. The deviations between four different observers with each other and also with themselves through a repeated second counting of the Sauter mean diameter $d_{32}$ are shown in Figure 3. The results of the $d_{32}$ are presented on the left ordinate and the number of counted particles $n$ on the right ordinate. All participants counted the same frame set twice. An example image from the chosen analyzed frame set is given in Figure 1(a). The image quality is very high due to the high refraction index of the used system (see Table 1) and the low dispersed phase fraction ($\varphi = 2\%$). That is why almost no overlapping effects occur. Even with those "simple" images the four different observers have a deviation of $\pm 5\%$ for the Sauter mean diameter and of $\pm 15\%$ for the number of particles found in the whole set (presented as the black squares). Surprisingly these deviations are increased by repeating the same counting for both the $d_{32}$ ($\pm 10\%$) and the number of identified particles ($\pm 30\%$). The standard deviation of one observer quantifying manually the same frame set is around 5%. These deviations are called human bias in this work as they show a distortion which is not systematic but influenced by the observing human and his experience and mental state. The newly developed image analysis algorithm will overcome these weaknesses of human bias. All distortion will be systematic and therewith interpretable or even avoidable.

The image processing and detection steps are described in section 4 and 5. For all following comparisons between the automated and the manual counted drop size distribution results, the observer (four) with the high reproducibility is used.
Table 1 – Overview of the working program, testing the image algorithm software

<table>
<thead>
<tr>
<th>dispersed phase</th>
<th>refraction index n (-)</th>
<th>φ (-)</th>
<th>T (mm)</th>
<th>H/T</th>
<th>D/T</th>
<th>origin of image data</th>
</tr>
</thead>
<tbody>
<tr>
<td>toluene</td>
<td>1.496</td>
<td>0.02, 0.10</td>
<td>150</td>
<td>1.0</td>
<td>0.33</td>
<td>this study and Maaß et al. 2011a</td>
</tr>
<tr>
<td>petroleum</td>
<td>1.428</td>
<td>0.10</td>
<td>150</td>
<td>1.0</td>
<td>0.33</td>
<td>Maaß and Kraume 2011</td>
</tr>
<tr>
<td>n-butyl chloride</td>
<td>1.402</td>
<td>0.45</td>
<td>155</td>
<td>2.3, 5.0</td>
<td>0.6</td>
<td>this study and Maaß et al. 2010</td>
</tr>
<tr>
<td>glass beds</td>
<td>1.485</td>
<td>0.10</td>
<td>200</td>
<td>1.0</td>
<td>0.31</td>
<td>Angst and Kraume 2006</td>
</tr>
</tbody>
</table>

Figure 3 – Analysis of the variance of the Sauter mean diameter and the number of observed drops by human bias in a single image batch at constant process parameters.

3.3 Computational specifications

The calculations were carried out on an Intel Quad-Core i7 with 2.6Ghz and 12GB RAM. The algorithms were implemented in Matlab 7.9 (R2009b). On this PC running Windows Vista, the developed software was able to reach a processing speed of 190 ms/frame for simple images. 50 to 150 images were taken for every investigated point in time and analyzed as complete batches. Following the guidelines given by Yan et al. (2009a), section 4 describes the image operations carried out for noise removal, sharpening and contrast enhancement.

4 IMAGE PROCESSING

The quality and variability in quality of the images obtained by the camera computer system greatly influences the success of the automated interpretation (Yan et al. 2009b). The first step
of the used method essentially consists of enhancing image information to facilitate its analysis which is the second step and is described in section 5.

Images were at first processed by modifying their intensity distribution to enhance certain features of the image. The processing technique involves several steps of contrast enhancement. In order to ensure best possible drop detection, the given image series are pre-filtered to remove irrelevant and misleading image information. The redundant information, like lighting patterns and dirt or particles on the lens only given by the optic system, can be removed by subtracting it from every image. Therefore the cumulative pixel by pixel average image is formed out of the whole batch of $n_B$ images $B_i$ (see eq. (1), with $n_B = 50$ for this example):

$$S = \frac{1}{n_B} \sum_{i=1}^{n_B} B_i$$

The corrected image $B'_i$ is therewith the subtraction of the original image $B_i$ (see Figure 4 - a) with the arithmetic mean image $S$ of the whole sequence (see Figure 4(c)), which is an approximation of the redundant information.

$$B'_i = B_i - S + \bar{S}, \quad \bar{S} - the \ average \ intensity \ of \ S$$

The result of eq. (2) is presented in Figure 4(d).

![Figure 4](image_url)

**Figure 4 – Image processing steps to remove redundant information during pre-filtering:** (a) and (b) example images from a whole set, (c) integrated average image from this set, (d) subtracted image (a) minus (c). Used system is n-butyl chloride/ water; dispersed phase fraction $\varphi = 45\%$, $d_{32} \approx 80 \mu m$.

Every pixel at position $(x,y)$ in the pixel matrix of the filtered image $B'_i$ is multiplied with the quotient of 255 and the maximum grey value of this matrix to use the whole spectrum of a gray value image. This standard amplification is shown in eq. (3) and Figure 5(b).
The robustness of automatic detection of structures in noisy pictures can be increased by the self quotient image method (SQI) (Gopalan and Jacobs 2010; Shashua and Riklin-Raviv 2001). These operation norms the intensity of every local pixel based on the local environment. This is carried out by a division of the processed image $\hat{B}_i$ pixel values by those of a smoothed version $X$ of itself (see Figure 5(c)):

$$Q(x,y) = \frac{\hat{B}_i(x,y)}{X(x,y)} \text{ with } X = B * K,$$

where $K$ is a 2-dimensional Gaussian kernel and * the convolution operator.

![Figure 5](image)

**Figure 5** – Image process steps under SQI: (a) one example original image, (b) image after image processing steps illustrated in Figure 4 plus a contrast amplification, (c) convolution of image (b) with a 2dimensional Gaussian kernel, (d) result of the self quotient (b)/(c).

The quantification of the influence of every neighbour particle at every coordinate is related to the convolution kernel $K$. The image $Q$ (Figure 5(d)) is created by emphasizing the changes of the intensities. This image is now not dependent on any illumination variation (see Figure 5) and processed to a stage where it can be analyzed.

**5 IMAGE ANALYSIS**

The particle recognition, presented in this section, essentially consists of three steps: The pattern recognition by correlation of pre-filtered images with search patterns, the pre-selection of plausible circle coordinates and the classification of each of these by an exact edge examination. These individual steps include the compilation of the search patterns for correlation.
5.1.1 Pattern compilation

A necessary condition for coherent results of the algorithm is a precise knowledge of the expected gray values to design meaningful search patterns. From example drops, which the user gives to the program, these drop patterns are automatically computed by the software for every representative optical system. It is necessary to provide example drops over the whole spectrum of expected sizes, since the drop appearance cannot be assumed to be equal or linear scalable over the radius r. Such example drops are given in Figure 6.

After the definition of example particles, the intensity signature curves of these examples are computed. The signature curve is the average value $b_j$ of the gray values in the image $B_j$ of a chosen object $j$ with the radius $R_j$. It is computed over a circumference within the image coordinates $B(x;y)$ at every radius $r$ which are for simplification treated with polar coordinates $(r, \alpha)$:

$$b_j(r) = \frac{1}{2\pi} \alpha \sum_\varphi B_j(r, \alpha), \quad \alpha = 0^\circ, 360^\circ, r = 1, 2, ..., \left\lfloor \frac{1}{2} R_j \right\rfloor$$  \hspace{1cm} (5)

Figure 7 displays the results of $b_j$ for all example drops from Figure 6. For comparison the distribution of $b_j(r; R_j)$ over the radius for different drop sizes, $r$ is always normed to the absolute radius $R_j$ of the corresponding toluene drops. Therefore the drop border is always at $r/R_j = 1$. The distribution are qualitatively similar in the region of $r/R_j = 1$ but different for the rest of $r$. The absolute values for $b_j$ at a constant $r$ for different drop sizes also vary with no certain rule.

The qualitative similarity at the boundaries of the different drops makes it possible to compute a pattern with an ideal distribution of $b_j$ over $r$. Such a distribution is shown in Figure 7 for the example drop pattern $T(r, \alpha)$ with a radius $R = 102$ pixel. Such patterns are then generated automatically for every possible drop size in a user defined diameter range and are used for correlation with the gray value distributions of all processed images.
Figure 7 – Signature curves $b_j$ of all example drops from Figure 6. Additionally the pattern $T(r,\alpha)$ for $R = 102$ pixel based on an interpolation of several drop examples is given in comparison with the example drop image $B(r,\alpha)$.

### 5.1.2 Pattern matching

Although the description of the signature curves $b_j$ in polar coordinates is more intuitive, the computation is carried out in Cartesian coordinates as images are organized in discrete elements following a Cartesian order. Therefore the following procedures are explained as they are implemented.

There are different image algorithms for structure detection available. In this project, two different principles are used for the analysis: a voting and a correlation procedure (CP). A frequently used CP is the normalized cross correlation (NCC). It is defined by Lewis (Lewis 1995) and shown in eq. (6). The pattern $T(x,y)$ defines a sought-after example structure in the image $B(x,y)$, in our case a gray value matrix which includes a particle boundary with a certain radius $r$. The distribution of $b_j$ for a pattern $T(x,y)$ of the radius $r = 102$ pixel is given in Figure 7. It is compared with the distribution of the example drops and also given in direct comparison with the correlating image segment $B(u,v)$ for a radius $r = 102$ pixel. Those are correlated with the NCC:

$$V(u,v) = \frac{\sum_{x,y} [B(x,y) - \bar{B}_{u,v}]T(x-u,y-v) - \bar{T}_{u,v}}{\sqrt{\sum_{x,y} [B(x,y) - \bar{B}_{u,v}]^2 \sum_{x,y} [T(x-u,y-v) - \bar{T}_{u,v}]^2}}$$

Here $\bar{B}_{u,v}$ is the average of the gray values below the sample pattern $T$ and $\bar{T}_{u,v}$ the average value from the sample pattern. The result $V(u,v)$ is a similarity measure for the wanted sample pattern $T$ of each local picture segment $B_{u,v}$, which is centred around $(u,v)$. The solution range is limited to -1 to 1. Maxima suggest an agreement with the sample pattern, negative values an opposed match.
The search for circular patterns for a range of possible radii is the most time consuming and critical process of the detection. The results of $V(u,v)$ for sample pattern $T$ with the radius 20 pixel is given in Figure 8(a). The intensities behave proportional to the centre probabilities, white being the lowest and the strongest tone (black) being the highest probability.

![Figure 8](image)

*Figure 8 – Original image (a) and the self quotient image for comparison (b); the results for the normalized cross correlation with a 20 pixel radius pattern (c) and results of the drop detection for all radii (d), the resulting from $T(r=20)$ are highlighted by a dotted line.*

The best possible centre points (these are local probability maxima) and radii are then checked in detail to finally be classified. This is done by decomposing the possible drop circle into segments and checking these for local validity of being part of a real drop. Note that thanks to the contrast enhancement, borders can be evaluated with image illumination invariance. The percentage of features needed to pass as circles are very much dependent on the optical characteristics of the system. The results for the detection after the classification are given in Figure 8(b) and compared with the processed original image Figure 8(c).

6 RESULTS AND DISCUSSION

6.1 Analysis of a solid-liquid system

Solid spherical glass particles were analyzed by the presented technique and the spheres were detected by the image processing and analysis techniques. The number of particles detected in one image was roughly 10 (see Figure 1(b) as an example picture). Figure 9 shows the comparative results between estimated distributions based on manual and automated detection and the number of detected particles $n$ in the whole frame set of 82 images. Both show almost the same results. The only differences are observed for the larger spheres which lead to a
deviation of the Sauter mean diameters of around ± 20%. The processing speed was maximized to facilitate online monitoring. The overall time for acquisition, processing and analysis of the whole batch of images is below 180 s.

![Figure 9 - Comparison of manual and automatic detected particle size distributions for bimodal glass beds from a frame set with 82 images.](image)

### 6.2 Analysis of liquid-liquid systems

#### 6.2.1 Dilute liquid-liquid system

The already introduced "simple" system with a low dispersed phase fraction was used (toluene/water; \( \varphi = 2\% \); see Figure 1(a) as an example image).

After optimizing the parameter set of the algorithm, quite promising results could be achieved. The cumulative number distributions \( Q_0(d_p) \) shown in Figure 10 are almost identical for the manual (man.) and the automatic (autom.) detection. They are presented by the dotted curves over the particle diameter for the right ordinate.

For a better differentiation the number density distribution \( q_0(d_p) \) is plotted on the left ordinate, presented as straight lines. The broadness of the distribution is slightly different and this becomes obvious by comparing the number density distribution of the automatic and the manual detection. The automatic counting shows a slightly more narrow distribution. That concurs with a more than doubled number of detected particles for the automatic detection (\( n = 553 \)) in comparison to the average value of the manual counted (\( n = 301 \)). Those deviations lead to a difference of 2.7 percent in the Sauter mean diameter.
6.2.2 High concentrated liquid-liquid systems.

The size of drops in agitated vessels is the result of two opposing phenomena: drop breakage and coalescence. When agitation starts, the bulk of the dispersed phase is pulled down into the continuous phase and is broken into small drops. The rate of drop break-up in this stage is much larger than the rate of drop coalescence. As a result the size of drops shows an exponential decrease during the transient stage (see the example for petroleum/water or n-butyl chloride/water in Figure 11). The rate of drop coalescence rises with the increasing number of drops with time during the transient stage. Eventually a steady state is reached where the rates of drop break-up and coalescence are balanced. To reflect this transient behaviour, 200 pictures were taken at several points in time over 60 or 80 min of stirring. The algorithm is able to count more than 250 drops on the digital images within 2 min at each time of measurement. For every different optical system, parameters for the pattern have to be adapted. To analyze the images effectively a single parameter optimization varying from 10 minutes (petroleum) to 4 hours (n-butyl chloride) was carried out. The algorithm also delivers the opportunity of suggesting those parameters based on 10 to 50 manually counted example particles. This automatic parameter generation was used for the toluene/water system.
Figure 11 – Transient behaviour of petroleum and n-butyl chloride drops after 1 and 60 min of stirring.

In Figure 12 the results for transient Sauter mean diameter are shown, for both the manual and the automatic counting of drops, for all three systems. It shows clearly that, despite numerous overlapping of drops in the images obviously occurring in such highly concentrated dispersion, the automatic algorithm works very well. It obtains almost the same values for the transient Sauter mean diameter that the manual observation for the three different systems attained.

The results for the toluene/water system are outstanding, taken into account that only once a little number of example drops had to be marked and all further data points were computed fully automated. The measured values of the algorithm correspond to the expected development of the Sauter mean diameter whereas the manual counting shows unexpected fluctuations. This can be explained by the fact that the absolute number of particles was higher with the automated analysis and therewith the confidence level of the data set.

Example pictures of the petroleum/water system (see Figure 11) show, how difficult it is even for humans to identify the drops. The drops both reflect strongly and are low transparent at the same time. Thus chaotic and ambiguous shade effects occur. In each picture there are more than 50 drops (see Figure 11(b)), but only approximately 5% of them can be clearly analyzed by humans. However, the results of the manual and automatic counting are in good agreements.

Notice that the automatic counting only needed 15% of the manual counting time. The best results were achieved for the n-butyl chloride/water system. The results show lowest deviation between the two methods (man. & autom.). The steady state values for n-butyl chloride/water after 60 min of mixing have a deviation of 4% (d_{32,man.} = 175.8 µm; d_{32,autom.} = 182.9 µm) for petroleum/water 6.8% after 80 min stirring and for toluene/water 4.6%, also after 80 min of stirring.
6.3 Drop size monitoring

Finally a drop size monitoring over 120 min with the n-butyl chloride/water system ($\varphi_d = 45\%$) was carried out. The results of this mixing experiment are shown in Figure 13. Two endoscope probes were used in parallel. The data from endoscope A are analyzed manually while the data from endoscope B are determined with the presented image algorithms. No additional parameter adaptation was carried for the algorithms as the values from the former n-butyl chloride experiment were used. Two stirrer speeds have been investigated. The first period of mixing over 60 min was carried out under a stirrer speed of 350 rpm while the second 60 min were agitated with a lower stirrer speed of 250 rpm.

With the use of this set-up, the spatial distribution of the drop sizes, the influence of the stirrer speed and the quality of the automated image algorithm for drop size monitoring was evaluated.

The discussion of the results will be subdivided as the two stirrer speed regimes. The mixing during the first 60 min showed clearly two opposed dispersions. The expected oil in water (o/w) emulsion was observed at the bottom of the reactor but a water in oil (w/o) emulsion occurred at the top. Due to the density differences of the two used organics, the oil phase became the continuous phase at the top, while the water became the continuous phase at the bottom. However, the stirring over time was able to distribute the dispersed phase homogeneously throughout the reactor. That led to a change in the emulsion characteristics which were monitored at sampling point A. The w/o dispersion turned into a water in oil in water (w/o/w) double emulsion. That is reflected by an increase in the Sauter mean diameter, starting at minute 5 for the next 20 min in Figure 13 for the upper sampling point. Only the outer drop sizes of the o/w/o drops were counted for the determination of the $d_{32}$. With increasing stirring time, the double emulsion broke up and became the expected o/w dispersion.
As the oil phase was not homogenously distributed in the beginning of the process, low oil concentrations at the bottom can be assumed. Therefore, the drop sizes are smaller than expected in the beginning of the process at sampling point B. After a homogeneous distribution of the oil phase is reached (around the point in time of 40 min), the drop sizes at the bottom of the reactor increase. This seemingly surprising result can easily be explained with the local increase in dispersed phase fraction. This reason for a drop size increase even for coalescence hindered systems is in good agreement with other studies in our research group (Kraume et al. 2004; Maaß et al. 2011c) and also with studies from other scientists (El-Hamouz 2009; Khakpay and Abolghasemi 2010; Razzaghi and Shahraki 2010).

The results for the drop sizes after the stirrer speed change are not controversial with other results. The decrease in the energy dissipation rates leads to a decrease of the breakage rate in the system under a critical value. Therefore, no further drop size decrease was observed. As the system was stabilized using 1 mg polyvinyl alcohol per 1 g n-butyl chloride, also no coalescence occurred. This is in good agreements with recent studies (Maaß et al. 2011b). The manual as well as the automated analysis reflect this behavior. The size deviation between both counting methods is lower than 10%. The preciseness of the algorithm results is in the bandwidth of the expectable measurement error range described in literature (Maaß et al. 2011a; Ritter and Kraume 2000). Although the automated detection introduces a small deviation to the manual determination, it allows a continuous drop size monitoring. With the help of the automated analysis the computer generated drop size results for every minute throughout the process. 100’s
of 1000’s of drops have been measured each time. That would not be possible with manual work.

All results obtained for the liquid-liquid systems are summarized in a parity plot in Figure 14. Almost all deviations between manual and automated detection are lower than 10 percent.

![Figure 14 – Parity comparison of all results obtained for different liquid-liquid systems.](image)

7 SUMMARY AND OUTLOOK

Image analysis has become a powerful tool for the work with particulate systems, occurring in chemical engineering. A major challenge is still the manual work load which comes with such applications. Additionally manual quantification also generates variance by different observers, which is shown in this study. Therefore the full automation of those systems is desirable. In this work we have introduced an image recognition tool to automatically count and measure particles in multiphase systems. Up to now this software is limited to spherical particles but it will be extended to irregular shapes as well.

In each of the processes, parameter optimization is necessary to save computing time and improve error quotient and hit rate. This is achieved by marking a few drops manually and letting the software extract their optical characteristics. These include the steepness and regularity of the particle border as general optical behavior. These values vary strongly from system to system but in one particular system solely depend on the drop radius.

The automatic drop recognition is most effective with high contrast backlight images with a high drop density. The high contrast ensures a good hit rate and low error quotient while the density ensures that the computation renders more drops recognized per minute. Nevertheless the proposed software was proven to be very robust and able to compete with the human eye in complex images. The computation time is nearly proportional to the pixel-quantity being
processed and is depending on the images twice to fifty times faster than manual counting. The user is able to define thresholds for the error quotient and the hit rate in order to guide the program into further parameter optimization.

The program has reached hit rates of 95% with an error quotient under 1% and a detection of 250 drops per minute for simple images and hit rates of 40% with an error quotient of under 5% and a detection of 10 drops per minute for some difficult images. A parallelization on different cores in a multi-core PC will decrease those calculation times dramatically. Therefore online process observation and control is possible.

**SYMBOLS**

- $B_i$: individual original picture
- $\hat{B}_i$: processed image
- $b_j$: average gray value
- $d_{32}$: Sauter mean diameter (m)
- $D$: stirrer diameter (m)
- $d$: Euclidean distance (pixel)
- $H$: liquid level height (m)
- $K$: convolution core
- $n$: number of particles (#)
- $n$: refraction index (-)
- $n_B$: number of images in one batch (-)
- $q_0$: number density distribution (1/m)
- $Q$: self quotient image
- $Q_0$: cumulative number distribution (-)
- $R_j$: absolute Radius of an individual particle (m)
- $r$: control variable
- $S$: cumulative average image from a whole sequence
- $S$: average pixel value of S
- $s$: stirrer distance (m)
- $T$: pattern
- $T$: vessel diameter (m)
- $T_{u,v}$: average gray value from the according pattern
- $V$: result for NCC
- $X$: convoluted image
- $\alpha$: angle (°)
- $\varphi$: dispersed phase fraction (-)
ABBREVIATIONS

CP   correlation procedure

NCC  Normalized Cross Correlation

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LITERATURE


