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An Automatic Algorithm for Determination of the Nanoparticles from TEM Images Using Circular Hough Transform

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Highlights

- Automated measurement of the number and the average primary diameter of the nanoparticles from TEM images
- This method is based on the modified version of the hough transform with pre-processing modifications on TEM images.
- The method has been tested on several TEM images with different complexity in the images.
- It has less than 5% difference over 11 different TEM images, relative to manual sizing

• Abstract

Nanoparticles have a wide range of applications in science and technology, and the size distribution of nanoparticles is one of the most important statistical properties. Transmission electron microscopy (TEM) or X-ray diffraction is commonly used for the characterization and measuring particle size distributions, but manual analysis of the micrographs is extremely labor-intensive. Here, we have developed an image processing algorithm for measuring

particle size distributions from TEM images in the presence of overlapped particles and uneven background. The approach is based on the modified circular Hough transform, and pre and post processing techniques on TEM image to improve the accuracy and increase the detection rate of the nano particles. Its application is presented through several images with different noises, uneven backgrounds and overlapped particles. The merits of this robust quantifying method are demonstrated by comparing the results with the data obtained through manual measurement. The algorithm allows particles to be detected and characterized with high accuracy.

Keywords: *Hough Transform, nano particle, TEM image, image processing*

1. Introduction

Nano-particles that are used in a wide range of applications as a result of possessing unique properties arising from their nano-scale dimensions. They are going to be of significant current interest in a wide range of applications as a result of their chemical [Sen et al., 2015], optical [Vishwas et al., 2014], magnetic [Hatamie et al., 2016], mechanical [Ribeiro et al., 2015], thermal [Fu et al., 2013] and electronic [Kim et al., 2013] properties. Depending on the applications, the size and distributions of the nanoparticles are a primary concern, and the uniformity of the dispersed particles, highly effects on the quality of the product performance.

The methods and systems that can rapidly assess dispersed nanoparticle populations are highly desirable [Nontapot et al., 2013]. To analyze the nano-particle distributions, typically transmission electron microscopy (TEM) is used as a comprehensive technique for nanoparticle characterization, which relies on the acquisition of the images at scales approaching a single atom; once a representative group of images is obtained, the next task is to explore the characterization and the statistical analysis on the size distribution of the nano-particles. The statistical data of the nanoparticles are included counting as many particles as possible, ideally a few thousand. Primary particle diameters and counting step are usually measured manually from digitized TEM images, which are still common in the literature [Trandafilović et al., 2012; Ancona et al., 2014; Dastanpour and Rogak, 2014; Lovell et al., 2015; Zhou et al., 2015]. Nevertheless, such manual sizing methods are exceedingly laborious and time-consuming, and often result in a few hundred or fewer particles being counted, hence yield poor statistics despite all one's effort. In addition, this procedure is inherently open to human errors and thus is subjective [Woehrle et al., 2006; Gontard et al., 2011].

According to the above reasons, image processing methods are of interest in counting a relatively large number of particles, and can afford more accurate morphology characterization of nano particles, which have done recently on TEM images [Grishin et al., 2012; Bescond et al., 2014; Oshida et al., 2013; Karin et al., 2013]. But even using digital image processing tools, the quantification of the sizes and distributions of nanoparticles is a difficult task. That is due to the very small size of the objective particles of interest, which particles may be in contact or overlapped with each other. This difficulty is exacerbated by the fact that the TEM image may be corrupted by some fluctuations, arising in the imaging process, or during its transmission, or even during reproduction of the image.

In the following, the state-of-the-art of image processing methods for analysis the nanoparticles in TEM images, are discussed. Pyrz et al. [Pyrz and Buttrey, 2008] performed a nanoparticle size distribution analysis to study the capabilities of nanoparticle synthesis. Based on their results, for effective nanoparticle characterization, the proper selection of imaging type (bright vs dark field), magnification, and analysis method (manual vs automated) is critical for the subsequent analysis efficiency, and the proper determination of the particle-background boundary.

Karin et al. [Karin et al., 2013] focused on the particulate emissions from diesel engine, where PM (Particulate matters) structures were presented with TEM images. They used an image processing method, including black and white operation to estimate the distribution of carbon platelet length and atom density of PMs.

De Temmerman et al. [De Temmerman et al., 2014] developed a semi-automatic image processing method in which the primary particles are detected based on watershed segmentation. In their study, the minimal size and overlap coefficient were measured based on a Euclidean distance map. The focus of their research was to measure the minimal size in one dimension of primary particles in aggregates.

Du [Du, 2015] used low-pass filter and Wiener filter to denoise the TEM images. His results showed that the developed nonlinear filtering algorithm, can efficiently reduce the noises without noticeable artifacts with the contrast of variation in the background and defects. However, his algorithm is particularly suitable for quantitative TEM image, but cannot perform statistical analysis and the size distribution of the nanoparticles.

One of the most recent studies of this subject, has been performed by Dastanpour et al. [Dastanpour et al., 2016]. They developed a method for automatic determination of the average primary particle diameter based on the variation of the 2-D pair correlation function at different distances from the main skeleton of the aggregates. The accuracy of their method is comparable to the best available algorithms for primary particle sizing which can be used for analysis of TEM images, but their method cannot provide information on the size distribution of the particles in TEM images.

The above-mentioned studies either focuses on reducing the noises or object determination in nanoparticles of TEM images, while all methods have been performed in the special domain (matrix form). For the applications of TEM images that particle detection is too sensitive to different noises, uneven backgrounds and over lapped particles, the transform functions, would be a robust quantifying technique.

Grishin et al. [Grishin et al., 2012] used Hough transform in the TEM images for automated identification and measurement of primary particle size distribution in individual aggregates. However, in their study, the accurate results can only be acquired when prior information on the size range of the primary particles in each aggregate is known. Their model relies on the assumption of perfectly spherical structures for primary particles, and can only detect primary particles touching the edge of the aggregate.

Here, in order to improve the sensitivity and specificity of the nano particles in TEM images, an image processing algorithm, based on coupling the operations in special domain with the modified Circular Hough Transform (CHT), has been developed. The proposed method, consequently measures accurately the size distribution of the objective particles in the nanoparticles. This method can be used as an alternative approach for automatic measurement of the

particle diameters and particle size distribution in TEM images. Examples of its application are demonstrated in several TEM images with different chemical and imaging conditions.

2. Proposed Method

Generally the nano particles have a roughly circular shape. CHT is a powerful tool for detecting circular object in the transform domain which is highly reliable [Yao and Yi, 2016]. Hence, an efficient CHT-based algorithm, is proposed for recognition of the circular shapes from the TEM images. In general the CHT, considers the border of the objective particles as the feature points, which are transformed into 3D space, while based on the votes in the transform space, the circles in the image, are considered (discussed in detail in section 2.3). Also the CHT provides parameters to reduce the search time for finding objects based on a set of edge points [Jain and Jain, 2012]. In the first step of the CHT, the border of the objective particles, as the feature points or the foreground pixels of the image, should be recognized. Hence detecting the foreground pixels, representing the border of the subjective particles, plays an important role in CHT. This step has been improved by the following procedures.

2.1. Pre-Processing

To capture the structural features of the nano particles as the foreground objects, several modifications have been implemented, which increase the accuracy of detecting and standing out the objects. These modifications include the techniques for denoising, edge sharpening and edge detecting, as explained in the following.

2.1.1. Image Denoising

Quite often a microscopy TEM image gets corrupted by some random fluctuations which generally arise in the process of acquiring the image and lighting conditions, or during its transmission, or even during reproduction of the image [Kushwaha et al., 2012]. These noises of microscopy imaging, which are generally presented as impulse noises, corrupts the TEM images [Shanmugavadivu and Jeevaraj, 2014]. So the linear filters are not able to effectively eliminate impulse noise, as they have more tendency to blur the edges of an image. On the other hand, nonlinear filters are suited. Hence, in order to attenuate the image noise, a modified median filter, based on directional median operation, was applied to the image. In this procedure, every pixel of the image is substituted by the median of its neighbor pixels based on the following algorithm:

1. Consider each pixel in the image
2. Select the neighboring pixels in all eight directions with a domain of two-pixel
3. Sort the selected pixels in order based upon their intensities
4. Replace the original value of the pixel with the median value

Figure 1a presents an example of TEM image (gold nanoparticles) where the above algorithm is implemented. In order to see the effect of the filtering clearly, a part of TEM image is zoomed as shown in figure 1b. This process works well in preserving the image details, while lets any eventual pixel outliers, i.e., the pixels which diverge

considerably from the surrounding ones, discards. However, after a median filtering, a slight decrease in the image sharpness occurs. Hence, after a median filtering, a sharpening filter improves the quality of the image.

2.1.2. Edge Sharpening

Edge sharpness is very important in the visual quality and the boundaries of the nano particles in TEM image. Most commonly adopted methods increase the contrast of the edges to achieve sharpening [Leu, 2000]. To improve the visibility of such images, histogram equalization and high pass Fourier filter, have been used. This technique modifies the dynamic range and the contrast of an image by altering the intensity histogram to a desired shape. While using Fourier high-pass filter, enhances the edge of the objective particles with retaining the high frequency information within the image, and reducing the low frequency information. This filter that enhances the edge contrast of the image, is an attempt to improve its acutance (apparent sharpness). In other words, this technique, coupling the histogram equalization and high pass filtering, identifies sharp edge boundaries in the image with a desired intensity. Figure 1c illustrates the processed image of the last step (after directional median filter), while Figure 1d is the result after edge sharpening; as is obvious, after edge sharpening, the objective particles stand out clearly from the background. This is the most difficult for small particles, which inherently have less contrast.

2.2. Foreground Detection

The next step for capturing the structural features of the nano particles, is the edge detectors. There are several methods for edge detection and extraction, such as Sobel [Mathur et al., 2016], Roberts [Romero-Manchado and Rojas-Sola, 2015] and Canny algorithm [Tang et al., 2016].

However, due to the importance of this procedure, Sobel filter, which is based on looking-up the maxima of the first derivative, is supported to 2nd-derivative filter, which is based on looking-up zero crossings of the 2nd-derivative. In such way of coupling two edge filters, the operator avoids misleading by noise which they falsely recognize as edges. This is done by running a 2x2 window over the image; if both positive and negative values, appear in the window, a zero crossing occurs. The regions where detected by both Sobel and “Mexican hat” filters are considered as the edge. This procedure is shown in figure 1e. The final step in the edge detection, is the use of hysteresis operator, in which, pixels are marked as either edge, non-edge and in-between; this procedure is based on threshold values. Here, for the pixels that are in-between, if they are connected to the edge pixels, these are marked as the edge pixels as well. The result of this edge detector is a binary image, in which the white pixels closely approximate the true edges of the original image as shown in Figure 1f.

2.3. Hough Space

After detecting the foreground features in the TEM image, the CHT is used to transform the given feature points from the image space into accumulator votes in the parameter space, or the Hough space. In the 2D space, the following equation indicates to a circle located at the center (a, b) with radius r :

$(x-a)^2 + (y-b)^2 = r^2$	Eq. 1
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If (x,y) is considered as a foreground pixel of the image, the parameters (a,b,r) in 3D transform space can be found by Eq.1. Similar to traditional Hough transform, an accumulative matrix, based on votes in the transform space, is constructed and local maximums will be considered as parameters of circles in the image. Nevertheless, if in the real time applications, the TEM image includes a huge number of nano particles in a wide range of the particle size, the large amount of storage, high cost of computation and not-well recognition of all nanoparticles will be the major drawbacks of the CHT. To tackle these problems, beside the above mentioned pre-processing techniques, several modifications to the CHT have been implemented to both increase the accuracy of the measurement and increase the detection rate of the nanoparticles, or more commonly to reduce its computational complexity. For complexity reduction of 3D computation, centers can be calculated in 2D and in this regard, radiuses will be computed in 1D space [Yuen et al., 1990]. In the other words, the circles with the constant radii are considered and the true centers will have the highest votes in the accumulator matrix.

In the next step, to increase the performance of CHT, the edge orientation information has been applied. This method notes that the orientation of an edge point on the boundary of a circle, is in the direction of its center [Atherton and Kerbyson, 1999]. This modification reduces computational requirements by plotting arcs in the accumulator space. This task can be subdivided into following procedures; first, the process is accomplished on the thresholded edge map. In the second step, in order to enhance the hot spots, the resulting (a,b) -space is convoluted with a Mexican hat filter. The (a,b) -space is then thresholded, and centers of the hot spots found. In the fourth step, we accumulate in r -space in order to find the most observable circle with the given center. Then setting a suitable threshold allows for concentric and overlapped circles. Finally the radius of each given circular objective is estimated. It's detailed as follows:

2.3.1. Accumulator Matrix Calculation

In this step, the CHT is used to transform feature points of the image space into the accumulated votes of the parameter space, or the Hough space. These pixels are used according to Eq. 1 for voting to (a,b) parameters by considering constant radius. These votes are stored in accumulator matrix. This procedure is repeated for all radius ranges while accumulator matrix is updated. The CHT accumulation matrix in a surface plot, is shown in figure 2b. The peaks, which are very clear, correspond to the locations of the centers of the circles.

2.3.2. Centers Detection

The array elements that contain the highest number of votes are used to indicate the presence of the pattern, and accordingly the local maximums of accumulator matrix are considered as circle's centers, as shown in figure 2c. The scaled magnitudes of accumulator matrix peaks for each circle are illustrated in figure 2d.

2.3.3. Radii Estimation

Using centers obtained in the previous section, transform space reduces to 1D radius space. The corresponding radiuses are computed based on radial histogram around each center [Grishin et al., 2012; Jain and Jain, 2012].

2.4. Hierarchical Order and Image Inpainting

The TEM images may contain small and large particles simultaneously, while CHT is sensitive to the range of the diameter. If the size of the particles is significantly different, CHT causes a false circle detection, and if the threshold is reduced to avoid the false circles, some true particles will be eliminated. Hence, when the diameter range is rather large, the accuracy of CHT may reduce. To address this issue, a tricky thing that shall be regulated well, is the r-space thresholding. This parameter need to be adjusted in such way that can find objects in all ranges as well the concentric or partially overlapped objects correctly. Hence we implement CHT in a hierarchical order; the diameter range of the particles is divided into some smaller ranges and in each sub-divided range, CHT is applied. In the first step, CHT is implemented on the sub-division of the largest particles. After detecting the largest circles, they are completely filled with using the background pixels around them through image inpainting techniques. Image inpainting is almost a new and important topic in image processing which is the technique of reconstruction a damaged image in an undetectable form. While for our application, this technique, reconstructs the detected circles by the pixels from outside of the given circles; the proposed inpainting algorithm operates based on joining the interpolation of gray levels on the radiant/isophotes (line of equal gray values) directions, while iteratively propagates information from outside of the detected circling along isophotes in the selected region for inpainting. In this paper, partial differential equation (PDE) based inpainting presented in [Bertalmío et al., 2006] is used. With the assessment of this technique, which applies after each iteration, the detected circles are filled and considered as the background for the next iterations, consequently the sensitivity parameter can be increased with iteration to detect the rest particles. This procedure is repeated for smaller ranges until the CHT is applied to the sub-division of the smallest particle. The hierarchical order technique helps to increase the precision of the detection by increasing the sensitivity parameter in the higher order. In the other words, the detection performance remains robust with respect to the parameters while it could be quite sensitive without using hierarchical order. Figure 3a shows this procedure on a sample of TEM image, while figure 3b illustrates a superimposed image of detected circles by CHT and the original image. In this figure, CHT has been applied four times; the detected particles in the first step are illustrated in blue and the results of the second to fourth steps are shown in red, green and yellow respectively. This allows for a quick visual inspection to test the accuracy of the analysis. As seen from the above image, the circle detector of modified CHT works quite well, even for the objects that are partially occluded by others. Finding such objects is possible with the above modified transform of CHT, if enough of the boundary is visible.

The whole procedure of the algorithm is illustrated in the graphical flowchart of figure 4; this graphical flowchart shows that for a precise foreground-detection, a pre-processing stage, including image denoising and sharpening, is performed, as mentioned above. The accumulator matrix is constructed by considering a fixed radius in each step. As can be seen in this figure, the radius increases from minimum value until it reaches to the maximum allowable range. After accumulating votes for all radius range, the center of circles are detected. The next stage is radius estimation of each center, which is performed with using foreground neighborhood histogram. The proposed algorithm uses CHT in the hierarchical order. In each iteration the detected circles are wiped out with image inpainting. The PDE-based image inpainting [Bertalmio et al., 2006] is utilized in the proposed method. In the next iteration the sensitivity parameter is increased to detect the rest circles. As this figure illustrates, the hierarchical procedure is repeated until the maximum iteration (the stopping criterion) is reached and after that the detected centers and radii are reported. According to wide range of experiments, the maximum iteration 4 is an appropriate choice which is used in the all experiments of this paper.

2.5. Statistical Analysis

After obtaining the detected circles, corresponding to the objective particles of the nano-particle, the statistical analysis can be performed to characterize and express quantitatively the statistical information in TEM image. Generally, statistical analysis makes sense of, and draws some inferences, from the given TEM images. The measurement of individual nanoparticle with spherical particles is straight forward. However, when nanoparticles have an irregular shape, their particle size cannot be directly defined, hence accurate size statistics can be more complicated to obtain.

According to the CHT, the approximation for non-spherical particles is to determine the cross-sectional area of the particles and either convert the area to an equivalent spherical diameter, or split to two spherical particles that convert the majority of the region of the particle. In such cases, the indirect definition "sphere-equivalent diameter" is used, where the particle diameter is approximated by a circle which has the most matching with the region of the object. Under this approximation, when Hough transform characterizes a certain particle as a circle, its diameter is defined based on the most similarity of the particle to the circle. The performance of the proposed algorithm could be affected if the particle shape is far from circular or pseudo circular forms. Since a large number of nanoparticles are needed for the high-quality statistical analysis, the work is facilitated when the analysis through the above outlined algorithm is used. The most common descriptive statistical indicators to present the detailed characterization of dispersion of the nano-particle is the histogram, which graphically summarizes the proper distribution model for the data.

3. Experiments

TEM analyses of samples were performed at Shahid Bahonar University of Kerman on a Philips CM-12 TEM with a 70 μm lens operating at 100 kV and with a 2.0 Å point-to-point resolution. The nano suspension solution containing

nanoparticles was analyzed with a VinCary 50 UV-vis Spectrophotometer for detecting the nanoparticles. Transmission Electron Microscope (TEM) images of the nanoparticles were prepared by CEM 902A (Zico Germany) with accelerating voltage of 100 KV. X-ray diffraction (XRD) patterns of the powders were obtained using Philips analytical diffractometer. A Scanning Electron Microscope (CAMSCAN, 2600 MV) is used to energy dispersive X-ray spectroscopy (EDS) analysis of the samples. For more details of the experimental setup and instruments, please see [Hashemipour et al., 2011]. Sample preparation, stage positioning, image shifting and focusing were done manually by the operator, based on the decisions the automation software produced.

4. Results

For testing the our method, nine TEM images, in different chemical and imaging conditions, were passed through the above outlined algorithm to be analyzed, then the results were compared with the manual measurement. Among the tested TEM images, three samples, which have the most complexity in measurement, have been provided in detailed. The first TEM image, shown in figure 5a, is a nano cluster of the silica supported Ni catalyst (5% wt of Ni).

The processed image of the given TEM image, which is a superimposing of detected circles on the original image, is shown in Figure 5b. This figure shows, although the given TEM image contains some veins and uneven background due to Ni catalyst, the proposal CHT could detect almost all particles. Figure 6 illustrates the size distribution of the nanoparticles given by proposal CHT.

These 106 particles, detected by our algorithm, yield an average diameter of 4.67 nm, while these values for manual measurement is respectively 102 and 4.7 nm. As the detected particles at the border of the image, were truncated by the image acquisition system, they were not included in these results. The next sample of TEM images, presented in figure 7a, is related to the Al_2O_3 nanoparticles in Diethanolamine; this image includes overlapped and concentrated nano particles, which is even difficult to separate and count the nanoparticles by eye.

The proposed method has been applied to the given image and the output is illustrated in figure 7b. This allows for a quick visual inspection for the accuracy of the detection. As seen from the above image, although a few complicated particles are not recognized correctly, the circle detector of modified CHT works quite well, even for the nano particles that are concentrated or partially occluded by others.

The size distribution of the nanoparticles of the given TEM image is illustrated in figure 8, which is obtained by the proposal CHT. For this TEM image, the detailed characterization of the dispersion of the nanoparticles is difficult even for manual measurement. However the above outlined CHT could separate and recognize 54 particles, and

according to our approximation the average diameter is 15.6 nm. These values for the manual measurement is 56 and 15.9 nm.

Figure 9a, shown the third sample of TEM image, which is related to copper nanoparticles synthesized at concentration ratio of 4 (the ratio of reducing agent (NaBH_4) to precursor (CuSO_4)). This TEM image provides view of non-circular shapes of nanoparticles with un-uniform contrast and dark background, while the objective particles do not stand out.

The result of the processed image is presented in figure 9b. The resulting image by proposal CHT, shows a good recognition for the position of the nano particles. The statistical results of the size distribution of the nanoparticles are presented in figure 10. However the above outlined CHT could recognize 27 particles with the average diameter of 30.2 nm, while these values for manual measurement is 25 and 31.4 respectively. This difference between the measurements is due to the inherent operation of the CHT; if the shape of a non-circular object is close to two connected circular objects, then the algorithm approximates the non-circular particles by two circular objects which has the most matching with the region of the object. The subjected particles are obvious in image 9b.

The above mentioned TEM images are ones with different complexity of the nano cluster and imaging conditions, which shows the capability of the proposed algorithm for different conditions. Table 1 summarizes the statistics of nanoparticle populations of 6 other samples of TEM images of the silica supported Ni catalyst with different Nickel loading.

Beside the above results, in order to prove the validity and reliability of the proposed method, a TEM image, with overlapped particles is taken from available literature. Therefore, the obtained results and driven conclusions can be considered statistically meaningful for a more reliable comparison. Hence a TEM image from the research of rastogi et al. [Rastogi et al., 2012] has been taken; this TEM image is related to gold nanoparticles (Au NP) with using sodium borohydride as reducing agent and bovine serum albumin as capping agent (figure 11).

In their study, the distribution of the nanoparticles was obtained with Philips CM200 (New York, USA) transmission electron microscope (TEM). This typical TEM image has been analyzed by our algorithm; the detected nanoparticles are marked in the figure 11b; as is obvious, our algorithm is highly accurate to detect the whole range of the nanoparticles, as well the overlapped ones.

The particle size distribution of the given TEM image is illustrated by histogram in figure 12, which compares the particle size distribution obtained by our method and the results of [Rastogi et al., 2012]; here (fig. 12), the red bars of histogram present the hydrodynamic diameter, obtained from a Malvern Zetasizer Nanosystem, while the blue bars show the measurement assessed with the modified CHT. This figure shows a good agreement between two

measurements, as well proves the suitability and reliability of the proposed method. According to the measurement of our method, the given TEM image (fig. 11) has 102 particles with an average diameter of 8.4 nm, while the average diameter, reported in [Rastogi et al., 2012] is 8.6 ± 2.8 nm and number of nanoparticles has not been reported in their article. Evidently, the proposed method offers high accuracy of the recognition, and consequently precise measurement of particle dimensions with consistent results.

5. Conclusion

The measurement of particle size distributions from TEM images is often difficult, especially when noises, uneven or overlapped nano particles in TEM image are presented. Here, this problem was addressed by using a novel image processing method, based on modified CHT, for automatic and robust recognition and counting the nano particles from the TEM images.

A nonlinear filtering algorithm has been developed by directional median filter coupled with contrast adjustment and high pass filtering to overcome the peak intensity attenuation and edge sharpening operation. Afterwards the modifications in an accumulative matrix of transformation space, edge orientation information and inpainting operation, results reduction in complexity and improvement in accuracy for detecting circular and semi-circular particles. The visual inspections on the analyzed images, showed a high accuracy on the results, especially for the TEM image including circular nanoparticles as well as the regions of the image with overlapped particles or noisy and uneven backgrounds. Using this approach, TEM images have been directly analyzed and compared with the data obtained from the manual measurement, which shown a small error of less than 5% in calculating the average diameter and particle counting. Based on the findings of this study, the merits of the method is the ability to analysis accurately the nanoparticles with a very high number of particles, which makes it suitable to process large stacks of TEM images. The proposed image processing method could also calculate the average, shortest and longest dimensions.

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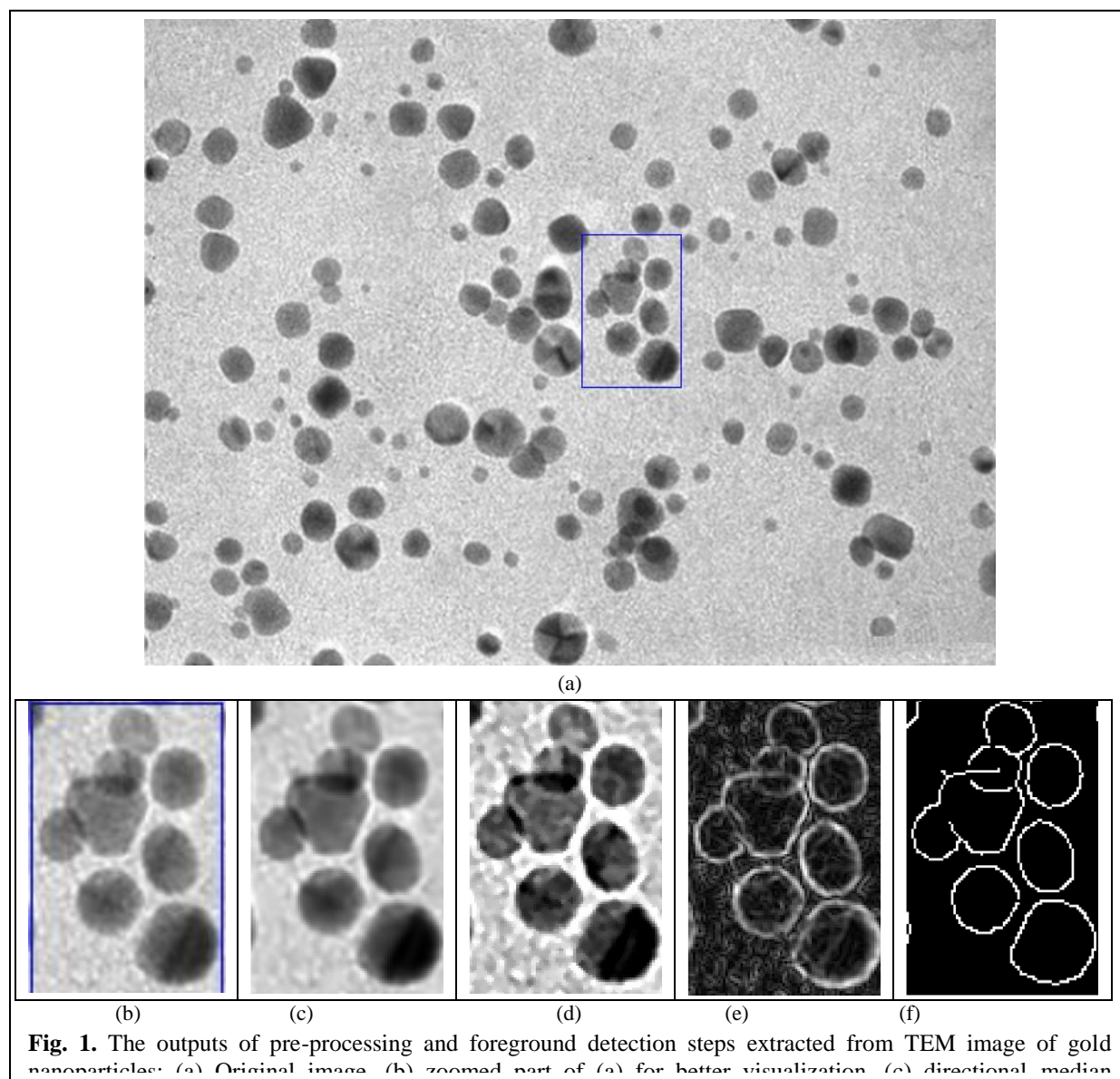
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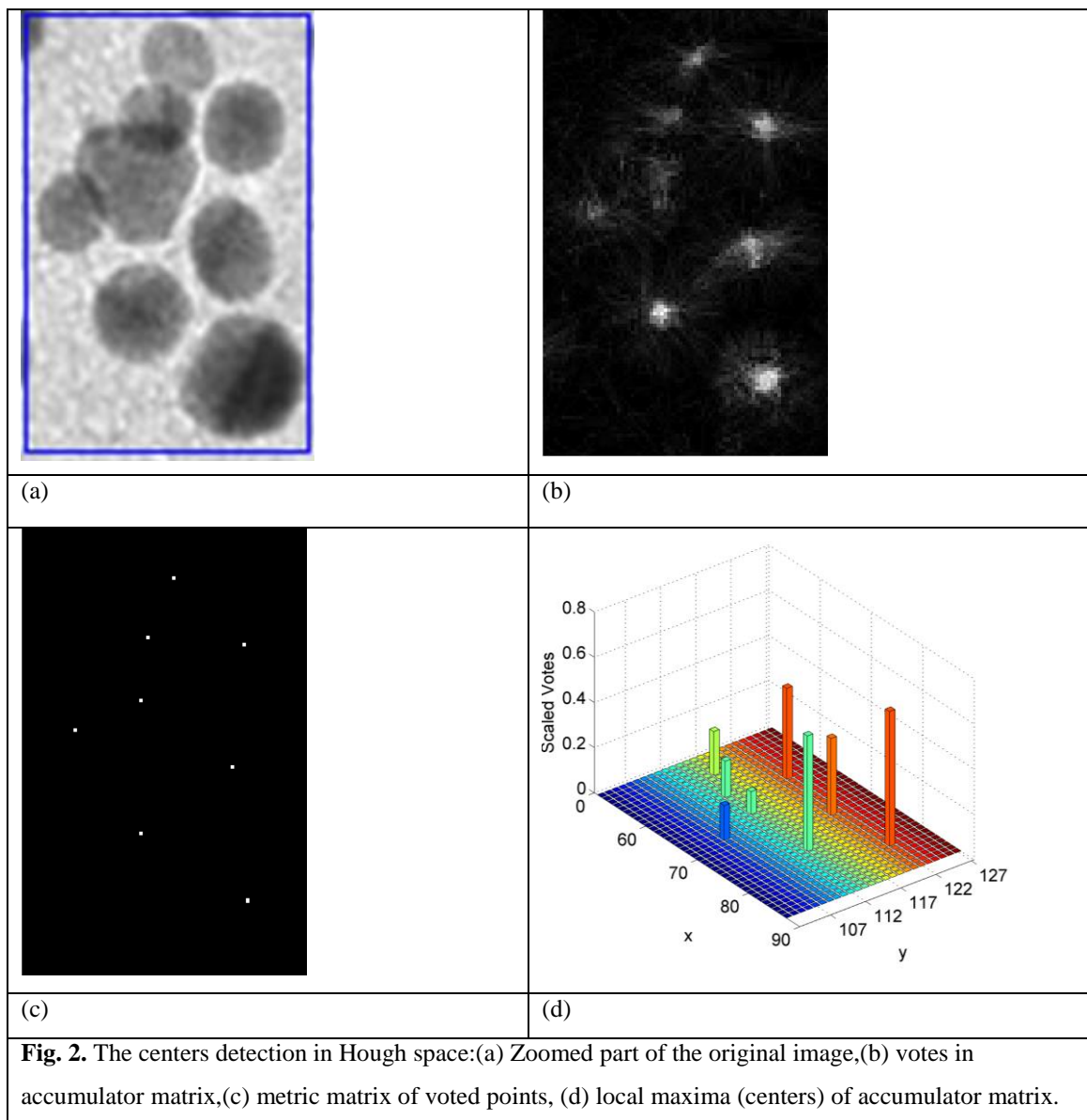
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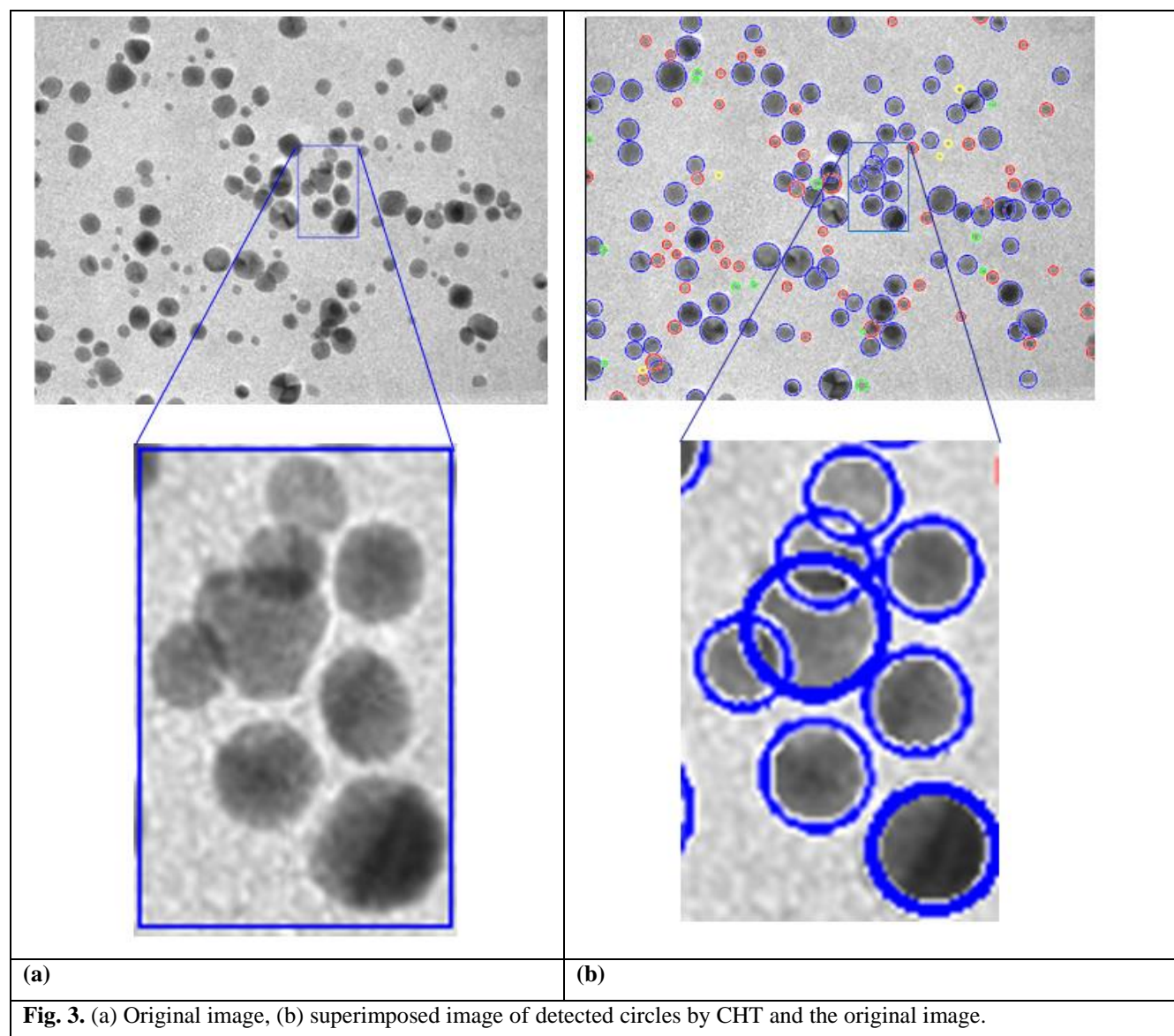
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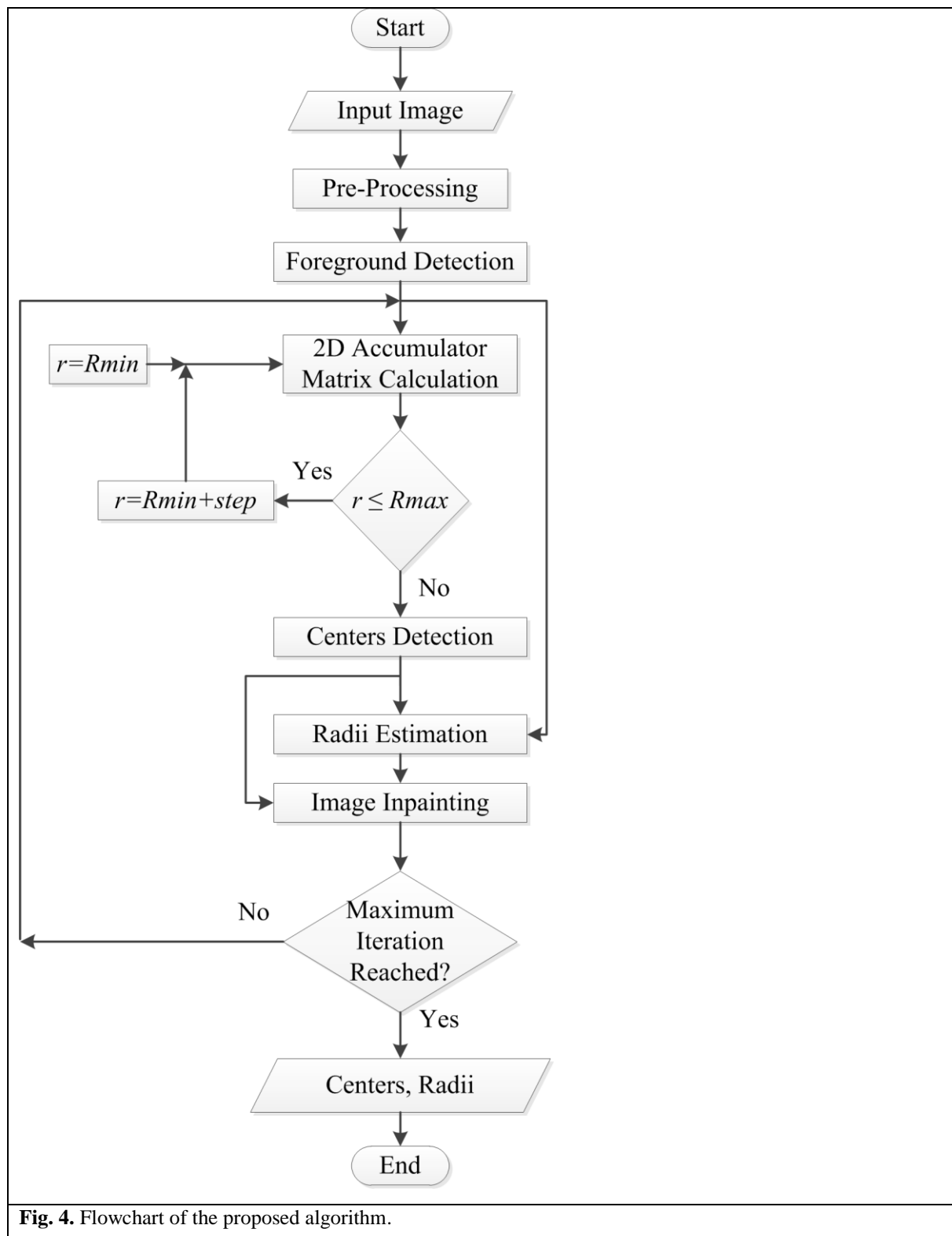
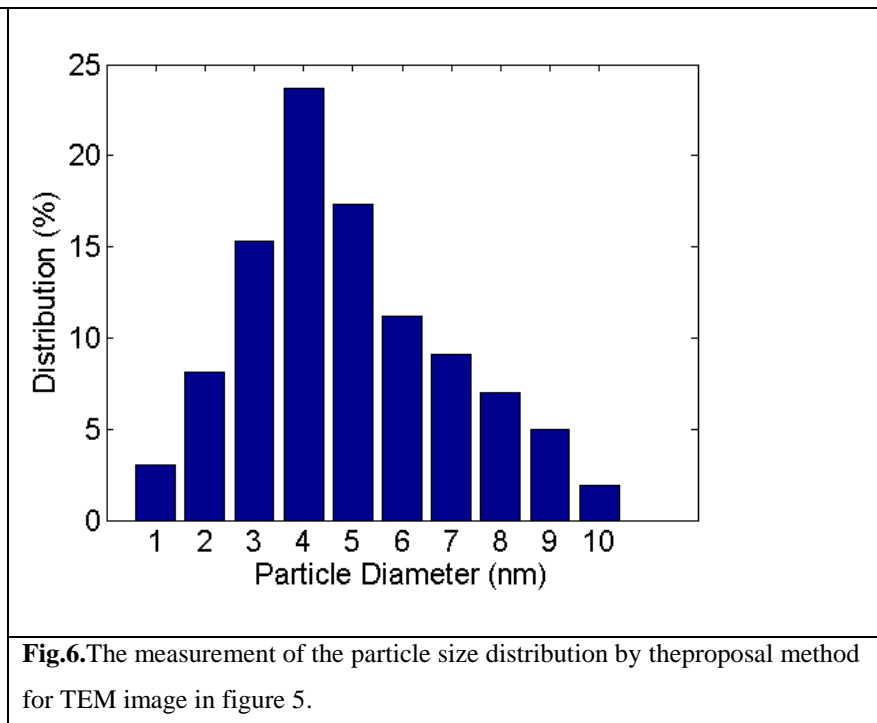
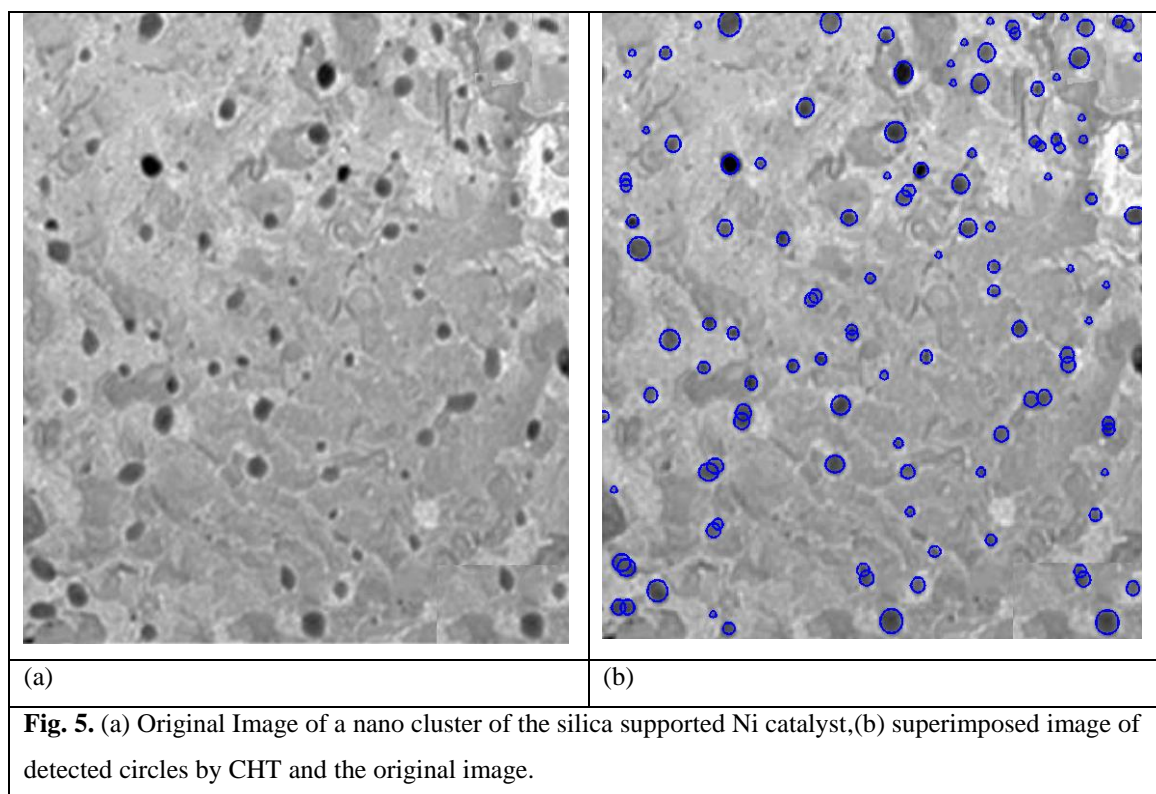
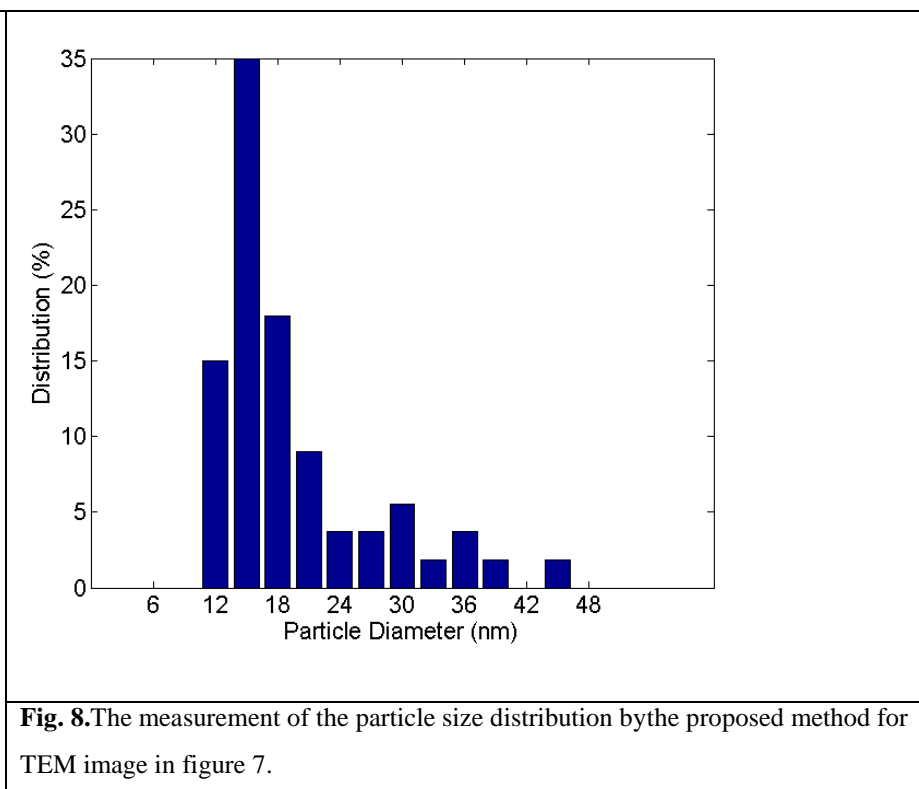
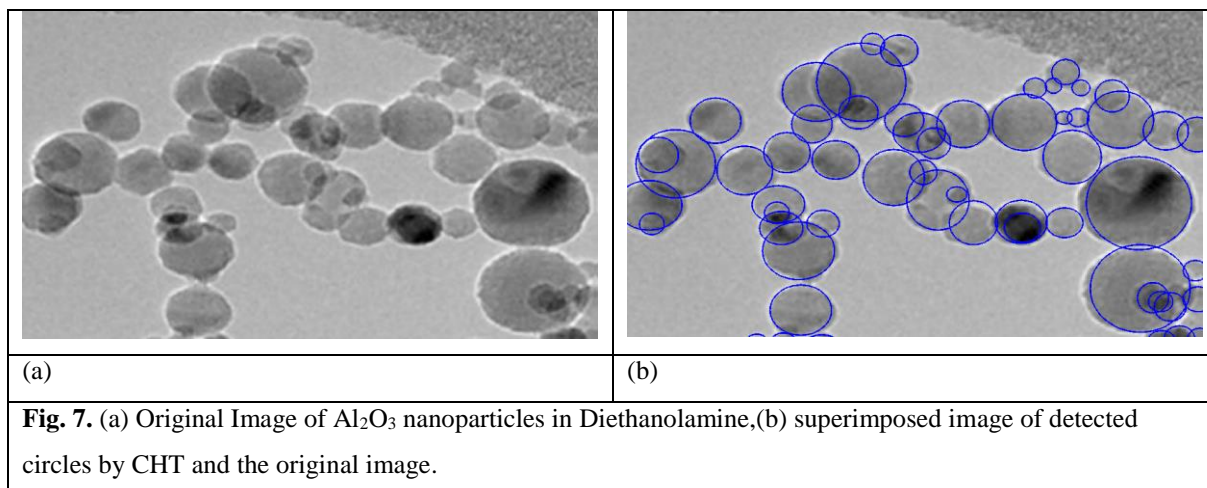
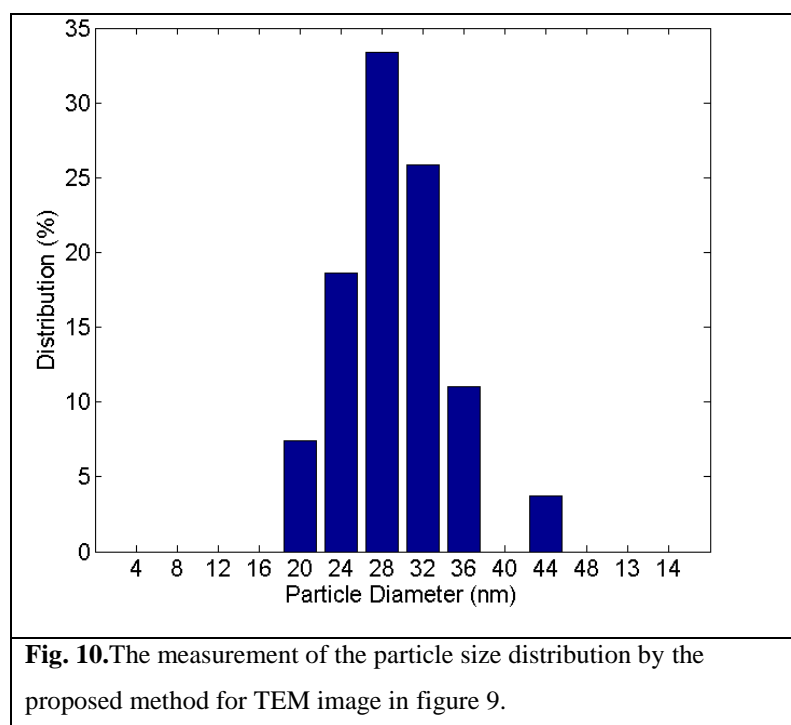
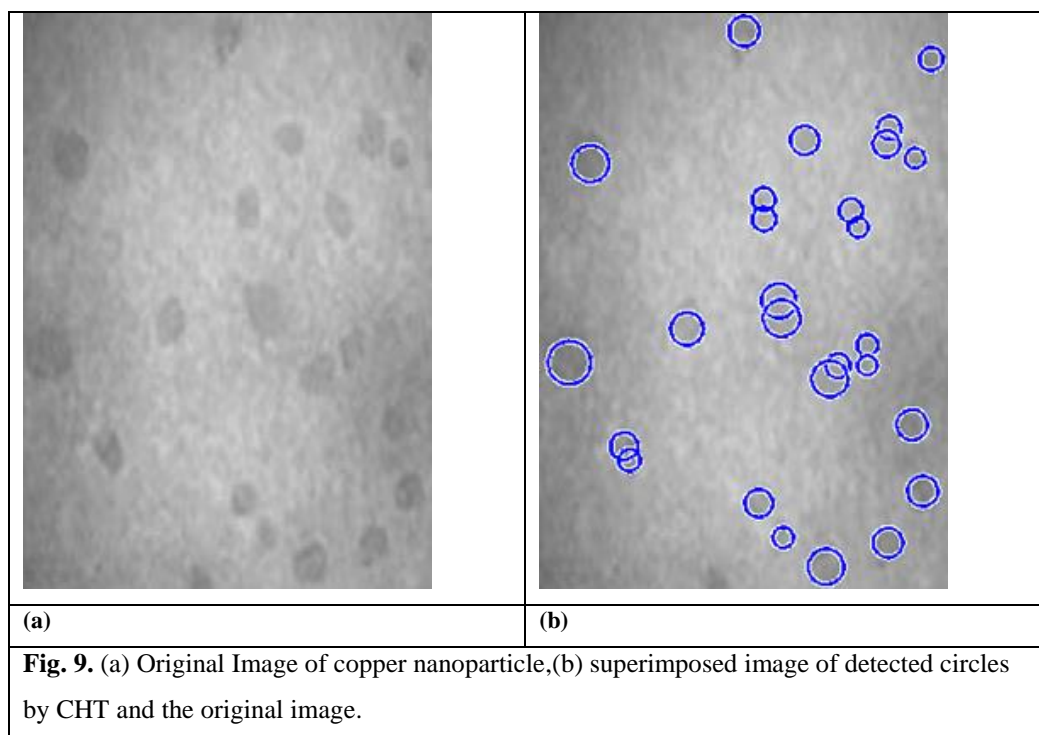


Fig. 4. Flowchart of the proposed algorithm.







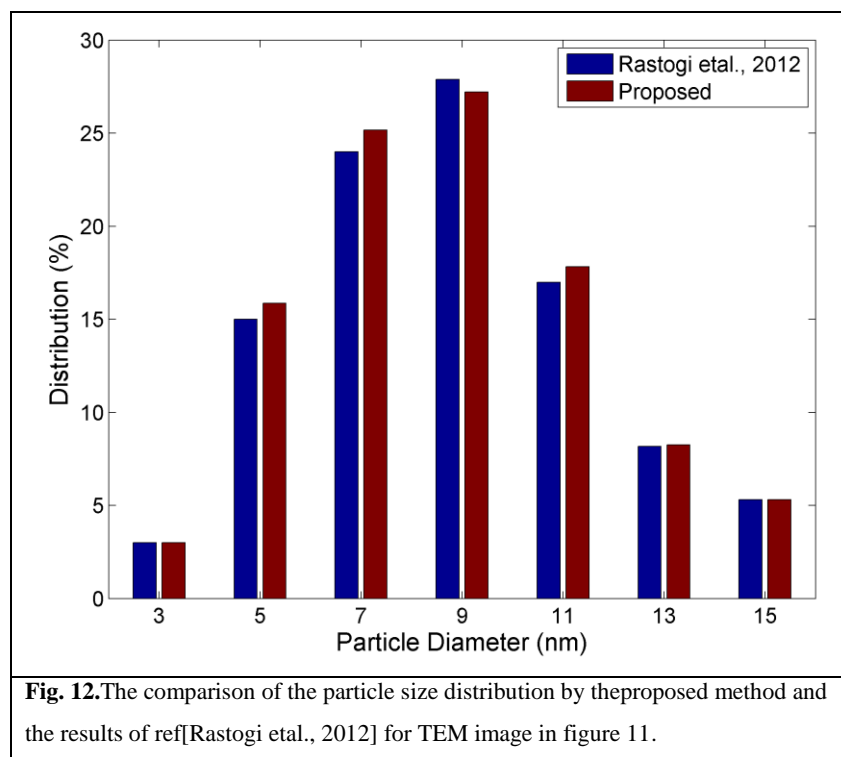
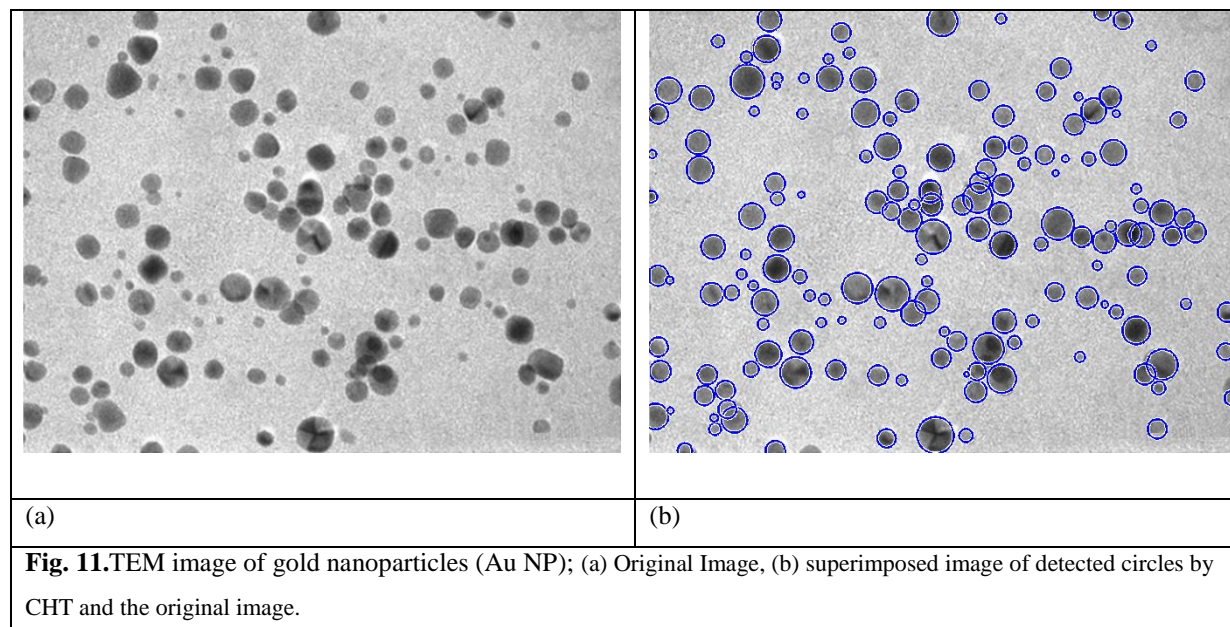


Table 1. Mean diameter and the number of nanoparticles obtained from the one-by-one inspection and image processing method for TEM images of the silica supported Ni catalyst with different Nickel loading

images	Nickel loading	Number of particles		Mean of diameter(nm)	
	% wt	Our method	manual	Our method	manual
Sample 1	2	94	91	3.4	3.4
Sample 2	10	85	79	7.3	7.4
Sample 3	15	71	67	10.1	10.4
Sample 4	20	54	51	15.9	16.4
Sample 5	25	39	37	18.7	19.1
Sample 6	30	26	25	26.4	27.5