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An Improved Machine Learning Algorithm for Silver Nanoparticle Images: A Study on Computational Nano-Materials

Parashuram Bannigidad[1], Namita Potraj[1]*, P M Gurubasavaraj[2]

1 Dept. of Computer Science, Rani Channamma University, Belagavi, 591156, Karnataka, India
2 Dept. of Chemistry, Rani Channamma University, Belagavi, 591156, Karnataka, India

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* **Corresponding author**.

namitapotraj@gmail.com

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Abstract

Objectives: To study the determination and classification of Scanning Electron Microscopy (SEM) images of silver nanoparticles using digital image processing techniques and classifying the images using various machine learning classifiers namely; SVM, K-NN, and PNN classifiers. **Methods:** Segmentation techniques namely; Fuzzy C-Means (FCM) and K-Means were applied to extract geometric features from SEM images of silver nanoparticles. The size of nanoparticles was determined and classified based on various nano applications. The categorization of silver nanoparticles was done based on the geometrical feature value, i.e. area, and was further classified into three categories such as; 0-100 nm, 101-200 nm, and 201-500 nm using both the segmentation techniques. Further, the proposed method extracts texture features such as; kurtosis, skewness, and entropy and used SVM, K-NN, and PNN classifiers to classify the silver nanoparticle SEM images. The proposed results are analyzed and interpreted by chemical experts to observe the effectiveness of the proposed method. **Findings:** To improve the classification accuracy of Silver nanoparticle images, textural features such as; kurtosis, skewness, and entropy were extracted and SVM, K-NN, and PNN classifiers are used to classify the silver nanoparticle images. The performance of the proposed algorithm is evaluated based on performance evaluation measures such as; specificity, sensitivity, true predictive value, and false positive value. Higher TPV, specificity, and sensitivity values show promising results. It is observed that the SVM classifier has 75.00%**,** K-NN has 62.50%, and PNN yielded an accuracy of 93.00%. **Novelty:** Traditional synthesis and characterization techniques are inefficient and time-consuming and require more space and infrastructure to analyze the size of the nanoparticle. The current study circumvents these limitations. The silver nanoparticle images used in the present study are synthesized using rudanti fruit seeds, and the process is named as green synthesis process or biological method. This method is widely used because it does not use anytoxic chemicals, is inexpensive, and is environmentally friendly. In the previous work, only geometrical features namely; Area (pore size), and range-wise pore size were calculated depending on the application-oriented. The present study employs two phases; the training phase (knowledge base) and the testing phase(classification). The training phase includes segmentation, feature extraction, and categorization depending on the applications. The testing phase extracts the textural features such as; kurtosis, skewness, and entropy, and classifies the silver nanoparticles by using SVM, K-NN, and PNN classifiers.

Keywords: Silver nanoparticles; Image segmentation; FCM; KMeans; SVM; KNN; PNN

1 Introduction

Nanotechnology offers the possibility to change the properties of a material by controlling the size of the material, which has driven research into a wide range of potential applications for nanomaterials. Nanotechnology is a revolutionary path for technological development that focuses on material management at the nanometer scale (one billion times smaller than a meter) [1]. It is used in a broad scale of scientific fields (medicine, biotechnology, ecology, pharmacy, electronics, and others). Nanomaterials have a wide range of definitions used by different scientific committees, national authorities, and international organizations. International Standard Organization (ISO) defines a nanomaterial as "a material having any external dimension in the nanoscale or having an internal structure or surface structure in the nanoscale". Nanomaterials are increasingly becoming a part of our daily lives. They are used in pioneering products and processes. Further, Nanomaterials are classified into various categories, namely; nanoparticles (NP), nanofibre, nanoplate, etc. Nanoparticles are tiny objects with all external dimensions at the nanoscale. The scope of research is also expected shortly to enable materials with properties previously unknown. In the past decade, the biosphere has seen visible growth in the application of nanoscience and nanotechnology, leading to great advances in the development of new nanomaterials. Nanoparticles synthesized from nanometer-sized metals, either destructively or constructively, and to change the properties of their respective metals are metal oxide-based nanoparticles.

Silver is classified as a metal-based nanoparticle. The synthesis of nanoparticles is a significant process that involves the synthesis using biological and chemical methods. silver metal oxide nanoparticles have been successfully used in biomedical diagnostics and therapeutics for many years [2]. Nanosilver has found numerous applications in medicine [3], wherein it was exploited as an antibacterial, antifungal, antiviral, and anti-inflammatory agent. Because of their excellent intrinsic properties and wide range of applications, nanoscale analogues of gold, silver, platinum, and palladium in particular have gained prominence [4]. The discovery of recipes that yield nanomaterials with defined optical properties is costly and time-consuming. The synthesis process is a vital method that decides the even-sized formation of nanoparticles. Over the decades, there have been many changes in the synthesis process and applications of silver nanoparticles. The recent progress of silver nanoparticles in terms of synthesis and electronic applications [5], shows the evolution in the synthesis process which is application-oriented. Silver nanoparticles represent a new research direction involving a new class of materials with promising applications in biological, biomedical, and pharmaceutical domains, and it has demonstrated significant potential and implications in the treatment of bacterial infections [6]. Silver nanoparticles (AgNPs) are widely used as antimicrobial agents in personal care products [7]. A design method is proposed by Kaur A et.al. [8], in which a glycopeptide antibiotic called vancomycin is loaded on antibacterial citrate-capped silver nanoparticles to increase its activity against bacteria. Roelofs Dicki et.al.[9] investigated how surface coating and particle size are the primary factors explaining the earthworm's transcriptome-wide responses to silver nanoparticles. Combining silver nanoparticles with small amounts of antibiotics increases their antimicrobial efficiency, yielding excellent results *in vitro* and promising *in vivo* bacterial eradication.

The SEM images A, B, C, D, E, F, and G are obtained from the synthesization followed by characterization methods as shown in Figure 1.

1.1 Literature survey

Only a few researchers have made significant contributions to the field of nanoparticle image analysis. The following are some examples of related work in the field of nanoparticle image analysis using various segmentation techniques.

The present study focuses on the segmentation of Silver SEM nanoparticle image analysis and classification. The silver SEM nanoparticles were synthesized using a green synthesis process which used rudanti fruit seeds. Earlier work on a similar dataset was carried out wherein the silver SEM nanoparticle images were synthesized using rudanti fruit extract.[10]. Parashuram Bannigidad et.al[11] made an effort to automate a tool that can determine the size of TEM images of Iron Oxide Nanoparticles using the Gaussian Mixture Model - Expectancy Maximization (GMM-EM) segmentation technique to extract various geometrical features from the nanoparticles. Mekki-Berradaey et. al.[12] showed a two-step framework for a machine learning-driven high-throughput microfluidic platform that can produce silver nanoparticles with the desired absorbance spectrum in a short period. The combination of nanotechnology and computer science is currently involved in a wide range of theoretical and practical requests[13]. These applications can help shape the future of computer science by making smaller and more efficient computer parts, improving data transmission, and facilitating device communication. Simultaneously, computer applications can improve experimental research in nanotechnology by simulating the behavior of nanoparticles at a lower cost and in less time. Digital image processing aids different kinds of nanomaterials to find their size and structures economically. Lehan Yao et.al.[14] explained the method to achieve nanoparticle segmentation in liquid-phase TEM videos using a U-Net-based workflow to map the interaction landscape, anisotropic etching, and self-assembly kinetic laws. Eryka et. al.[15] used digital image processing techniques such as principal component analysis (PCA), multiple linear regression (MLR), and second-order regression on SEM images of silver nanoparticles for the comparative analysis of the presence of chemical oxygen demand in wastewater. Elif Emil Kaya et. al.[16] proposed a search algorithm based on an image-processing technique to obtain particle size and particle-size distribution from SEM images of silver nanoparticles. Parashuram Bannigidad et. al.[17] automated a system that extracted the pore from the experimental Al_2O_3SEM images and extracted the geometrical features; nanopore wall thickness, pore size, and porosity using five different segmentation techniques: global thresholding, active contour, K-means, region growing, and watershed.

Madallah et.al.[18] used weighted spatial Fuzzy C-Means clustering techniques to find the spatial information of brain MRI images. Amanda[19] developed a new algorithm named iterative label spreading to identify a pattern in high-dimensional feature spaces of silver nanoparticle images and compared it with K-Means, ward agglomerative, and DBSCAN clustering methods. Parashuram et. al.[20] estimated the size and structure of Boron nanoparticles using Fuzzy C-Means and K-Means segmentation techniques.

2 Methodology

For experimental purposes, silver nanoparticles were obtained using silver nitrate, a green synthetic process with varying pH by keeping the concentration (mM), time (min), and temperature (^{0}C) constant. The details of chemical compositions used for the preparation of silver nanoparticles during green synthesis are given in Table 1 .

Table 1. The details of chemical compositions for the preparation ofsilver nanoparticle

Image	Concentration (mM)	Time (min)	Temperature(^{0} C)	pH
A	1	30	90	3
B	1	30	90	5.3
C	1	30	90	6
D	1	30	90	10
E	1	30	90	7
F	1	30	90	7.5
G	1	30	90	11

The purpose of this study aims to develop an algorithm to analyze the silver nanoparticle SEM images with the Green synthesization process. The proposed study also provides an automated tool to determine the size of silver nanoparticles in SEM images. Figure 2 shows a flow diagram that outlines the procedure of the proposed work for segmentation using FCM and K-Means clustering methods.

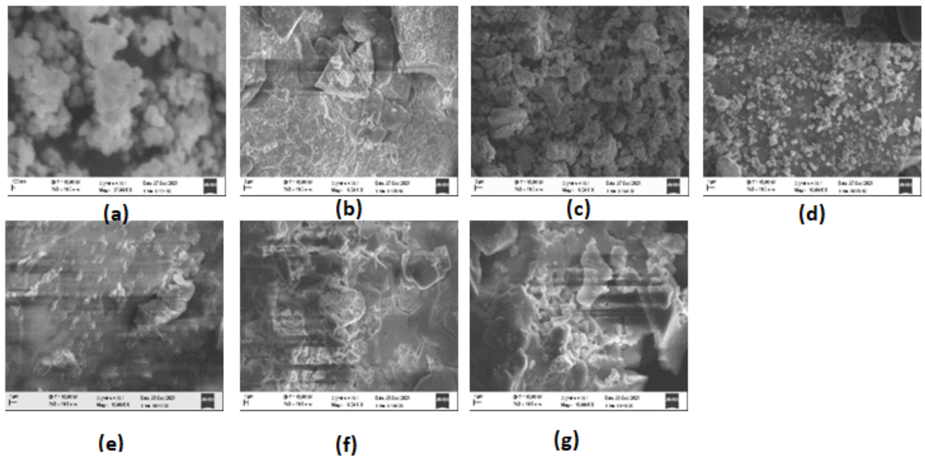

Fig 1. SEM images of silver nanoparticles synthesized at varying pH

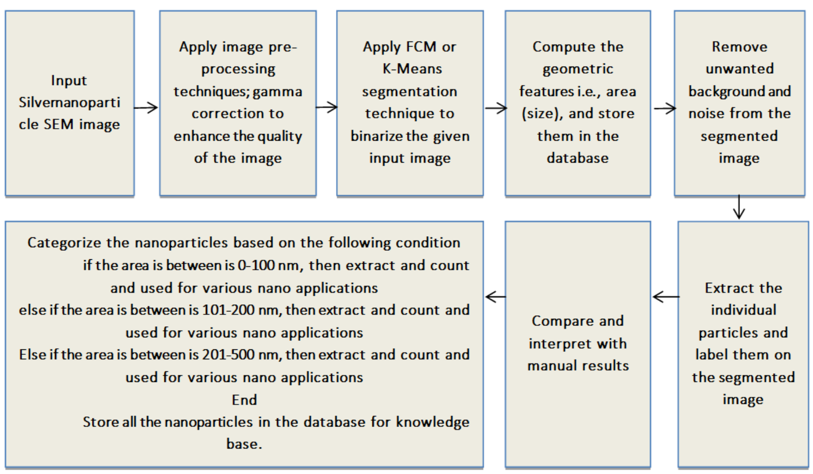

Fig 2. The flow diagram of the proposed study uses FCM and K-Means clustering methods

2.1 Categorization phase

Algorithm I: Segmentation, feature extraction, and categorization of silver nanoparticle s.

Step 1: Input the SEM images of silver nanoparticles
Step 2: Apply image pre-processing techniques
Step 3: Perform segmentation by using clustering methods namely; Fuzzy C-Means and K-Means clustering techniques.
Step 4: Compute the geometric feature; i.e. area for all nanoparticles and store them in the database.
Step 5: Remove unwanted background noise from the segmented image in step 4.
Step 6: Identification of individual nanoparticles and labeling them on the segmented image
Step 7: Compare and interpret the manual results
Step 8: Categorize the nanoparticles under the following conditions

if the area is between 0-100 nm, then extract and count suitable for various nano applications
else if the area is between 101-200 nm, then extract and count suitable for various nano applications
else if the area is between 201-500 nm, then extract and count suitable for various nano applications
end
store all the silver nanoparticles in the database for the knowledge base.

2.2 Classification phase

Algorithm II: Preprocessing, feature extraction, and classification of silver nanoparticle s.

Step 1: Input the SEM images of silver nanoparticles
Step 2: Apply image pre-processing techniques: applying filters, enhancing the images
Step 3: Perform texture-based feature extraction on SEM images of silver nanoparticles
Step 4: Perform classification using SVM, K-NN, and PNN classifiers
Step 5: Predict the images whether it is silver or non-silver nanoparticle using different classifiers.

The proposed method employs various classifiers namely; SVM, K-NN, and PNN applied to silver nanoparticleSEM images to predict the positive and negative classification. Figure 3 shows the flow diagram of feature extraction and classification of SEM images of silver nanoparticles.

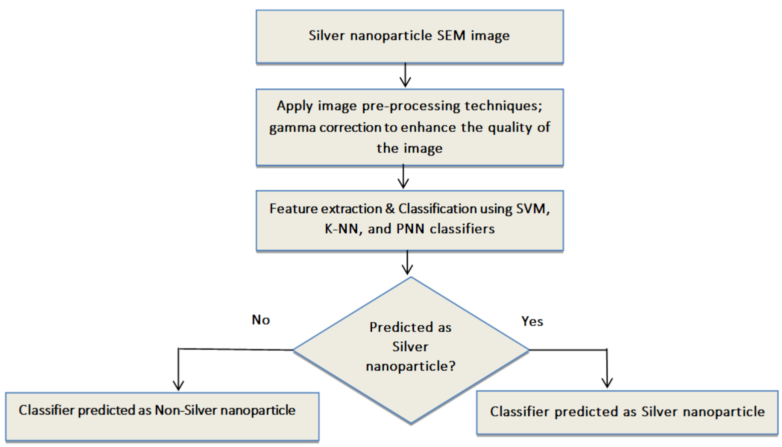

Fig 3. The flow diagram of feature extraction and classification of SEM images of Silver nanoparticles

A. Preprocessing

The key objective of the preprocessing of silver nanoparticle SEM images is to resize and apply filters to remove the unwanted background and debris and enhance the images. The input images are resized to 483 X 717 to remove the bottom line then applied gamma correction to the resized images to remove the noises and boost or increase the tone of the images.

B. Segmentation

The segmentation is a crucial part of the algorithm since the good extraction of nanoparticles depends on the appropriate segmentation process. The fuzzy C-Means (FCM) segmentation technique is applied which acts as a pre-process to support the classification and recognition process and is used as the extraction process of silver nanoparticles of SEM images. To check the robustness of the algorithm we have used another segmentation technique named K-Means The geometrical feature; i.e. the area(size) of each nanoparticle is extracted using both clustering methods namely; Fuzzy C-Means and K-means. The Fuzzy classification uses fuzzy value partitioning to assign a membership function for each group to the sample. Affiliation value plays an important role in the clustering process and performs classification. It is a more flexible and robust process for handling noise and uncertain data. Similarly, K-Means is another clustering algorithm used for separating the area of interest from the background.

C. Feature extraction

This algorithm explores the geometrical feature area(size) of each nanoparticle of the silver nanoparticle SEM image. The area of a nanoparticle can be evaluated as follows;

Area= Total number of pixels in an extracted nanoparticle.

D. Classification

The proposed work includes classification algorithms that explore the texture features for classification. The various texture features extracted in the proposed algorithm are; kurtosis, skewness, and entropy of a grayscale image. The performance evaluation of the proposed algorithm is measured by statistical features such as; sensitivity, specificity, true predictive value, and false positive value.

3 Results and Discussion

For experimentation, a total of 336 nanoparticles from various SEM images of silver Nitrate samples (A, B, C, D, E, F, and G) are studied which are obtained from the synthesization and characterization process. The geometrical features of each image are extracted using MATLAB R2018a software on Intel(R) Core^{TM} i5-10210U CPU@1.60GHz system. The experimentation is carried out by applying segmentation techniques namely; Fuzzy C-Means and K-Means. The SEM images of silver nanoparticles A, B, C, D, E, F, and G (Figure 4 (i)) are resized. To extract the feature value of each nanoparticle, the images are segmented using the clustering technique Fuzzy C- Means (Figure 4 (ii)). Followed by removing unwanted background noise from the segmented images. The extraction of individual nanoparticles and labeling them on the segmented image (Figure 4 (iii)) is performed. To check the robustness of the algorithm, we have also used another segmentation method, i.e., K-Means clustering to extract the nanoparticles from SEM images of silver nanoparticles. The input images are resized (Figure 5 (i)), followed by applying the K-Means Clustering technique (Figure 5 (ii)), then the removal of unwanted background from the images, and finally extraction of individual nanoparticles and labeling them (Figure 5 (iii)).

The geometrical feature value; the area is calculated for each labeled nanoparticle using Fuzzy C-Means and K-Means clustering algorithms. The nanoparticles are categorized into various sizes; 0-100 nm, 101-200nm, and 201-500 nm for all the seven (A, B, C, D, E, F, and G) SEM images of silver nanoparticles which are used by different applications as shown in Table 2.

Table 2. Geometric feature value; area of SEM images of silver nanoparticles A, B, C, D, E, F, and G

Images	**FCM**			**K-Means**		
	Area 0-100 nm	**Area 101-200 nm**	**Area 201-500 nm**	**Area0-100 nm**	**Area 101-200 nm**	**Area 201-500 nm**
A	05	03	02	00	00	04
B	86	02	02	06	00	00
C	73	11	07	80	14	09
D	74	18	10	81	20	11
E	13	02	04	14	02	02
F	21	00	00	23	00	00
G	21	03	02	09	00	01
Total	**293**	**39**	**27**	**213**	**36**	**27**

Based on experimentation, it is observed that the minimum size of the area of the silver nanoparticles is suitable for wide applications such as; medical, food, health care, consumer, industrial, preservatives in cosmetics, and anti-acne preparations of cosmetics. The antibacterial activity of various metal nanoparticles, including silver colloids, is closely related to their size. That is the smaller the silver core, the greater the antibacterial activity. Therefore, Chemists and scientists look at the nanoparticles within the minimum range of size of nanoparticles. Hence the categorization of individual silver nanoparticles is very much essential. As per the categorization of size(area) of each nanoparticle, it is observed that using the Fuzzy C-Means segmentation technique a total of 293 nanoparticles are extracted, and are in the range of 0-100 nm, 39 nanoparticles are in the range of 101-200 nm, and 27 nanoparticles are in the range of 201-500 nm. Similarly using the K-Means segmentation technique 213 nanoparticles are in the range of 0-100 nm, 36 nanoparticles are in the range of 101-200 nm, and 27 nanoparticles are in the range of 201-500 nm. Figure 6 shows the graphical representation of the number of nanoparticles that are in the range of 0-100 nm, 101-200 nm, and 201-500 nm using both Fuzzy C-Means and K-Means segmentation techniques. As per the experimentation, it is observed that the Fuzzy C-Means segmentation technique extracts more (293) nanoparticles when compared with the K-Means segmentation technique (213), which ranges in the standard range of 0-100 nm. Hence, the Fuzzy C-Means segmentation technique is suitable for silver nanoparticles as per the required applications.

The proposed method also employs various classifiers for the classification of silver nanoparticles, namely; SVM, K-NN, and PNN applied to silver nanoparticle SEM images to predict whether the sample image is a silver nanoparticle image or not. Table 3 shows statistical feature values and the average classification accuracy of the proposed method using the SVM classifier, K-NN classifier, and PNN classifier. Figure 7 depicts a comparative analysis in terms of class accuracy of SVM, K-NN, and

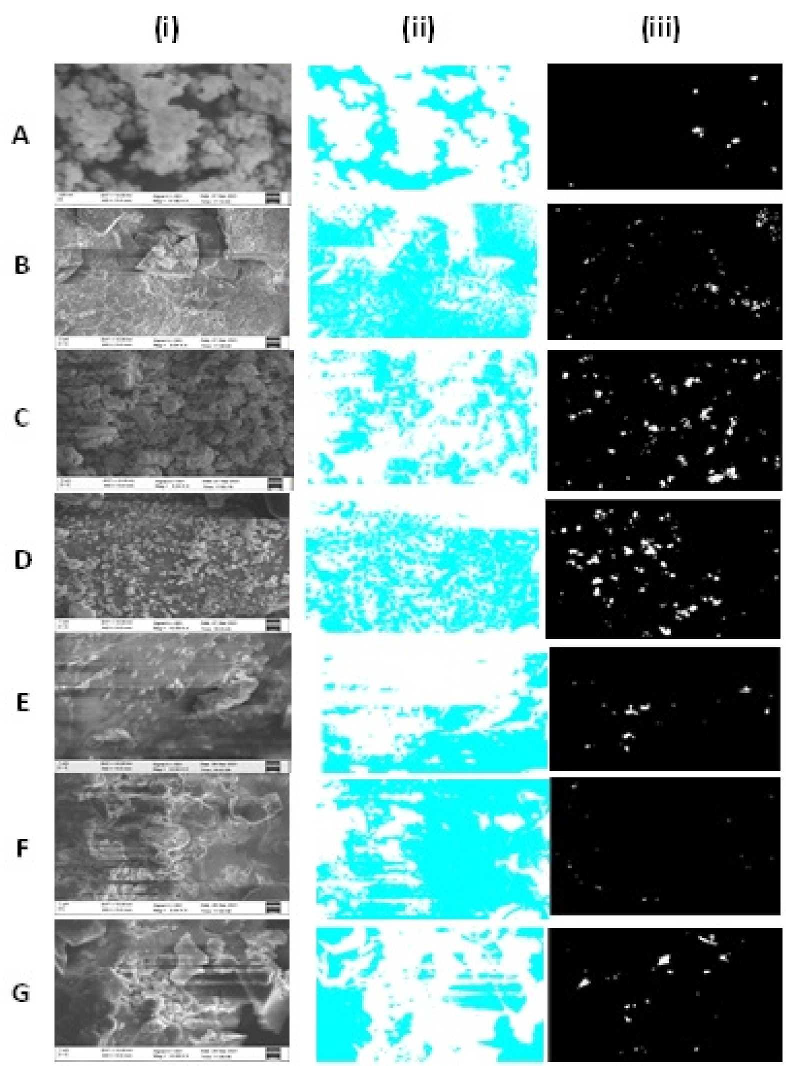

Fig 4. Segmented images using Fuzzy C-Means method (a) Original SEM images of silver (b)Fuzzy C-Means (c) Extracted and labeled nanoparticles

PNN classifiers. The performance of the quality of the nanoparticle images is evaluated based on the following performance evaluation measures

- Sensitivity(St)= Measure the percentage of true positives (TP) Correctly identified from a particular set of input images are shown in equation (1)

$$St = TP/(TP + FN) * 100 \tag{1}$$

- Specificity (Sp)= Measures the percentage of false negative (FN) correctly identified from a particular set of input images shown in equation (2)

$$Sp = TN/(TN + FP) * 100 \tag{2}$$

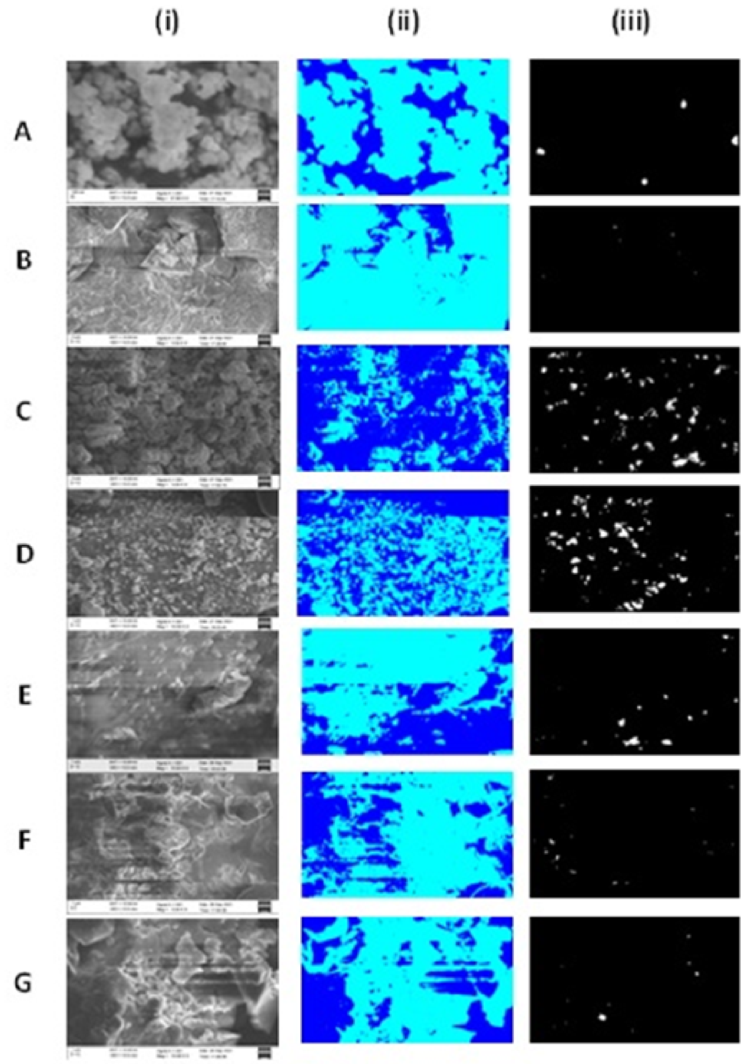

Fig 5. Segmented images using K-Means method (a) Original SEM images of silver (b) K-Means(c) Extracted and labeled nanoparticles

- True predictive value(TPV): It indicates the percentage of the total input set that shows the correct classified images given in equation (3)

$$TPV = (TP/(TP+FP)) * 100 \tag{3}$$

- False positive value(FPV): Indicates the percentage of the total input set that shows the correct misclassified images is given in equation (4)

$$FPV = (FP/(FP+TN)) * 100 \tag{4}$$

Table 3. The statistical features values and classification accuracy of the proposed method using SVM, K-NN, and PNN classifiers

Classifiers	Statistical features				Class Accuracy
	St	Sp	TPV	FPV	
SVM	100.00%	50.00%	66.67%	50.00%	75.00%
K-NN	100.00%	25.00%	57.14%	75.00%	62.50%
PNN	100.00%	87.50%	88.89%	12.50%	93.00%

St-Sensitivity, Sp- Specificity, TPV-True positive value, FPV-False positive value

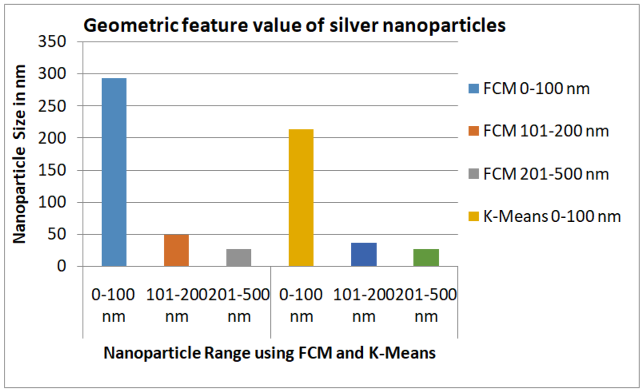

Fig 6. Graphical representation of the number of nanoparticles that are in the range of 0-100 nm, 101-200 nm, and 201-500 nm using Fuzzy C-Means and K-Means segmentation techniques

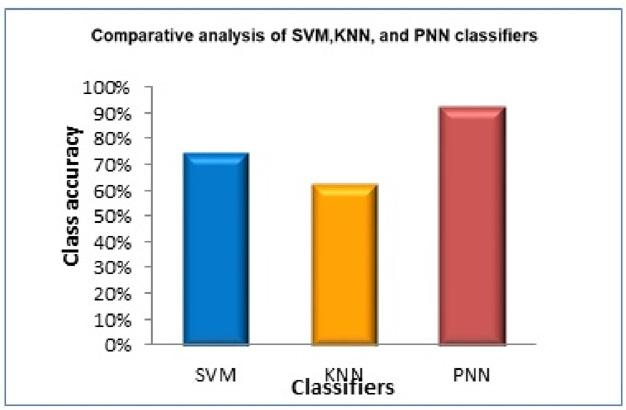

Fig 7. The bar chart depicts a comparative analysis of SVM, K-NN, and PNN classifiers

4 Conclusion

The current study focuses on the segmentation and classification of silver SEM nanoparticle images. silver SEM nanoparticles were created using rudanti fruit seeds known as the green synthesis process. Previously, work on a similar dataset was completed, but there is a minor difference between the current and previous work's synthesization processes. The previous work used rudanti fruit extract to synthesize silver SEM nanoparticle images, whereas the current work uses rudanti fruit seeds to synthesize silver SEM nanoparticle images. Only the geometrical feature Area (pore size) and range-wise pore size were calculated in previous work, which was application-oriented. The current study employs two algorithms: algorithm I for silver nanoparticle segmentation, feature extraction, and categorization, and algorithm II for silver nanoparticle preprocessing feature extraction, and classification. and algorithm II for silver nanoparticle preprocessing, feature extraction, and classification. This will help scientists and researchers use silver SEM nanoparticles in a variety of applications.The objective of this present study is the categorization and segmentation of SEM images of silver nanoparticles which is based on the standard size of nanoparticles centered on the applications such as medical, food, health care, consumer, industrial, preservatives in cosmetics, and anti-acne preparations of cosmetics. The segmentation techniques namely; Fuzzy C-Means and K-Means are applied to SEM images of silver nanoparticles and 336 nanoparticles are obtained which are further classified in the range of 0-100 nm, 101-200 nm, and 201-500 nm. It is observed that the Fuzzy C-Means segmentation technique yields more nanoparticles (293) that are in the standard range of 0-100 nm than the K-Means segmentation technique (213). Hence as per the interpretation and analysis with chemical experts, it is observed that the Fuzzy C-Means segmentation technique is suitable for the segmentation and categorization of SEM images of silver nanoparticles. The proposed method also extracts the performance measuring approaches and uses SVM, K-NN, and PNN classifiers to classify SEM images of silver nanoparticles and predict the positive and negative classifications. The performance of the proposed algorithm is evaluated based on the performance rating scales such as kurtosis, skewness, and entropy. A higher true predictive value (88.89%) using the PNN classifier shows promising results. It is observed that the SVM classifier provides 75.00% class accuracy, the K-NN provides 62.50%, and the PNN provides 93.00% class accuracy.

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