1. Algorithm Description

1.1 The proposed algorithm for morphology/shape recognition and core center detection

**Algorithm 1: Morphology/Shape Recognition**

```plaintext
Input: histogram-adjusted SEM image X
Output: set of detected core centers \( P = \{ p_1, \ldots, p_n \} \), binary image \( X_b \), shape type \( M \)

1. \( X_b^1 \leftarrow \text{medianBlur(binarize}(X)) \); \( X_b^2 \leftarrow \text{medianBlur(binarize}(X, \text{invert}=1)) \)
2. \( X_b \leftarrow X_b^1 \); \( X_b^2 \leftarrow X_b^2 \)
3. for \( i \leq \text{maxErosionCount} \) do
   4. \( X_b^1 \leftarrow \text{erode}(X_b^1, \text{kernelSize}=3) \); \( X_b^2 \leftarrow \text{erode}(X_b^2, \text{kernelSize}=3) \)
5. \( S^1 = \{ s_{1}^1, \ldots, s_n^1 \} \leftarrow \text{segmentation}(X_b^1) \); \( S^2 \leftarrow \text{segmentation}(X_b^2) \)
6. initialize set of core segments, \( C^1 \leftarrow \emptyset \); \( C^2 \leftarrow \emptyset \)
7. for \( s_{1,i}^1 \) in \( S^1 \) do
   8. if \( \text{isBackground}(s_{1,i}^1) \) or \( \text{isBorder}(s_{1,i}^1) \) or \( \text{numAdjacency}(s_{1,i}^1) > 1 \) then
    9. Continue
10. \( r_s^1 \leftarrow \text{area}(s_{1,i}^1) \); \( r_c^1 \leftarrow \text{area(convexHull}(s_{1,i}^1)) \)
11. if \( (r_s^1 / r_c^1) > \text{solidity_thres} \) then
    12. \( C^1 \leftarrow C^1 \cup s_{1,i}^1 \)
13. repeat 7 for \( S^2 \)
14. if \( |C^1| \geq |C^2| \) then
    15. \( X_b \leftarrow X_b^1 \); \( S \leftarrow S^1 \); \( C \leftarrow C^1 \)
16. else \( X_b \leftarrow X_b^2 \); \( S \leftarrow S^2 \); \( C \leftarrow C^2 \)
17. initialize set of core centers, \( P \leftarrow \emptyset \)
18. for \( c_i \) in \( C \) do
    19. \( P \leftarrow P \cup \text{centroid}(c_i) \)
20. initialize set of shell segments, \( C_a \leftarrow \emptyset \)
21. for \( c_i \) in \( C \) do
    22. \( C_a \leftarrow \text{adjacentSegment}(c_i, S) \)
    23. if \( \text{centroid(validContour}(c_i)) \) in \( c_i \) then
        \( C_a \leftarrow C_a \cup c_i \)
    24. if \( |C_a| / |C| < \text{valid_ratio} \) then
        \( M \leftarrow \text{core-only} \)
25. else \( M \leftarrow \text{core-shell} \)
```
1.2 The proposed algorithm for core-shell size measurement

**Algorithm 2: Core-shell Size Measurement**

**Input:** histogram-adjusted SEM image $X$ and its binary image $X_B$

**Output:** core sizes $F$, shell sizes $G$

1. Initialize core and shell sizes, $F \leftarrow \emptyset$, $G \leftarrow \emptyset$
2. Initialize core and shell segments, $C \leftarrow \emptyset$, $S \leftarrow \emptyset$
3. Initialize core centers, $P \leftarrow \emptyset$
4. $Z \leftarrow \text{imageStatistics}(X)$
5. $L = \{l_0, \ldots, l_n\} \leftarrow \text{prune}($segmentation$(X_B), Z)$
6. For $l_i$ in $S$ do
   - If not isBackground($l_i$) and not isBorder($l_i$) and numAdjacency($l_i$) $== 1$ then
     - $C \leftarrow C \cup l_i$
   - $C \leftarrow \text{filterOutliers}(C)$
7. For $c_i$ in $C$ do
   - $p \leftarrow \text{centroid}(c_i)$
   - $P \leftarrow P \cup p$
   - $F \leftarrow F \cup \text{measureSize}(p, c_i)$
   - $S \leftarrow S \cup \text{adjacentSegment}(S, c_i)$
8. Initialize input and marker images for watershed, $X_B' \leftarrow X_B$, $X_M \leftarrow \text{ones}$
9. $n \leftarrow 2$
10. For $c_i$, $s_i$ in $C$, $S$ do
    - For $(x, y)$ in $c_i$ do
      - $X_M(x, y) \leftarrow n++$
      - $X_B'(x, y) \leftarrow 0$
    - For $(x, y)$ in $s_i$ do
      - $X_M(x, y) \leftarrow 0$
11. $X_M \leftarrow \text{watershed}(X_M, X_B')$
12. For $p_i$ in $P$ do
    - $G \leftarrow G \cup \text{measureSize}(p_i, X_M)$
Figure S1: Examples of successfully measured nanoparticles using the automatic pipeline

- **Core-only**
- **Core-only (Rod)**
- **Core-shell**
- **Core-shell (Rod)**
Figure S2: Examples of somewhat poorly measured nanoparticles using the automatic pipeline, together with improved sizes using the semi-automatic pipeline, respectively.

A, B: failure cases of scale bar text information retrieval. The provided manual scale selection function enabled to measure sizes correctly.

C: Poorly measured core sizes due to incorrect segmentation. D: Improved core measurement using manually chosen parameters.
Figure S3: Examples of incorrectly measured nanoparticles

A: Incorrect morphology classification due to vague core boundary. B: Incorrectly measured sizes due to occlusion