Chapter 4

Results and Analysis

4.1 Introduction

In this work, the effect of different ambient gases and pressures on the powder characteristics and the wire explosion process has been investigated. In order to investigate the ambient gas effect, explosions of copper wires in argon, nitrogen, air and nitrogen-argon mixtures at 500 mbar have been carried out. On the other hand, investigation on the ambient pressure effect has been carried out by exploding copper wires in 1 bar, 500 mbar, 100 mbar and 50 mbar nitrogen and air ambient.

In the following sections, the experiment results for the study of the effect of different ambient gas will first be shown. The results for the study of the effect of different ambient pressure will be given in the section thereafter. For each study, the results and analysis of the electrical signals (current, voltage and light emission signals) will first be discussed. After that, powder properties as characterized by TEM, FESEM and XRD will be shown.

Experimental parameters for the investigation of the ambient gas effect are summarized in Table 4.1 while that for the investigation of the ambient pressure effect are summarized in Table 4.2.

Table 4.1: Experimental parameters used in the investigation of the ambient gas effect.

1	Capacitance	1.85 μF
2	Charging voltage	10 kV
3	Wire material	Copper (Annealed)
4	Wire diameter	125 μm
5	Wire length	6.1 cm
6	Ambient pressure	500 mbar
7	Ambient gas*	(i) 100% argon (ii) 100% nitrogen (iii) 75% nitrogen + 25% argon (iv) 50% nitrogen + 50% argon (v) 25% nitrogen + 75% argon [(iii)–(v): Nitrogen-argon mixtures]
8	Powder collector	(i) Membrane filter (ii) Glass substrate
9	Powder characterization	(i) TEM (ii) XRD

* The percentage of each gas represents the percentage of the partial pressure of the gas.

Table 4.2: Experimental parameters used in the investigation of the ambient pressure effect.

1	Capacitance	1.85 μF
2	Charging voltage	10 kV
3	Wire material	Copper (Annealed)
4	Wire diameter	125 μm
5	Wire length	6.1 cm
6	Ambient pressure	(i) 1 bar (ii) 500 mbar (iii) 100 mbar (iv) 50 mbar
7	Ambient gas	(i) Air (ii) Nitrogen
8	Powder collector	(i) Membrane filter (ii) Glass substrate and silicon substrate
9	Powder characterization	(i) TEM (ii) FESEM (iii) XRD

4.2 Investigation of the ambient gas effect

4.2.1 Current, voltage and light emission signals

From the experiment, it has been observed that the current, voltage and light emission signals have almost similar waveforms when the wires are exploded in 500 mbar nitrogen and nitrogen-argon mixtures. The typical waveforms are shown in Figure 4.1. This mode of explosion is known as wire explosion with current pause because there is a period where the current flowing across the circuit is near to zero. On the other hand, wire explosion in 500 mbar argon ambient produced signals as that shown in Figure 4.2. In this case, no current pause has been observed while arc discharge is observed to occur at an earlier time. This mode of explosion is known as wire explosion with immediate discharge. These two types of waveforms as mentioned above have been discussed by Cash (Cash, 1999) and Kotov (Kotov, 2003).

In this work, it has been observed that the current pause formed after the first spike has average temporal length in the range of 7.0 – 7.5 μs for the case of 100% nitrogen, 75% nitrogen + 25% argon and 50% nitrogen + 50% argon. However, the length is approximately 1.2 μs for majority of the wire explosions in the ambient of 25% nitrogen + 75% argon, although several explosions with longer current pause (~ 7 μs) have also been observed.

From the current and voltage signals, the deposited energy has been calculated based on the method described in section 3.6.1. The average deposited energies at different stages of wire explosion under different condition are summarized in Table 4.3.

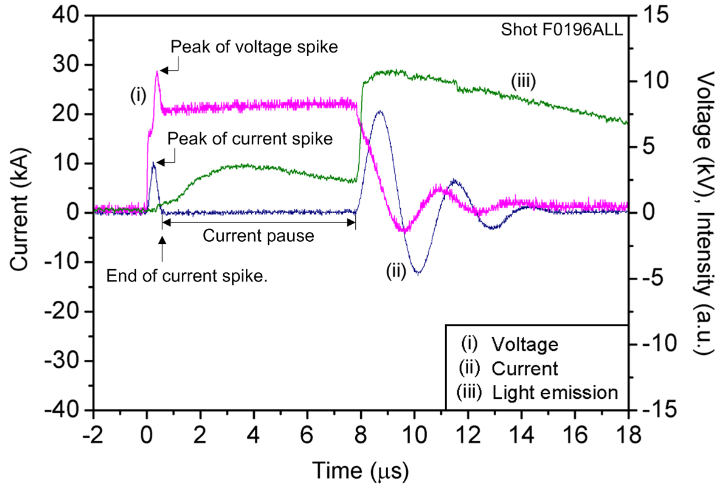


Figure 4.1: Typical current, voltage and light emission signals obtained from wire explosion in 500 mbar nitrogen and nitrogen-argon mixtures.

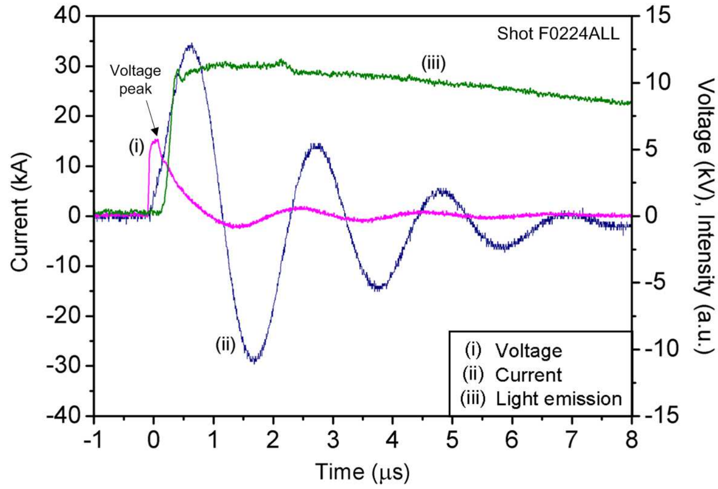


Figure 4.2: Typical current, voltage and light emission signals obtained from wire explosion in 500 mbar argon.

Table 4.3: Average deposited energies at different stage of wire explosion in different ambient gases.

Ambient gas composition	Average deposited energy (J)		
	Peak of current spike	**Peak of voltage spike / Voltage peak**	**End of current spike / Current dip**
100% N_2	4.78 ± 0.38	18.00 ± 0.83	24.10 ± 2.10
75% N_2 + 25% Ar	3.45 ± 1.23	16.59 ± 0.95	21.44 ± 1.03
50% N_2 + 50% Ar	3.22 ± 0.98	15.08 ± 3.40	20.55 ± 2.84
25% N_2 + 75% Ar	2.23 ± 0.21	16.43 ± 1.23	20.46 ± 0.72
100% Ar	-	1.02 ± 0.32	-

Since no current spike has been observed on current signal obtained from wire explosion in 100% argon ambient, only the average deposited energy at the time of the voltage peak is given in Table 4.3. The voltage peak represents the time when the discharge occurs. After the discharge has formed, the current flowing through the wire material will not be distinguishable from that flowing through the discharge. Besides that, the amount of current flowing through the wire will also be very low compared to that flowing through the discharge. The standard deviation of the deposited energy at different stages is also given in the table. The energies required to bring the solid copper wire at room temperature (25 ^{o}C) to various states are summarized in Table 4.4. The calculation of the energies is shown in Appendix A.

Table 4.4: The energies required to bring a solid copper wire at room temperature to various states.

Melting point reached.	Complete melting of the whole wire.	Boiling point reached.	Complete boiling of the whole wire.
2.7 J	4.1 J	7.9 J	40.1 J

Several observations obtained from the comparison of data in Table 4.3 and 4.4 are summarized below:

(1) Wire explosion in ambient containing nitrogen:

(a) Only in the case of wire explosion in 100% nitrogen ambient that the wire has been fully melted at the time the peak of current spike is reached.

(b) Wire has reached its boiling point at the point where the peak of voltage spike is reached.

(c) Wire has not been fully vaporized before the first current spike ended.

(2) Wire explosion in pure argon ambient:

(a) Wire has not reached the melting temperature at the point when the arc discharge is formed.

In the next two sections, the particle size distribution and the crystal structures of the powders produced in different ambient gases will be presented. Further discussion related to the signals and the powder characteristics will be carried out in chapter 5.

4.2.2 Particle size analysis based on TEM images

From the TEM images, the size of particles obtained has been measured. The measured diameter has been categorized into 20 classes in the range of 1 – 99 nm. The particle size distribution for each condition has been calculated by adopting the log-normal distribution function. From the calculation, the median diameter and the geometrical standard deviation for the nanopowders have been obtained. The median diameter and geometrical standard deviation are summarized in Table 4.5. The change of median diameter with ambient gas composition is illustrated in Figure 4.3.

Table 4.5: Particle size analysis for nanopowders obtained from wire explosion in different ambient gases.

Ambient gas composition	Median diameter (nm)	Geometrical standard deviation (nm)
100% N_2	31.2	1.5
75% N_2 + 25% Ar	33.4	1.4
50% N_2 + 50% Ar	32.7	1.4
25% N_2 + 75% Ar	33.3	1.3
100% Ar	37.5	1.4

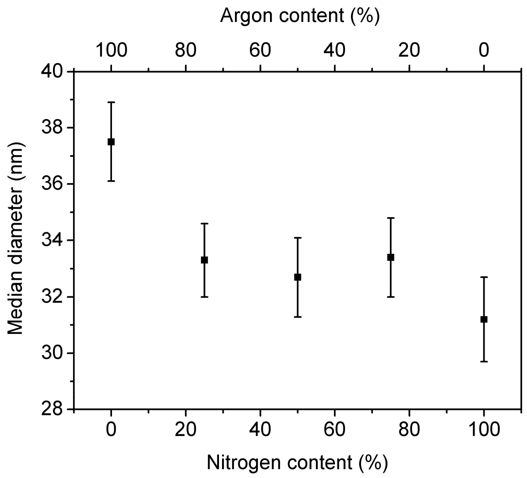


Figure 4.3: Median diameter of nanopowders produced by wire explosion in different ambient gases.

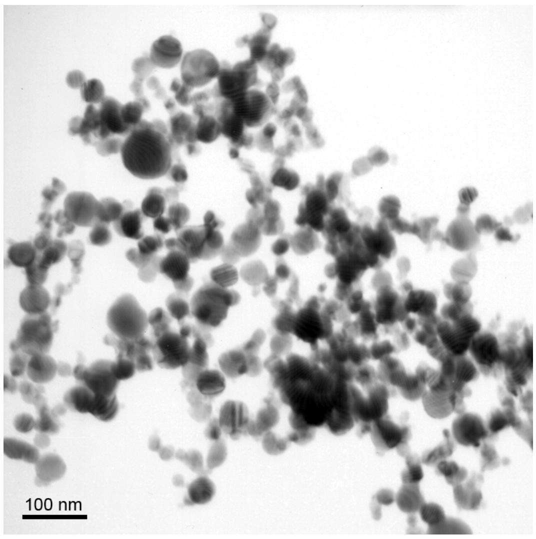


Figure 4.4: Typical TEM image of nanopowders obtained from wire explosion in nitrogen, argon and nitrogen-argon ambient.

4.2.3 Crystal structure of the powders determined from XRD analysis

From the XRD results, it is observed that the powders obtained from wire explosion in the five different ambient gases gave the same XRD pattern. A typical XRD pattern is shown in Figure 4.5. Three peaks have been observed on the XRD pattern. By comparing the value of 2θ for each peak with the database value, they are found to be corresponding to the face-centered cubic copper crystal structure. No peaks corresponding to copper oxide or nitride have been observed.

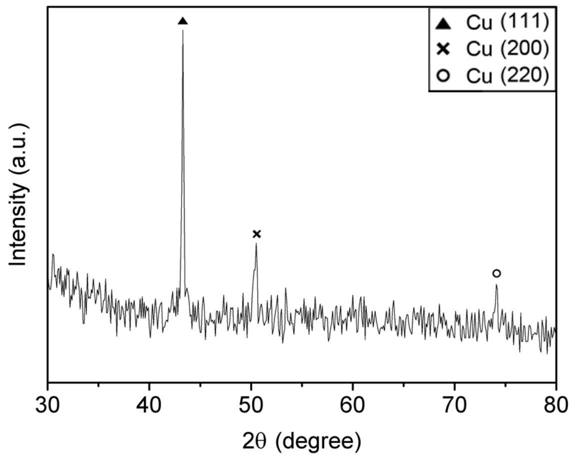


Figure 4.5: Typical XRD pattern for samples obtained from wire explosion in different ambient gases.

Table 4.6: Summary of XRD analysis for samples obtained from wire explosion in different ambient gases.

Gases	Compositions	2θ (hkl)	Lattice
100% N_2 75% N_2 + 25% Ar 50% N_2 + 50% Ar 25% N_2 + 75% Ar 100% Ar	Cu	43.5 ^{o} (111), 50.4 ^{o} (200), 74.0 ^{o} (220).	Face-centered cubic

4.3 Investigation of the ambient pressure effect

4.3.1 Current, voltage and light emission signals

The signals obtained from nitrogen and air wire explosion at the same pressure are generally observed to be the same. When the applied pressure is 1 bar and 500 mbar, wire explosion with current pause has been obtained. It has been observed that the current pause has average duration of about 7.9 μs for the case of air while it ranges between 6.8 – 7.5 μs for the case of nitrogen. As the pressure is further reduced to 100 mbar, no current pause has been observed. In this case, the discharge occurred at an earlier time after the peak of the first current spike but before the current spike ended. This has formed a dip in the current signal as shown in Figure 4.6(c). At an ambient pressure of 50 mbar, the formation of discharge occurred at even earlier time. The signals have general features similar to that for wire explosion in 500 mbar argon as shown in Figure 4.2.

Wire explosion in the ambient of 100 mbar is categorized as wire explosion with current dip while that in the ambient of 50 mbar is categorized as wire explosion with immediate discharge. The main difference between the two wire explosions is the time of occurrence of the discharge where the discharge occurs at earlier time in the case of 50 mbar compared to that of 100 mbar. From the current and voltage signals, the deposited energy has been calculated based on the method described in section 3.6.1. Average deposited energies at different stages of wire explosion in different condition are summarized in Table 4.7.

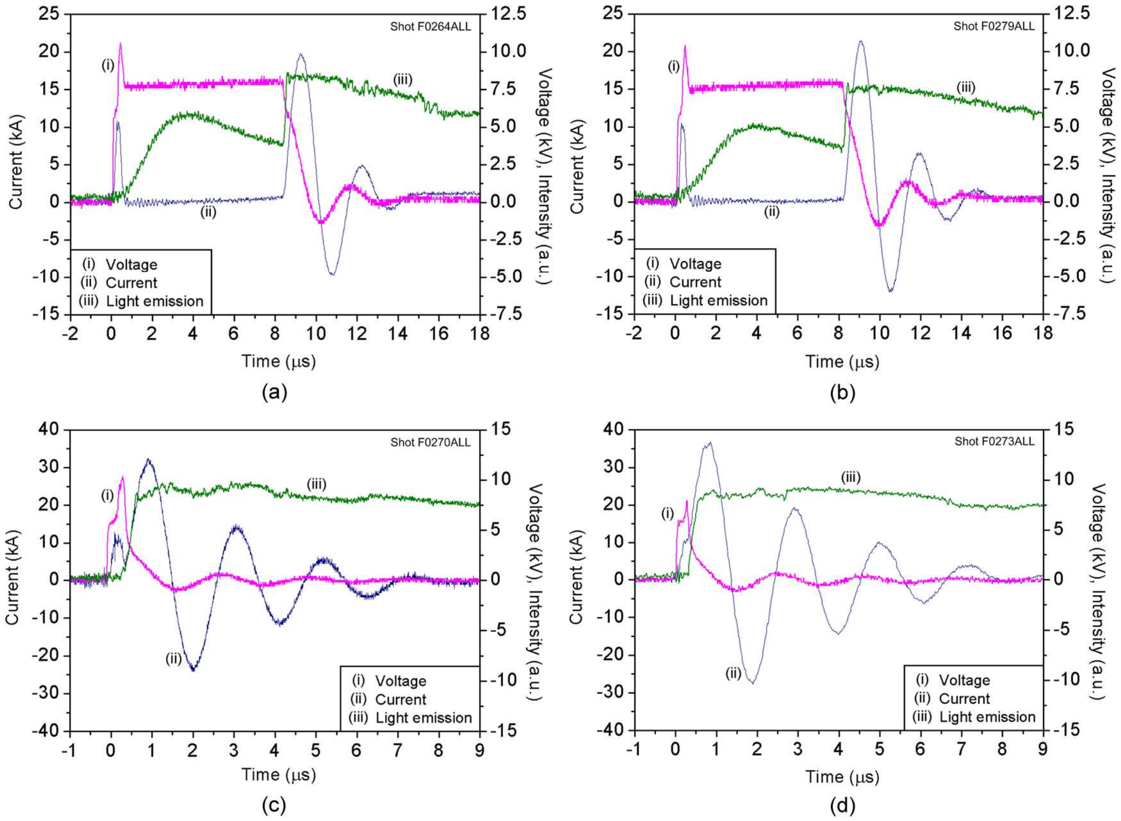


Figure 4.6: Typical current, voltage and light emission signals obtained from wire explosion in air and nitrogen at different pressures. (a) 1 bar, (b) 500 mbar, (c) 100 mbar and (d) 50 mbar.

Table 4.7: Average deposited energies at different stage of wire explosion in different ambient gases and pressures.

Ambient gas composition and pressure	**Average deposited energy (J)**		
	Peak of current spike	**Peak of voltage spike / Voltage peak**	**End of current spike / Current dip**
Air			
1 bar	4.56 ± 0.81	16.56 ± 1.08	25.03 ± 0.95
500 mbar	5.58 ± 1.86	18.43 ± 0.90	25.79 ± 1.58
100 mbar	4.94 ± 1.61	20.10 ± 1.26	25.70 ± 2.10
50 mbar	-	3.38 ± 0.83	-
Nitrogen			
1 bar	4.21 ± 1.72	17.34 ± 0.73	25.10 ± 2.13
500 mbar	4.78 ± 0.38	18.00 ± 0.83	24.10 ± 2.10
100 mbar	6.05 ± 1.77	19.03 ± 3.55	28.34 ± 4.61
50 mbar	-	3.13 ± 0.85	-

Since no current spike has been observed on current signal obtained from wire explosion in 50 mbar air and nitrogen ambient, only the average deposited energy at the voltage peak is given in Table 4.7. This is similar to the case of 100% argon ambient in section 4.2.1. The value after the plus-minus sign represents the standard deviation of the deposited energy at different stages.

Comparing the data in Table 4.7 with the energy required to change the temperature and state of the solid wire as shown in Table 4.4, several observations have been made and summarized below:

(1) Wire explosion in ambient with pressure of 1 bar and 500 mbar:

- (a) Wire has been fully melted at the time the peak of current spike is reached.
- (b) Wire has reached its boiling point at the point where the peak of voltage spike is reached.
- (c) Wire has not been completely vaporized before the first current spike ended.

(2) Wire explosion in ambient with pressure of 100 mbar:

- (a) Wire has been fully melted at the time the peak of current spike is reached.
- (b) Wire has reached its boiling point at the point where the peak of voltage spike is reached.
- (c) Wire has not been completely vaporized when the arc discharge has formed.

(3) Wire explosion in ambient with pressure of 50 mbar:

- (a) Wire has not reached the melting temperature when the arc discharge has formed.

In the next two sections, the particle size distribution and the crystal structures of the powders produced in different ambient gases will be presented. Further discussion related to the signals and the powder characteristics will be given in chapter 5.

4.3.2 Particle size analysis based on TEM images

Analysis similar to that described in sub-section 4.2.2 has been carried out, in order to determine the particle size distribution of powders obtained from the case of nitrogen and air. The median diameter and geometrical standard deviation are summarized in Table 4.8 and 4.9. The changes of median diameter with ambient gas pressure are illustrated in Figure 4.7 and 4.8. Figure 4.9 shows the typical TEM images of the nanopowders obtained from the case of nitrogen and air.

It can be seen in Table 4.8 that the median diameter for nanopowder produced in 500 mbar nitrogen is basically smaller than that shown in Table 4.5 which has been obtained under same condition. This will be discussed in section 5.3.1.1 in chapter 5.

Table 4.8: Particle size analysis for nanopowders produced by wire explosion in nitrogen at different pressures.

Pressure (mbar)	Median diameter (nm)	Geometrical standard deviation (nm)
1000	34.4	1.6
500	27.4	1.6
100	33.5	1.7
50	31.1	1.5

Table 4.9: Particle size analysis for nanopowders produced by wire explosion in air at different pressures.

Pressure (mbar)	Median diameter (nm)	Geometrical standard deviation (nm)
1000	31.3	1.6
500	31.0	1.5
100	26.4	1.6
50	23.6	1.6

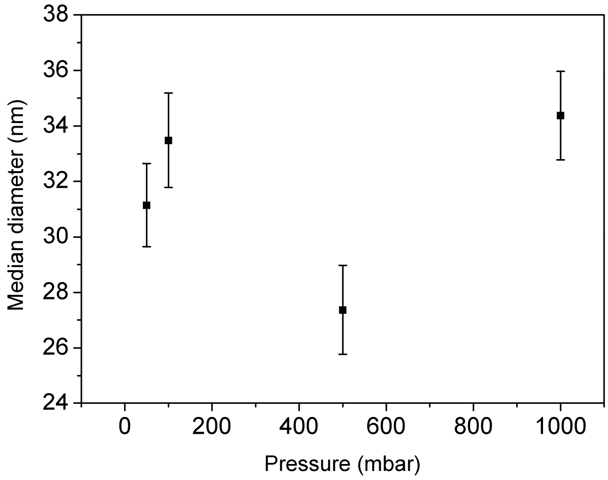


Figure 4.7: Median diameter of nanopowders produced by wire explosion in nitrogen at different ambient pressures.

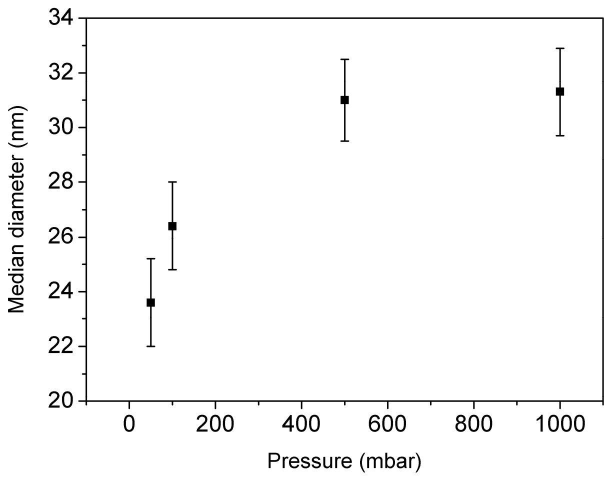
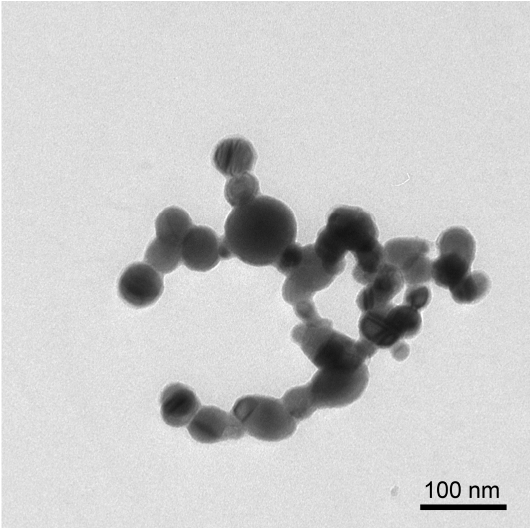
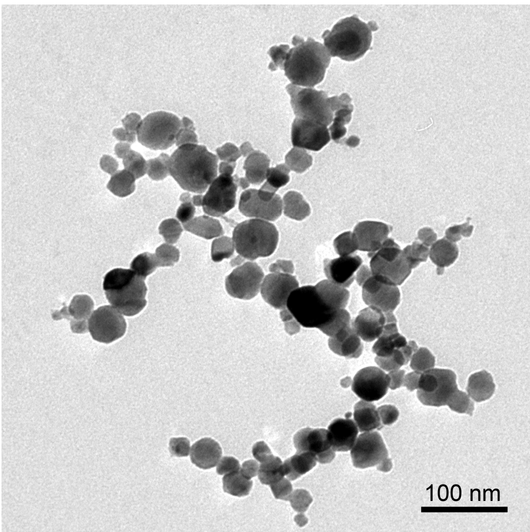


Figure 4.8: Median diameter of nanopowders produced by wire explosion in air at different ambient pressures.



(a)



(b)

Figure 4.9: Typical TEM images of nanopowders obtained from wire explosion in (a) nitrogen and (b) air.

4.3.3 Crystal structure of the powders determined from XRD analysis

From XRD results, pure copper has been observed in all samples produced from wire explosion in nitrogen. Meanwhile, copper oxide occurred in all samples produced from wire explosion in air. The results are summarized in Table 4.10 and 4.11. No peaks corresponding to copper nitride have been observed.

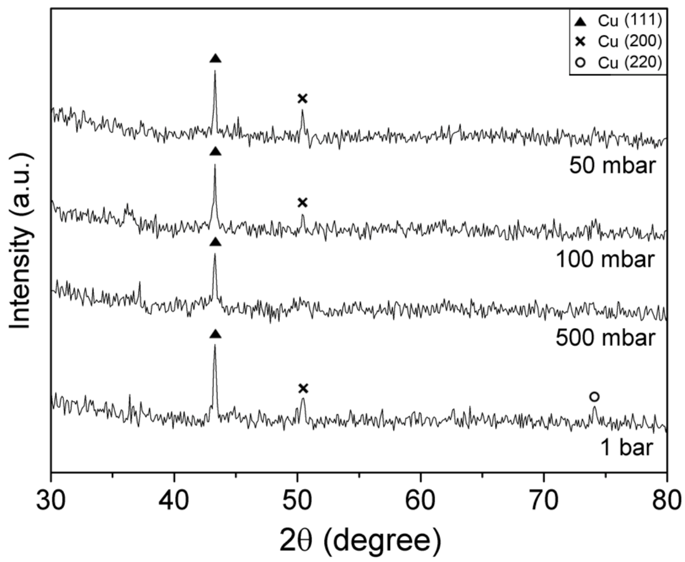


Figure 4.10: XRD patterns for samples obtained from wire explosion in nitrogen at different pressures.

Table 4.10: Summary of XRD analysis for samples obtained from wire explosion in nitrogen at different pressures.

Pressure	Compositions	2θ (hkl)	Lattice
1 bar	Cu	43.5 ^{o} (111), 50.4 ^{o} (200), 74.0 ^{o} (220)	Face-centered cubic
500 mbar	Cu	43.5 ^{o} (111)	Face-centered cubic
100 mbar	Cu	43.5 ^{o} (111), 50.4 ^{o} (200)	Face-centered cubic
50 mbar	Cu	43.5 ^{o} (111), 50.4 ^{o} (200)	Face-centered cubic

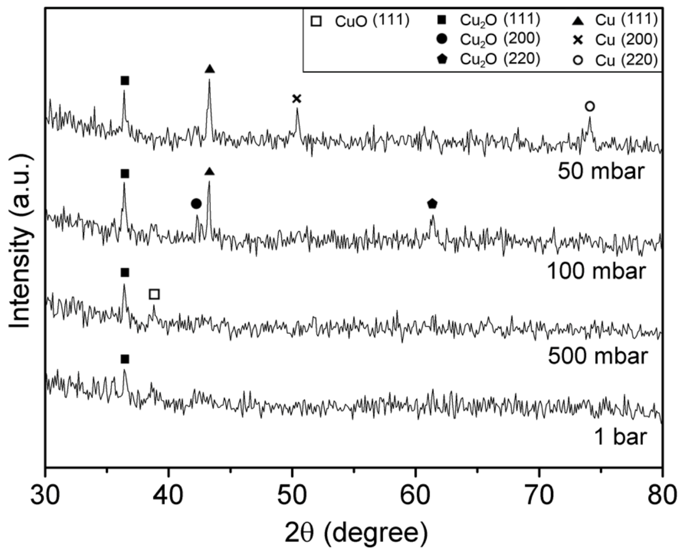


Figure 4.11: XRD patterns for samples obtained from wire explosion in air at different pressures.

Table 4.11: Summary of XRD analysis for samples obtained from wire explosion in air at different pressures.

Pressure	Compositions	2θ (hkl)	Lattice
1 bar	Cu_2O	36.5 ^{o} (111)	Cubic
500 mbar	CuO	38.8 ^{o} (111)	Base-centered monoclinic
	Cu_2O	36.5 ^{o} (111)	Cubic
100 mbar	Cu	43.5 ^{o} (111)	Face-centered cubic
	Cu_2O	36.5 ^{o} (111), 42.4 ^{o} (200), 61.6 ^{o} (220)	Cubic
50 mbar	Cu	43.5 ^{o} (111), 50.4 ^{o} (200), 74.0 ^{o} (220)	Face-centered cubic
	Cu_2O	36.5 ^{o} (111)	Cubic

4.3.4 Physical structure and appearance of powders as observed by FESEM

From FESEM, agglomerated and non-agglomerated nanoparticles as well as micron-sized spherical particles have been observed in all samples produced in nitrogen and air. The agglomerated nanoparticles show a web-like structure at low microscope magnification. Typical images showing the physical structure and appearance of the powder captured by FESEM are presented in Figure 4.12 to 4.14.

In Figure 4.12, we can see the web-like structure and micron-sized spherical particles (bright white dots) lie on top of the silicon substrate. This image has been captured at low magnification of 5000x. At higher magnification as shown in Figure 4.13, we can see that the web-like structure is actually formed by agglomerated nanoparticles. Meanwhile, non-agglomerated nanoparticles have also been observed, as shown in Figure 4.14.

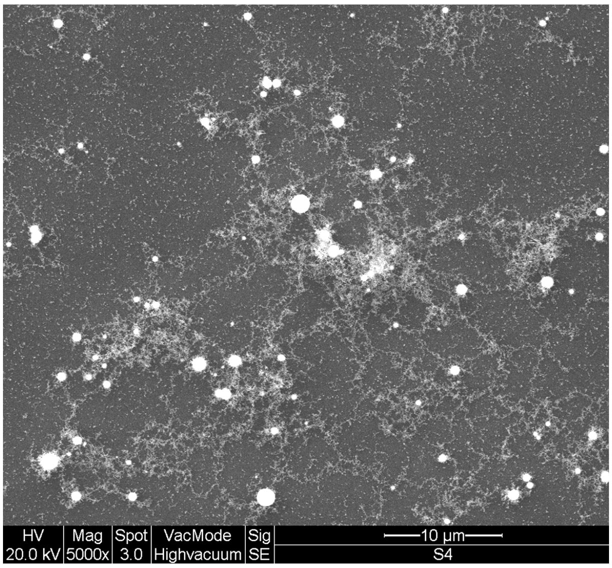


Figure 4.12: Typical FESEM image of web-like structure and micron-sized particles.

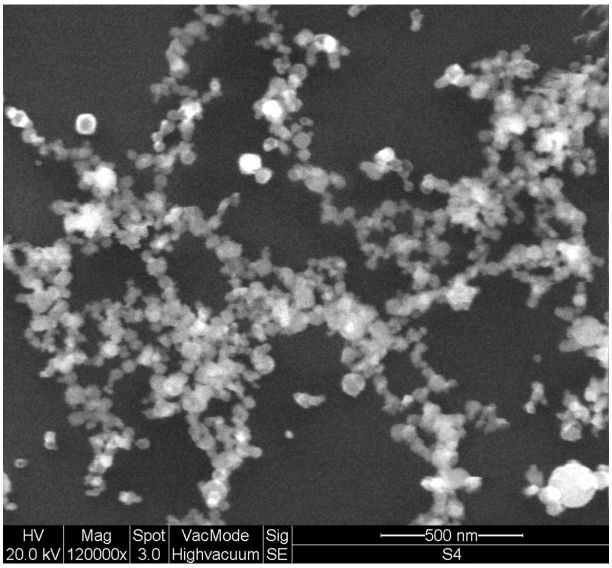


Figure 4.13: Typical FESEM image of agglomerated nanoparticles.

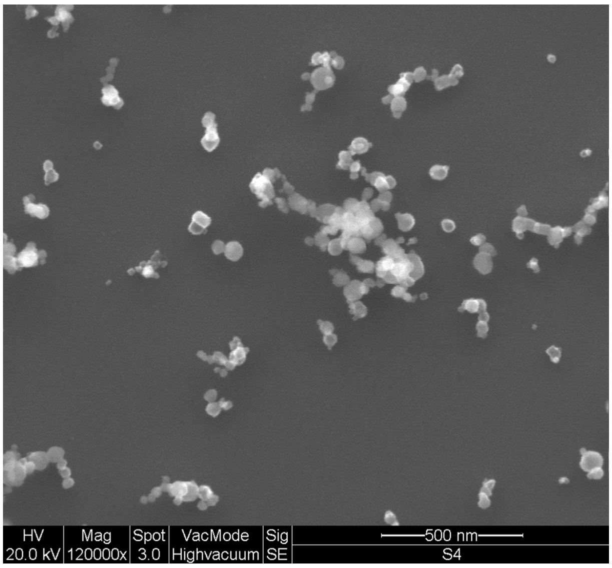


Figure 4.14: Typical FESEM image of non-agglomerated nanoparticles.