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Nanophase Ni particles produced by a blown arc method

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(Received 6 June 1994; accepted 22 October 1994)

Nanophase Ni particles (<10 nm in diameter) were produced by a blown arc method. A helium gas stream directed at the arc reduces the Ni vapor concentration and increases the quench rate. The helium gas velocity is the predominant factor influencing the size of the Ni particles. Gas velocities of 20 m/s and 56 m/s (at 26.6 kPa total helium pressure) resulted in Ni particle sizes of 13 nm and 7 nm, respectively.

Most nanophase particles produced by current methods suffer from surface contamination by impurities, which can greatly change the desired properties of a bulk sample. The inert gas condensation method[1] is one of the cleanest ways to produce nanophase materials. Because the material is physically evaporated in the absence of any precursors, there are no contaminants left on the surface of the particles, and this eliminates a common problem with chemical methods. However, this method results in very small bulk samples due to the low production rate, and the properties measured from these small samples may not represent those of bulk nanophase materials.

The arc method[2] has a higher production rate than the conventional inert gas condensation method, but produces larger particles (usually 20 to 50 nm in diameter) as a result of the higher vapor concentrations generated by the high temperature arc. To maintain the higher production rate and reduce the particle size, a blown arc setup was developed in which a helium gas stream directed at the arc reduces the Ni vapor concentration and increases the quench rate. The goal of this work was to optimize the gas velocity for our blown arc setup so as to produce Ni particles smaller than 10 nm.

Previous experiments using forced gas flow over a wide range of velocities have produced nanophase metal particles. For instance, Iwama and Hayakawa[3] used a 25 m/s helium gas stream to study silver particles, and Haas *et al.*[4] studied Cu and Pd particles with a supersonic helium gas stream. Both of these involved resistance heating, rather than arcs.

Ni vapor was created by generating an arc between a W-cathode rod and a Ni-anode block. Once the arc was started, a helium gas jet was directed through the arc. The particles were carried by the He to the collection chamber where they were captured on a liquid nitrogen cold trap. The particles were transferred to a microscope grid after the experiment and observed using a TEM.

The system was maintained at 26.6 kPa helium pressure during the run, and the current used to generate

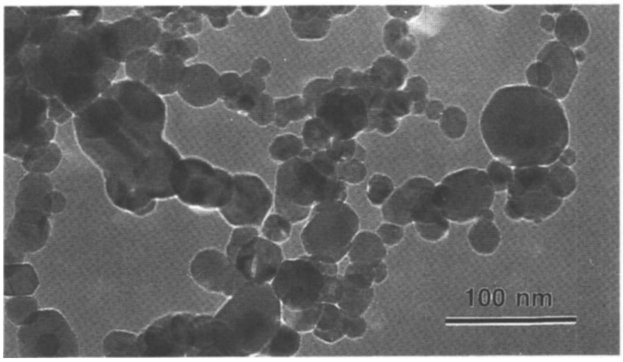

FIG. 1. A TEM micrograph of the 9 m/s specimen (average size 27.6 nm).

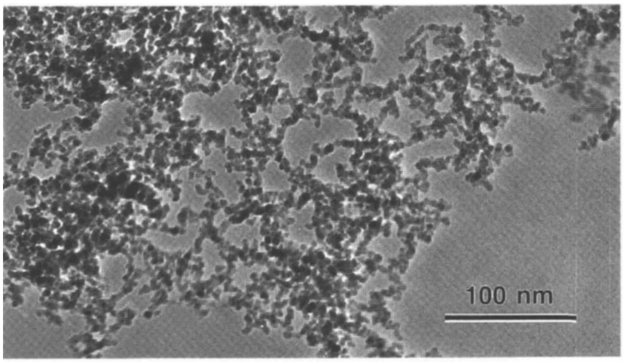

FIG. 2. A TEM micrograph of the 56 m/s specimen (average size 6.9 nm).

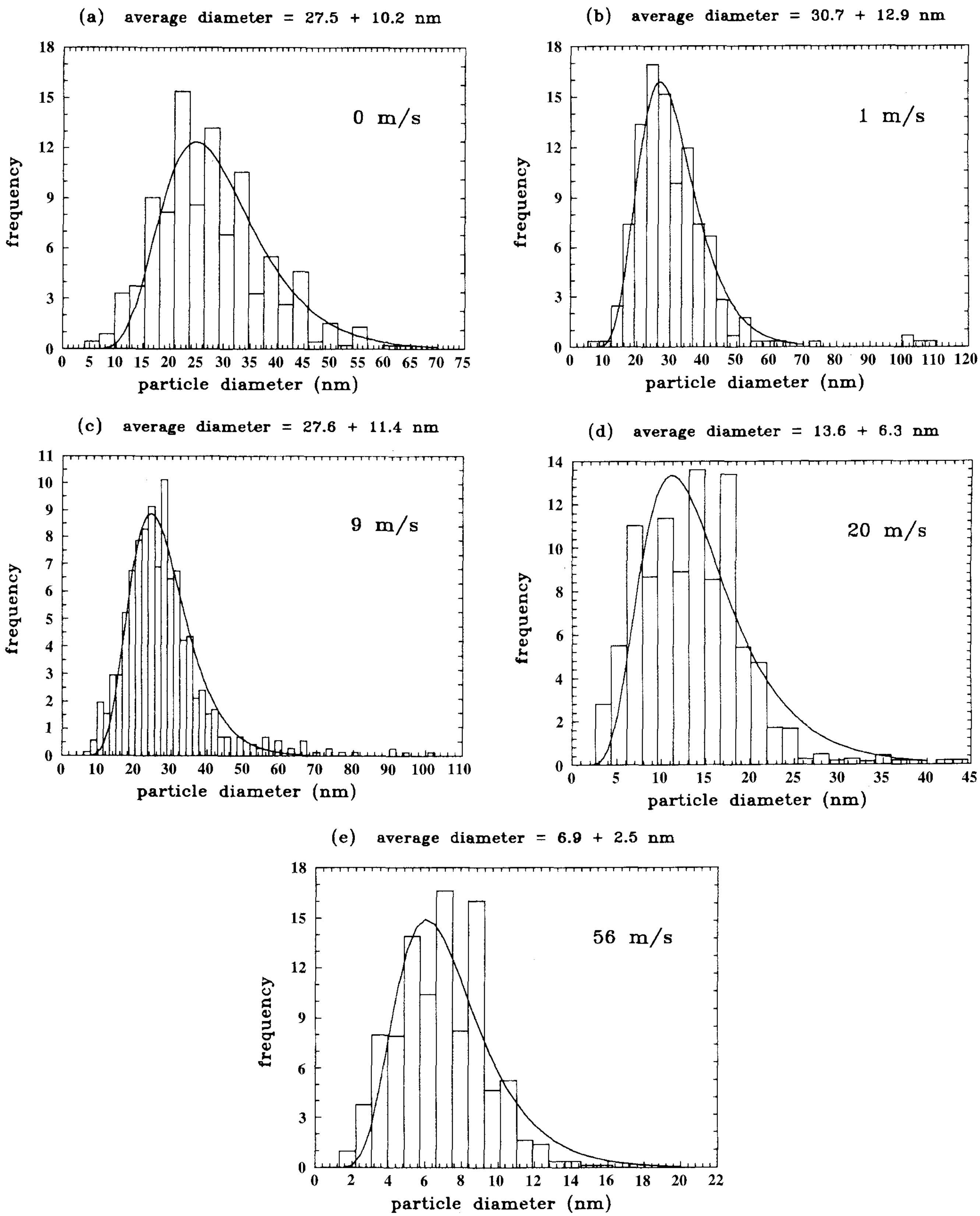

FIG. 3. Particle size distribution histograms of five specimens: (a) 0 m/s, (b) 1 m/s, (c) 9 m/s, (d) 20 m/s, and (e) 56 m/s. The superimposed curves represent log-normal distributions.

TABLE I. Particle sizes from the blown arc experiments.

Exp. no.	Gas velocity m/s	No. of particles	Average diameter, nm	Standard deviation, nm	Relative standard deviation	Geometric standard deviation
1	0	455	27.5	10.2	0.37	1.41
2	1	284	30.7	12.9	0.42	1.37
3	9	712	27.6	11.4	0.41	1.36
4	20	1397	13.6	6.3	0.46	1.53
5	56	1540	6.9	2.5	0.36	1.44

the arc was 130 A at 20 V. To produce a small arcing spot which was focused on the Ni block, the total helium pressure had to be kept at 26.6 kPa or higher. Experiments were conducted using five different gas velocities: 0, 1, 9, 20, or 56 m/s.

Several factors other than the gas velocity can influence the final particle size in an arc system,[5] including the total pressure, the gas species, and the source temperature. However, when compared to the effect of the velocity, these factors are insignificant. Therefore, only the effect of the velocity was considered in our final analysis

A TEM micrograph of the 9 m/s specimen is shown in Fig. 1. The particles are connected in chains, as is frequently observed for nanophase particles collected from an aerosol.[5] Static He and a 1 m/s jet produced very similar particles. A few large particles (>50 nm) mixed with the smaller near-average can be seen. Figure 2 is a TEM micrograph of the 56 m/s specimen with the same magnification as Fig. 1. It shows the same chain structure but with a much smaller average particle size. Note that all the particles have about the same size, and no oversize particles are seen.

The sizes of the Ni particles from the five experiments were measured manually from TEM micrographs, and the results are shown in Fig. 3. Most particles were close to spherical shape; the diameters of the nonspherical particles were determined by averaging the lengths of the longest and shortest axes. The accuracy of the measurements was limited by the resolution of the micrographs, which resulted in an uncertainty of about 2 nm at lower magnification down to about 0.5 nm at the magnification used for measuring the 56 m/s specimen. The size distributions of all five samples are close to log-normal. The curves shown superimposed on the histograms in Fig. 3 represent the log-normal distributions; they match reasonably well with the histograms. The average particle diameters of these five specimens are listed in Table I. Figure 4 shows the average particle size versus velocity, illustrating clearly the size reduction that occurs with a blown arc. The average sizes of the 0, 1, and 9 m/s specimens show no significant differences, while at 20 ms the entire distribution begins to shift to lower sizes. At 56 m/s the average particle size is much lower and the standard deviation is narrower.

The fitted log-normal curves are plotted against particle size in Fig. 5. This clearly shows the effectiveness of a blown arc in reducing the particle size and also demonstrates the existence of a threshold gas velocity below which the particle size is not measurably affected by the gas velocity. The threshold lies between 9 and 20 m/s. The five geometric standard deviations σ (defined as the size at 84.13% divided by the size at 50% in a log-probability plot) derived from the log-normal distribution curves lie within the empirical range proposed by Granqvist and Buhrman.[6] Their empirical rule for crystalline inert-gas evaporated particles is $1.36 \leqslant \sigma \leqslant 1.60$; our range is from 1.36 to 1.53.

A common problem with the arc method is the occurrence of large particles, usually 100 nm and sometimes larger, caused by instability in the arc. Because large particles were found in the production chamber, we compared the samples collected from the cold trap where a few larger particles were present in the lower velocity cases. However, few large particles were found in the 56 m/s experiment (Fig. 2), so a high gas velocity may be the solution to this problem.

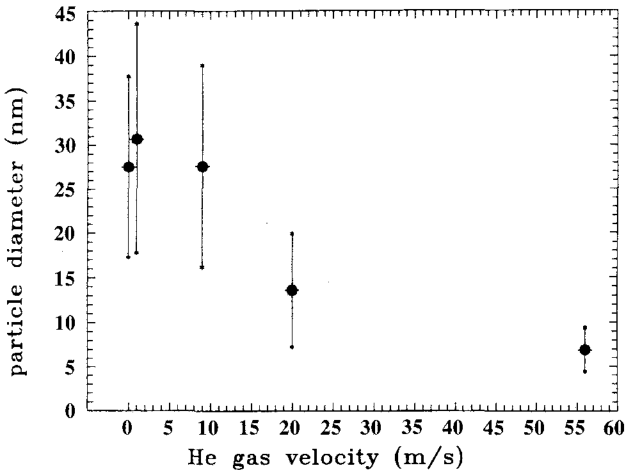

FIG. 4. Particle size as a function of gas velocity. Error bars represent one standard deviation.

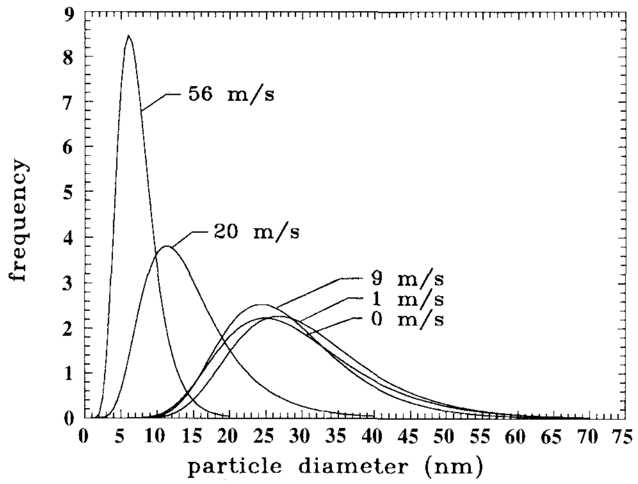

FIG. 5. Size distribution of specimens formed at five different blowing speeds as represented by their normalized frequency. The curves are the fitted log-normal distribution lines from Fig. 3.

In conclusion, the blown arc method was used successfully to produce nanophase Ni particles with average diameters below 10 nm. When the helium velocity was less than 10 m/s, the average particle size was about 30 nm, and was unaffected by velocity; while at velocities of 20 and 56 m/s, the average particle size decreased to 13 nm and 7 nm, respectively.

ACKNOWLEDGMENTS

This work was supported by National Science Foundation, Grant DMR-9202574, and by the MRL Program of the National Science Foundation, at the Materials Research Center of Northwestern University, under Award No. DMR-9120521.

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