

# Supercomputing of Cryogenic Fine Solid Nitrogen Particle Production Using Laval Nozzle for Physical Photo Resist Removal-Cleaning Technology

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The high thermal-fluid mechanical functionality of cryogenic fine solid particles of micron to nanometer size has been noted in ultrahigh heat flux cooling technology, which is applied to high thermal emission devices and nonaqueous physical nanodevice cleaning technologies.

To effectively use the high performance of such cryogenic solid particles in advanced nanotechnology, our laboratory has developed a new physical semiconductor cleaning method that employs cryogenic spraying.

In particular, given recent progress in developing high-aspect-ratio and fine-cantilever nanostructures in semiconductors, there is a strong expectation of an ultimate cleaning method that avoids physical damage. Because of the capillary force due to the surface tension of water that remains in the structure in the drying process, the fine structure is deformed, finally causing collapse or adhesions.

We have developed a new nonaqueous physical semiconductor-photoresist removal and cleaning method that employs cryogenic fine particulate spraying.

Cryogenic high-speed spraying of fine-solid nitrogen ( $\text{SN}_2$ ) in a nonaqueous resist removal and ultracleaning system for semiconductor wafers is investigated by an integrated supercomputational and experimental approach.

The fundamental characteristics of the cryogenic single-component fine  $\text{SN}_2$  particle production using a superadiabatic de\,Laval nozzle and an  $\text{SN}_2$  particulate spray for the physical photoresist removal and cleaning technology are investigated utilizing a new technique that couples measurements and computation.

The present study shows that high-speed ultrafine  $\text{SN}_2$  particles are generated continuously by the solidification of liquid nitrogen droplets induced by rapid adiabatic expansion of transonic subcooled two-phase nitrogen flow passing through the de\,Laval nozzle. Also clarified is the interactive effect of the ultrahigh heat flux cooling and impingement behavior of the  $\text{SN}_2$  particles on the resist removal performance.

We also investigate computationally the microscopic cryogenic particle heat transfer mechanism (which is difficult to obtain by conventional measurements) and the ultrahigh heat flux cooling characteristics and impingement behavior of a cryogenic fine- $\text{SN}_2$  particle on a heated wafer substrate.

Furthermore, we use laser optical measurements and particle-structure-interaction computing to investigate how the  $\text{SN}_2$  particle diameter, injection velocity, and impact angle affect the wafer substrate regarding the resist removal and cleaning performance.

Finally, we use scanning electron microscopy to analyze the microscopic condition of the resist-removed cleaned wafer surface and a new type of integrated time-of-flight secondary ion mass spectrometry (ToF-SIMS) and X-ray photoelectron spectroscopy (XPS) measurement to analyze the impurity removal characteristics.

According to this research, the resist removal characteristics associated with the impingement and cooling behavior of a single cryogenic fine- $\text{SN}_2$  particle on a heated wafer substrate were investigated computationally and experimentally in a microscopic approach.

The findings of this research are summarized as follows.

1. We conducted a new computationally assisted study of single-component cryogenic fine-particle production using mixed flow through a superadiabatic de\,Laval nozzle (converging-diverging nozzle).

This method continuously produces single-component fine solid nitrogen particles without helium refrigerant. Furthermore, almost spherical particles and a homogeneous particle diameter distribution can be obtained.

2. From the PIA measurement results,  $\text{SN}_2$  particles can be atomized to the order of several microns by using the present de\,Laval-nozzle-type fine- $\text{SN}_2$  spray production system. Especially in the dense spray core region,  $\text{SN}_2$  particle atomization is enhanced by the shear stress acting on the particles from the transonic high-speed axial spray velocity component. Therefore, finer particles are produced and have a high probability of being in that region.

3. It was found numerically and experimentally that the hybrid interactive effects of the fluid dynamic force by impingement of a fine- $\text{SN}_2$  particle and the thermomechanical effect of the resist due to the ultrahigh heat transfer characteristics contribute to the resist removal and cleaning process.

Also, the effect of ultrasonic atomization of the fine- $\text{SN}_2$  particulate flow on the ultraclean performance of the semiconductor wafer was found for the first time.

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