Cryogenic Single-Component Micro-Nano Solid Nitrogen Particle Production Using Laval Nozzle for Physical Resist Removal-Cleaning Process

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With the progress of high aspect ratio and fine cantilever in semiconductor nano structures, the ultimate cleaning method without use of water is strongly expected [1-3]. In the present study, the innovative characteristics of the cryogenic single-component micro-nano solid nitrogen ( $SN_2$ ) particle production using laval nozzle and its application to the physical resist removal-cleaning process are investigated by a new type of integrated measurement coupled computational technique.

The originality to be noted in the present study is that the continuous production of micro-  $SN_2$  particle is achieved by using single component gas-liquid two-phase flow of subcooled nitrogen through a laval nozzle. In the conventional method for solid nitrogen particle generation proposed by the authors [1, 2], the use of cryogenic helium gas were required as a refrigerant. However, since the helium is very expensive and it is specified in the strategic material and its rarity, there is a serious problem of cost for use in the cleaning process.

In order to overcome these problems, we have developed the new micro-nano SN<sub>2</sub> particle production method without use of helium refrigerant by using laval nozzle (converging-diverging nozzle). In this technique, micro-SN<sub>2</sub> particle is generated by adiabatic expansion of highspeed subcooled two-phase LN2 flow. When the flow was induced internal section of laval nozzle, the subcooled LN<sub>2</sub>-GN<sub>2</sub> flow is atomized and LN<sub>2</sub> droplet is generated at the throat, and high-speed ultra-fine SN<sub>2</sub> particle is continuously generated due to the freezing of LN2 droplet induced by rapid adiabatic expansion of subsonic subcooled two-phase LN<sub>2</sub> flow passing through the laval nozzle. To elucidate the detailed mechanism of microsolid nitrogen (SN<sub>2</sub>) generation, and integrated CFD analysis was carried out to clarify the cryogenic spray heat transfer mechanism that is difficult to obtain by conventional measurement. For the formulation of governing equations, the single component micro-SN<sub>2</sub> particle generation through the laval nozzle and the motion is governed by Navier-Stokes equations, continuity equations and energy equations. The defining feature of this phenomenon is the intense evaporation (and later condensation) that takes place at the interface of the gas and SN<sub>2</sub> particle phase. Since this instantaneous inter-phase mass exchange should be expressed in terms of enthalpy changes, we employed a simple finite-rate temperature-dependent source term to approximate the mass transfer occurring over an interfacial region. Figure 1 shows the computational result of the SN2-phase generation and volume fraction through the laval nozzle. It is numerically found that the SN<sub>2</sub> particle is continuously created by LN2 droplet freezing due to the latent heat release based on the adiabatic expansion of GN<sub>2</sub>-LN<sub>2</sub> subsonic two-phase flow just downstream of laval nozzle throat section.

Figure 2 shows the computational result of internal stress profiles of impinging  $SN_2$  particle and deformed resist. It is found that the interaction between  $SN_2$  particle and the resist is reasonably simulated. The magnitudes of stress in wafer resist increases with increase in deformation. The magnitude of pressure in  $SN_2$  particle and wafer resist

increases with SN<sub>2</sub> impingement and fragmentation. It can be numerically predicted that the resist removal performance is improved by the scraping effect of impinging micro-solid nitrogen particle fragment in the narrow trench section between the resist. Figure 3 shows the experimental results of the resist removal-cleaning using micro- SN2 spray produced by present laval nozzle method. As a result, the resist can be effectively removed and the ashing-less wafer surface cleaning has been achieved. These improved resist removal-cleaning performance is caused by the kinematic resist removal of extremely finer (nano-order diameter size) SN<sub>2</sub> particles. At the current technical stage, since it can be limited to produce in the dilute SN<sub>2</sub> particle number density condition, there is a little residual of resist in the trench part of the wiring pattern. However, we succeed in developing the single component cryogenic physical resist removal-cleaning method without use of gaseous helium.

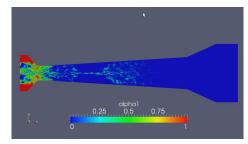


Fig. 1: computational result of the  $\mathrm{SN}_2\text{-}\mathrm{phase}$  generation and volume fraction through the laval nozzle

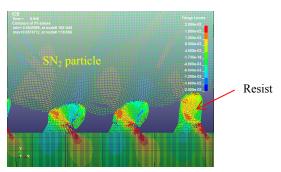


Fig. 2: Numerical result of the internal stress profile of impinging  $\mathrm{SN}_2$  particle and deformed behavior of resist

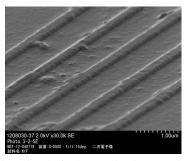


Fig. 3: Experimental result of resist removal performance using micro-SN<sub>2</sub> spray produced by single-component LN<sub>2</sub>-GN<sub>2</sub> high-speed two-phase flow through laval nozzle

## References

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