

Nanometre-Scale Visualization of Chemical Parameter Changes by T₁-Weighted ODMR Imaging Using a Fluorescent Nanodiamond

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1. pH Dependence of a Non-Carboxylated Nanodiamond.

We prepared non-carboxylated nanodiamonds as follows. The nanodiamond powder was electron-irradiated (2 MeV , $1.0 \times 10^{18}\text{ e}^-/\text{cm}^2$) and thermally annealed at $800\text{ }^\circ\text{C}$ for 2 hours under vacuum to give non-carboxylated nanodiamond characterized by FT-IR and XPS.

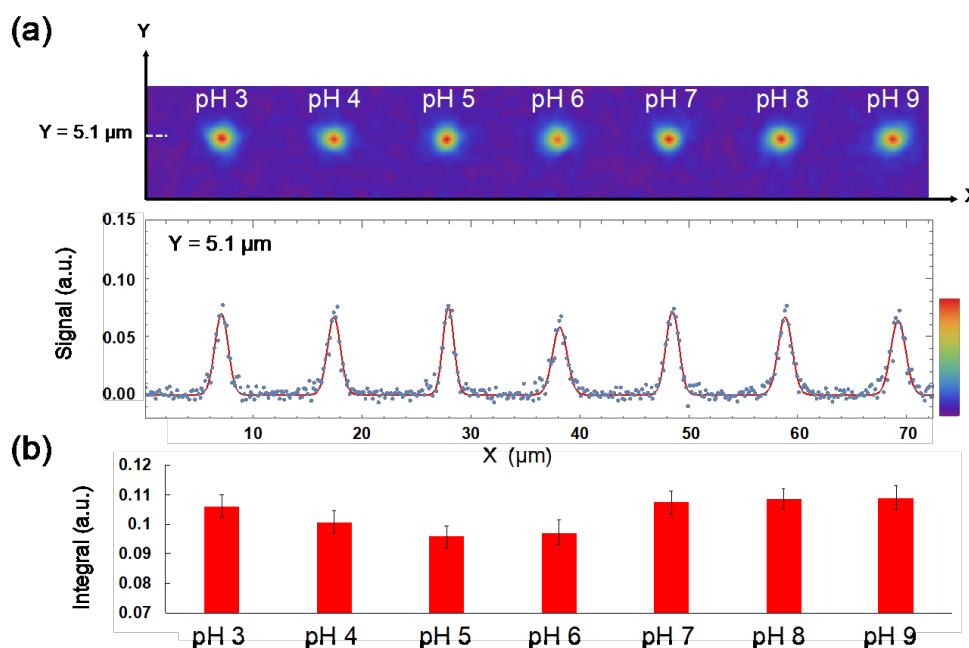


Figure S1. (a) T₁-weighted images of a non-carboxylated 100-nm nanodiamond in various pH conditions (top) and its signal intensity on a cross section Y = 5.1 mm plotted against X (bottom). The red line represents the best fit function consisting of seven Gaussian curves. (b) Definite integrals of the best fit Gaussian curves of each pH condition. Error bars represent the standard error for the fitting.

2. Spatial Resolution

pH affects the charge states of carboxy groups, which were bound to the nanodiamond surface. T_1 of NV centers in nanodiamonds varies depending on the charge states of the surface groups. Therefore, the pH sensing range is around the nanodiamond surface and 1 nm or less. On the other hand, the accuracy of nanodiamond's location estimated from its fluorescence gravity center was about 5 nm or less. When the heterogeneity of particle sizes or shapes is much more significant than the pH sensing range and location accuracy shown above, mainly, the particle heterogeneity limits the spatial resolution. Thus, the spatial resolution in our experiments is considered about 200 nm or less because of the large size distribution of the used 100-nm nanodiamonds (50–200 nm).