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# Optoelectronic properties and polar nano-domain behavior of sol–gel derived $K_{0.5}Na_{0.5}Nb_{1-x}Mn_xO_{3-\delta}$ nanocrystalline films with enhanced ferroelectricity

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High-quality lead-free piezoelectric $K_{0.5}Na_{0.5}Nb_{1-x}Mn_xO_{3-\delta}$ (KNNM*x*, $0 \le x \le 0.10$) films have been successfully deposited on Pt(111)/Ti/$SiO_2$/Si(100) substrates by a modified sol–gel method. The effects of Mn substitution on the microstructure, morphology, lattice vibrations, and optical and ferroelectric properties of the KNNM*x* films have been investigated in detail. All films are polycrystalline, crack-free and show a pseudo-cubic (pc) structure with a thickness of about 215 nm. Raman analysis indicates that the characteristic frequency of $v_1$, $v_5$ and $v_1 + v_5$ modes shifts towards lower wavenumbers with increasing Mn concentration. The optimal ferroelectric properties were obtained in the film doped with $x = 0.06$, whose remnant polarization ($2P_r$) and coercive field ($2E_c$) values at the applied electric field of 1000 kV $cm^{-1}$ are 51 μC $cm^{-2}$ and 265 kV $cm^{-1}$, respectively. The increased valence of $Mn^{2+}$, which is substituted at the $Nb^{5+}$ site as $Mn^{3+}$, plays an important role in reducing the amount of both oxygen vacancies and holes. In addition, the dielectric functions of the KNNM*x* films have been uniquely extracted by fitting ellipsometric spectra with the Adachi dielectric function model and a four-phase layered model (air/surface rough layer/KNNM*x*/Pt) in the photon energy range of 1.5–5.5 eV. The optical band gap ($E_g$) slightly decreases, while the high-frequency dielectric constant ($\varepsilon_\infty$) linearly increases with increasing Mn concentration. Moreover, temperature dependent optical dispersion behavior of the KNNM0.06 film has been investigated from 300 K to 800 K. The analysis of $E_g$ and the extinction coefficient ($\kappa$) reveals the correlation between optical properties and structural phase transition. Furthermore, a distinct in-plane (180°) polar nano-domain pattern with a well-defined rectangular phase hysteresis loop has been observed in the KNNM0.06 film from piezoresponse force microscopy (PFM) experiments. The present results could be crucial for potential multifunctional KNN-based device applications.