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In this paper we report nanocrystalline ZnO thin films deposited by varying the Ga concentrations and atmosphere gas, onto the glass substrates using spray pyrolysis technique at 450 °C substrate temperature. After deposition Ga-doped ZnO thin films were annealed at temperature 420 °C in vacuum. The morphological, structural, optical and spectral properties of synthetized thin films have been characterized by scanning electron microscopy, X-ray diffraction, Raman analysis, UV–Vis spectrophotometry and spectrofluorimetry. The XRD result shows hexagonal structure with preferential orientation along the (0002) plane and the dependence of the values of the full-width at half-maximum of this peak on the nature of the gas used in the synthesis. Also, it is found that the optical bandgap can be increased by increasing the doping level. The fluorescence spectra of ZnO thin films with 1%, 2%, 3% and 5% concentrations of Ga demonstrate that these nanostructured thin films can produce reactive oxygen species (ROS) such as singlet oxygen under ultraviolet light. Nanocrystalline ZnO thin films in function of the Ga concentration provide the phosphorescence lifetime of the charge separated states up to 102 ms.