We report a high-response hydrogen gas (H<sub>2</sub>) sensor based on a 50-nm-thick conductive Ga-doped ZnO (GZO) polycrystalline films. The GZO films were deposited on amorphous glass substrates at a temperature of 200 Celsius by ion plating with direct-current arc discharge. The Ga<sub>2</sub>O<sub>3</sub> content in the ZnO targets were 4 wt.%. Control of chemical states of adsorbed oxygen atoms on the ingrain surface and at grain boundaries and of oxygen-related point defects such as oxygen vacancies in the vicinity of the ingrain surface is essential to achieve hydrogen gas sensors showing a very strong and *immediate response* to this gas. We, thus, have been developing a novel after-arc plasma technique to generate electronegative oxygen ion (O<sup>-</sup>) for the control of the density and chemical states of the different type of defects above. The analysis of the data obtained by X-ray photoelectron spectroscopy (XPS) measurements for as-deposited GZO films indicated the presence of oxygen vacancies, O<sup>-</sup>, hydroxyl, oxygen molecule and water molecule. On the other hand, XPS study on the GZO films after the exposure to the  $O^{-}$  revealed that the intensity of the peak to the  $O^{2-}$  ions on the wurtzite structure of the hexagonal  $Zn^{2+}$  ion array increased, whereas the intensity of the peak associated with the  $O^{2-}$  ions that are in oxygen deficient regions within the ZnO matrix decreased. The above changes in the intensity of this component may be in connection with the variation in the concentration of the oxygen vacancies. This implied that some of the doped oxygen species should adsorbed on the surface of grain boundaries, trapping carrier electrons. This lead to the formation of a high and narrow energy barrier at a grain boundary in addition to the energy barrier owing to the nature of the grain boundaries such as discontinuity and disorder. In this study, we assume the chemical reaction limiting the performance of hydrogen gas sensors can be as follows: the reaction of H<sub>2</sub> with an O<sup>-</sup> ion adsorbed on a grain boundary produces a water molecule together with a free electron, resulting in a decrease in the electrical resistivity. The GZO films after the exposure to O<sup>-</sup> ions exhibited a sensitive and fast hydrogen response time within 1 second to H<sub>2</sub> gas of 0.25 % in air at a temperature of 330 Celsius. The use of extrinsic O<sup>-</sup> ions exposure with the after-arc plasma technique would be an effective way for the achievement of H<sub>2</sub> sensors exhibiting at lower operation temperature. We will propose a theoretical model of the H<sub>2</sub> sensing mechanism limiting the properties of the H<sub>2</sub> sensor.