Abstract: We demonstrate the ion-selective response of an electrolyte-gated carbon nanotube network-based field-effect transistor fabricated on a flexible polyimide substrate. Selective response toward the two prominent second messengers for cell-cell communication, namely K+ and Ca2+, is demonstrated by modifying the carbon nanotube network with different polymeric ion-selective membranes. The sensing mechanism relies on the transduction of the ionic signal in an electrical one due to an ion-activity dependent change of the membrane potential at the membrane/electrolyte interface, which leads to a change in the effective gate-potential affecting the charge transport in the semiconducting channel. These sensors can be successfully used to selectively detect concentrations of primary ions down to a concentration in the micrometer range even in solutions with a highly concentrated background of interfering ions. Our approach allows the realization of low-cost, flexible, portable and multipurpose biosensing devices.