The supercapacitance performance of birnessite is greatly hindered by poor electrical conductivity, especially when the areal mass loading of the active materials is high enough for practical application. Heterogeneous atom doping is an effective way for improving the supercapacitance performance of birnessite. Herein, we mainly investigate the influence of Fe doping on the crystal structure, electronic structure and capacitance performance of birnessite in detail by combining the experiments and theoretical calculations/simulations. It is found that Fe atoms mainly substitute the central trivalent Mn in \([\text{MnO}_6]\) octahedral after doping without changing the crystalline phase of birnessite. Meanwhile, the particle size and surface area of Fe-doped birnessite continuously increase with the increase of the content of Fe dopant. On the other hand, the electronic conductivity of the doped birnessite firstly increases and then decreases with the increase of the Fe content due to the reduced indirect band gap and the increased number of the boundary/grain interfaces. Based on these results, the influences of Fe doping on the supercapacitance performance of birnessite electrode with very high areal mass loading of \(\sim 10-12\ \text{mg cm}^{-2}\) are elaborately discussed related to morphology, structure, electrical conductivity, and ion diffusion properties.