Abstract: Piezoelectret films are prepared by modification of the microstructure of polypropylene foam sheets cross-linked by electronic irradiation (IXPP), followed by proper corona charging. Young's modulus, relative permittivity, and electromechanical coupling coefficient of the fabricated films, determined by dielectric resonance spectra, are about 0.7 MPa, 1.6, and 0.08, respectively. Dynamic piezoelectric d₃₃ coefficients up to 650 pC/N at 200 Hz are achieved. The figure of merit (FOM, d₃₃·g₃₃) for a more typical d₃₃ value of 400 pC/N is about 11.2 GPa⁻¹. Vibration-based energy harvesting with one-layer and two-layer stacks of these films is investigated at various frequencies and load resistances. At an optimum load resistance of 9 M Ω and a resonance frequency of 800 Hz, a maximum output power of 120 μ W, referred to the acceleration g due to gravity, is obtained for an energy harvester consisting of a one-layer IXPP film with an area of 3.14 cm² and a seismic mass of 33.7 g. The output power can be further improved by using two-layer stacks of IXPP films in electric series. IXPP energy harvesters could be used to energize low-power electronic devices, such as wireless sensors and LED lights.