In this study, a flexible magnetoelectric (ME) heterostructure of PZT/Ni was fabricated by depositing a (001) oriented Pb(Zr\textsubscript{0.52}Ti\textsubscript{0.48})O\textsubscript{3} (PZT) film on a thin, flexible Ni foil buffered with LaNiO\textsubscript{3}/HfO\textsubscript{2}. Excellent ferroelectric properties and large ME voltage coefficient of 3.2 V/cm.Oe were realized from the PZT/Ni heterostructure. The PZT/Ni composite's high performance was attributed to strong texturing of the PZT film, coupled with the compressive stress in the piezoelectric film. Besides, reduced substrate clamping in the PZT film due to the film on the foil structure and strong interfacial bonding in the PZT/LaNiO\textsubscript{3}/HfO\textsubscript{2}/Ni heterostructure could also have contributed to the high ME performance of PZT/Ni. (C) 2017 Author(s).