Pearlitic steel wire, with a representative sub-100 nm lamellar structure, is the strongest mass-produced steel with an excellent combination of formability and strength. This overview summarises investigations of cold-drawn pearlitic steel wire in the last decades, covering the microstructural evolution and strengthening mechanisms. Based on quantitative structural parameters, this overview covers a quantitative and extensive analysis of structure–strength relationships. By focusing on the structure, challenges and future strategy are outlined to further improve the mechanical behaviour and performance of pearlitic steel wire to widen its use in society.