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A series of six imidazole-derived thiosemicarbazones (**HL**<sup>1</sup>–**HL**<sup>6</sup>) and their copper(II) complexes (**1**–**6**) were synthesised and characterised by analytical, spectroscopic, electrochemical and single crystal X-ray diffraction techniques. In addition, solution studies and the results of antiproliferative activity in human cancer cell lines with some insights into the mechanism of cancer cell death are also reported. In particular, the substitution of one hydrogen at the terminal N-atom of the thiosemicarbazide moiety by a phenyl group resulted in slightly enhanced antiproliferative activity. **HL**<sup>3</sup> and **HL**<sup>6</sup> showed lower IC<sub>50</sub> values compared to **HL**<sup>1</sup> and **HL**<sup>4</sup> in MDA-MB-453 and LS174 cancer cell lines. The copper(II) complexes **3** and **6** exhibit a 2.4- and 4.7-fold increase of activity compared to parent proligands in MDA-MB-453 cancer cell line, respectively. The complex formation of the proligands with copper(II) increased their antiproliferative activity in all investigated cell lines. The cell cycle perturbations, apoptotic potential and the analysis of morphological changes in the A549 cell line induced by **3** and **6** revealed cytostatic rather than cytotoxic effects.