The composition of a planet’s core has important implications for the thermal and magmatic evolution of that planet. The elevated abundances of both S and C on Mercury’s surface measured by the MErcury Surface, Space ENvironment, GEochemistry and Ranging (MESSENGER) spacecraft, coupled with the low abundances of iron, suggest that the oxygen fugacity of the planet is several log units below the Iron-Wüstite buffer. These observations spark questions about the bulk composition of Mercury’s core. Here, we conducted C solubility experiments on Fe-Si metal mixtures (up to 35 wt% Si) at 1 GPa and 700–1800°C to determine the C concentration at graphite saturation (CCGS) in metallic melt and crystalline metal with varying proportions of Fe and Si to constrain the C content of Mercury’s core. All starting metals were completely enclosed in a graphite capsule to ensure C saturation at a given set of run conditions. All elements, including C, were analyzed using electron probe microanalysis. Precautions were taken to ensure accurate measurements of C with this technique including using the LDE2 crystal, the cold finger on the microprobe to minimize contamination and strengthen the vacuum, and an instrument with no oil based pumps. Our results, combined with those in the literature, show that composition is the major controlling factor for C solubility in Fe-rich metal with minimal effects from temperature and pressure. Moreover, there is a strong anti-correlation between the abundances of C and Si in Fe-rich metallic systems. Based on the previous estimates of <1–25 wt% Si in Mercury’s core, our results indicate that a carbon-saturated mercurian core has 0.63–5.3 wt% C, with 5.3 wt% C corresponding to an Si-free, Fe core and 0.63 wt% C corresponding to an Fe-rich core with 25 wt% Si. However, Ni and S are likely to be important components of Mercury’s core with Fe and Ni in chondritic proportions and some combination of S-Si-C in lower abundances (≤ 5 wt% each for S and C and up to 25 wt% Si). The Si content of the core remains the least constrained in the presumed Fe-Ni-S-C-Si mercurian core. The estimated FeO abundances in the mantle are consistent with a core that has <1 wt% Si, which would imply that bulk Mercury has a superchondritic Fe/Si ratio.