Spherical Fe-oxide concretions on Earth, in particular in Utah, U.S.A, have been investigated as an analogue of hematite spherules discovered in Meridiani Planum on Mars, in order to support interpretations of water-rock interactions in early Mars. Although several formation mechanisms have been proposed for the concretions on Earth and Mars, it is still unclear whether these mechanisms are viable because a precise formation process and precursor of the Fe-oxide concretions are missing. Here, we show evidence that Fe-oxide concretions in Utah and newly discovered Fe-oxide concretions in Mongolia, had spherical calcite (CaCO₃) concretions as precursors. Observed different formation stages of calcite and Fe-oxide concretions, both in the Navajo Sandstone, Utah, and the Djadokhta Formation, Mongolia, indicate the formation process of Fe-oxide concretions as follows: (1) calcite concretions initially formed by groundwater evaporation within aeolian sandstone strata; (2) the calcite concretions were dissolved by infiltrating Fe-rich acidic waters; and (3) mobilized Fe in acidic waters was fixed to form spherical FeO(OH) (goethite) crusts on the pre-existing spherical calcite concretion surfaces due to the pH-buffering dissolution reaction. The similarity between these Fe-oxide concretions on Earth and the hematite spherule occurrences in Meridiani Planum, combined with evidence of acid sulfate water influences on Mars, suggests that the Martian spherules also formed from dissolution of pre-existing carbonate concretions. Formation of recently discovered spherical-shaped nodules in Gale crater on Mars can also be explained by a similar process, although evidence of acid water influence is not obvious in lower strata of the Gale crater. The hematite spherules in Meridiani Planum and spherical nodules in Gale crater are possibly relics of carbonate minerals formed under a dense thick carbon dioxide atmosphere in the past.

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