

2000 年 2 月 14 日

機能材料 工学専攻	学籍番号	977272
申請者氏名	Liu Zhiguang	

指導教官氏名	梅本 実 土谷 浩一
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論 分 要 旨 (博士)

論文題目	Mechanically Driven Phase Transformation of Fullerene and Production of Fullerene Contained Metal Matrix Alloys (フラーレンの機械的誘起相変態とフラーレンを含んだ金属基複合材料の創製)
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(要旨 1,200字程度)

Carbon fullerene, since its discovery by Kroto and his co-researchers, has become a hot topic in chemistry and material engineering. Many novel chemical and physical properties of the new molecules had been discovered, contributing to the development of natural sciences. The discovery of superconductivity in the intercalation of alkali elements triggered a wide range research on the production of new materials containing fullerene molecules with novel properties. However, the conventional fabrication method will destroy the molecular structure at higher processing temperature. Mechanical alloying (MA), a solid state alloying process at ambient temperature, was therefore selected to produce the fullerene contained metal matrix composite alloys in the present study. In the thesis, fullerene (C<sub>60</sub>(C<sub>70</sub>)) fabricated through arc-discharge-soxhlet extraction was used for research. Mechanical milling of fullerene (C<sub>60</sub>(C<sub>70</sub>)), mechanical alloying of fullerene with carbide forming elements and non-carbide forming elements were investigated. The emphasis was put on the stability and basic structural evolution of fullerene molecules, the fullerene-metal mixtures and the properties of the resultant products.

Mechanical milling (MM) of pure fullerene is supposed to lay the basis to the whole work. It was found that mechanical milling could not destroy the molecular structure of fullerene (C<sub>60</sub>(C<sub>70</sub>)) but the long-range crystalline periodicity. An amorphous phase formed after 1000 hours milling was found to be fullerene (C<sub>60</sub>(C<sub>70</sub>)) polymer, including C<sub>60</sub> dimer. It is suggested that the existence of Fe contamination act as a catalyst to the "2+2" cycloaddition polymerization reaction of fullerene. By 'recrystallization' of the milled powders, two crystalline structures of fullerene polymers were observed, i.e., rhombohedral and tetragonal. It is argued that slow evaporation of toluene allows the small polymer clusters formed during MM to rearrange into bigger ones showing diffraction. Upon heating up to 678K, the toluene insoluble fullerene polymers were recovered to pristine monomers of fullerene.

Mechanical alloying of fullerene with strong carbide forming elements, such as Ti, V, Cr, Mo, W and Mn showed that fullerene has a stronger reactivity with metals than graphite. The carbide formation heat, as the driving force, plays an important role in the mechanical alloying process. Higher formation heat results in the direct formation of carbide in Ti-C systems, and the formation of carbide after the subsequent heating in V-C systems. In the systems with lower carbide formation heat, a mixture of metals with carbon is obtained even after heating the powders up to 973 K. Existence of residual fullerene molecules can be detected in W-fullerene and Mo-fullerene systems milled for 10 hours. It is suggested that the existence of metal atoms leads to the destruction of the fullerene molecules to small carbon pieces and therefore the easier formation of carbides.

Mechanical alloying of M-fullerene (C<sub>60</sub>(C<sub>70</sub>)), where M= Al, Cu, Fe, Ni, Sn, or Zn, were studied to explore the possibility of producing fullerene contained metal matrix alloys. The main results are as follows;

a. Fullerene was found to be present longer milling time according to the following order of MA powders: Zn, Al, Fe, Ni, Sn and Cu.

b. A higher hardness was observed for fullerene contained Al, while a lower hardness was observed for Zn-fullerene compacts, comparing to their graphite counterparts. The increasing hardness in Al is attributed to the formation of more homogeneously dispersed carbide due to the homogeneous dispersion of fullerene molecules. In contrary, the lower hardness of Zn-fullerene compact is suggested to be the absence of Zn carbide and existence of fullerene clusters in Zn matrix.

c. The stability of C<sub>60</sub> in a metal after MA could be classified according to the element group the metal belonged to on the periodic table.