Rapid synthesis and densification of single-phase Al-Cu-Fe quasicrystals by spark plasma sintering or microwave heating

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Introduction

Quasicrystals (QCs) are non-periodic long-range ordered solids with unique physical, chemical or mechanical properties. Their formation has been demonstrated for an impressive number of alloy systems. Beyond numerous Al-base QC-forming alloys, also non-Al quasicrystalline phases (e.g. of Ti- or Zr-base alloys) were investigated with respect to their potential use as high-capacity hydrogen-storage media or for their outstanding mechanical properties. Nevertheless, due to the wide availability of its constituents, the icosahedral ψ -phase of the Al-Cu-Fe alloy system remains most attractive for large-scale applications. Attention has been therefore increasingly paid to the synthesis of Al-Cu-Fe QC powders starting from the high-energy mechanical alloying of elemental powders followed by a heat treatment at elevated temperature. While brittle under ambient conditions, quasicrystals encounter further severe limitations in applications. Ultrafast heating processing techniques like spark plasma sintering (SPS) or microwave heating (MH) methods were then considered for the simultaneous synthesis and densification of suitable alloy powder precursors into bulk quasicrystalline solids. In this work, we discuss the rapid thermal processing of pre-alloyed nanopowders into bulk single-phase Al-Cu-Fe QCs by reactive sintering using either SPS or microwave heating, in particular with respect to the energy transfer density and to the role of electric current during densification and the crystallographic phase transformation of the alloys.

Precursor synthesis by mechanical alloying and structure evolution upon heating

The synthesis of single-phase complex metallic alloy structures by conventional heating methods usually extends over several hours or days.

High-purity (99.95%) elemental powders were mechanically alloyed using a Retsch PM400 MA ball-mill. Hexane was used as process control agent (PCA) to avoid excessive cold-welding and contamination from the milling equipment. The high-energy milling time for the formation of a homogeneous Al(Cu,Fe) cubic phase is between 6-10 hours. The sequence of phase transitions was investigated by DSC and TRXRD:

Spark plasma sintering of AlCuFe QCs

The main advantage of the SPS technique lies in the relatively short processing time (minutes) and in the *simultaneous* synthesis and densification of the precursor powders.

Upon pressure application, an initial shrinkage of the specimen occurs. The number of conduction pathways increases dramatically, thus leading to a sudden drop of the electrical resistance.

The second SPS densification step is related to the high-density nucleation of the icosahedral QC immediately above 600°C.

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Microwave heating: ultrafast synthesis of single-phase QC powders within seconds.

Using microwaves, single-phase quasicrystalline Al-Cu-Fe powders were obtained from high-purity wet-milled elemental powders in less than a few seconds as monitored *in-situ* by the TR-SXRD technique (J. Mater. Res. **23** (2008) 170). Single-phase quasicrystals are formed that retain their complex atomic structure upon cooling.

Conclusions

Spark plasma sintering and microwave heating are most effective technologies for the ultrafast sintering of nanomaterials. Both methods share a number of advantages over conventional sintering, i.e. rapid volumetric heating, use of time-varying electric currents and heat processing under pressure. Electromigration mechanisms are expected to allow for enhanced diffusion and phase transformation kinetics. All these factors support densification (e.g. by rapid heating across certain temperature intervals to bypass diffusion mechanisms not contributing to densification), while microstructural coarsening is kinetically suppressed.