

## Synopsis

The growing demand for energy is an important issue for the existence and development of humankind. In order to reduce the impact of conventional energy sources on the environment, attention should be paid to the development of new and renewable energy resources. Solar energy is a major environment friendly, renewable and sustainable energy source. Subsequently, with increasing drying up of the terrestrial fossil fuel, solar energy will potentially become an important part of the future energy structure. Concentrated solar power (CSP) technology is a productive way to make effective utilization of thermal energy from sun. Consequently, exploring high efficiency CSP technology is necessary. This has motivated the development of spectrally selective reflector and absorber materials.

Recently, reflectors have attracted considerable attention due to their potential application in CSP system. Reflector failure due to the formation of an oxide layer on metallic surfaces has been widely recognized as a major issue, often leading to CSP plant failure. Bronze (an alloy of copper and tin) mirrors, having lustrous reflective surface, were one of the candidate materials for traditional antique mirrors from ancient times. However, research involving these intermetallic materials for reflector application is still limited. The important drawbacks for these materials include complex processing conditions, brittleness, physical and chemical properties like oxidation, wettability etc. The phase composition, surface properties, free electrons density and hence the plasma frequency plays a critical role in controlling the reflectance of any materials. To this end, the main focus of this work was to develop reflector materials from alloys of Cu-Sn, Cu-Al, Cu-Sn-Zn or Cu-Sn-Al system and to understand the composition dependence reflectance properties of both bulk alloys and thin films of varying thickness.

In this perspective, the present dissertation demonstrates how one can adopt an alloy design approach to develop bulk optical reflector materials. Extending this to coatings, the efficacy of electrodeposition and thermal evaporation was also illustrated to show thickness dependent reflectance property. The importance of the development of new reflector materials for CSP technology is highlighted in chapter 1. The necessary theoretical fundamentals i.e., the physics aspect of the reflector functionality together with a review of the existing materials for reflector application are summarized in chapter 2. The general description of various experimental methodologies is made in chapter 3.

The first part of the present dissertation involves bulk alloy development and second part describes film fabrication. In a few cases, the scratch resistance and the influence of dust and humidity of the environment were also investigated. Chapter 4 demonstrates our attempt to develop a reflector material in the bulk cast form. This chapter describes the development of bulk Cu-Sn intermetallic for application as a solar reflector and further tuning them with tailored substitution of Zn or Al. A conventional metallurgical route (non-equilibrium processing techniques like vacuum arc melting and chill casting) was adopted. The second part (chapter 5 and 6) deals with thin film synthesis. The studies on intermetallic coatings of Cu-Sn alloys by electrodeposition are presented in chapter 5. Further, Cu-Al thin film deposition by thermal evaporation is explained in chapter 6. Some brief highlights of the major chapters of this dissertation are summarized below.

Chapter 4 mainly deals with the development of Cu-Sn intermetallic based solar reflector material, where further all substitution was performed using aluminium or zinc. Zinc was chosen due to its solid solution solubility with copper and aluminium was added because of its high plasma frequency. Chapter 4 essentially reports the rationale of reflector alloy design from a metallurgical and physics perspective. The results obtained with the baseline Cu<sub>41</sub>Sn<sub>11</sub> intermetallic and other alloy compositions have been analysed to bring out the influence of Al or Zn substitution to the baseline alloy. A detailed analysis of the phase assemblage utilizing Rietveld refinement of the X-ray diffraction (XRD) data and wavelength dispersive spectroscopy (WDS), attached to the electron probe micro analyser (EPMA) has been performed. The physical basis of higher reflectance has been explained on the basis of plasma frequency calculation of these alloy compositions. A good correlation between the phase abundance and plasma frequency was seen and is attributed to the high specular reflectance. Moreover, the Cu- 21.2 at % Al alloy composition, consisting of two phases Cu<sub>3</sub>Al ( $\omega_p = 6.47 \times 10^{15}$  rad/sec) and Cu<sub>0.78</sub>Al<sub>0.22</sub> ( $\omega_p = 19.25 \times 10^{15}$  rad/sec) phases, exhibited 89.5% specular reflectance and 83% solar reflectance with roughness at nanometre level and a hardness of 2.1 GPa. These results establish the suitability of an alloy design approach to obtain a new class of intermetallic reflector materials with a tailored combination of bulk specular reflectance and hardness.

Chapter 5 was focussed on the development of lustrous Cu-Sn coatings from an acidic sulfate based electrolyte containing electro generated metal ions. The deposition of individual metals and co-deposition of metals were carried out. Importantly, it has been demonstrated that lustrous coatings can be galvanostatically electrodeposited from acidic sulfate based electrolyte

in the presence of Laprol as an additive. The intricate designing of Cu-Sn codeposition was illustrated by systematic changes in the deposition conditions, such as applied current, bath composition and time of deposition. The quantitative analysis of the phase assemblage as well as compositional analysis of different phases was conducted using SEMEDS and XRD based Rietveld analysis. The coating thickness dependent surface morphology and specular reflectance was established. Essentially, we evaluated the properties such as scratch hardness and scratch adhesion, effect of dust and humidity, in reference to the projected CSP applications. Importantly, the useful combination of ~ 80 % specular reflectance and scratch resistance of Cu<sub>41</sub>Sn<sub>11</sub> film were demonstrated to be highly durable under local environmental conditions.

In chapter 6, a facile fabrication of Cu-Al thin films on flex glass substrate was demonstrated by systematically varying the applied current and rate of deposition in a thermal evaporation route. The metallic Cu-Al ingot (obtained from arc melting) was heated in vacuum by applying current, and was thermally evaporated onto a flexible glass substrate to obtain the reflector coatings. In particular, the Cu<sub>0.78</sub>Al<sub>0.22</sub> thin films with a plasma frequency  $\omega_p = 19.25 \times 10^{15}$  rad/sec, were fabricated on flexible glass substrates by resistive thermal evaporation. An attempt was made to analyze the relationship among the phase compositions, surface morphology, thickness, surface coverage and optical properties. Importantly, flexible Cu<sub>0.78</sub>Al<sub>0.22</sub> films with a specular reflectance of ~ 84 % in the solar region and scratch resistance at 900 mN load were obtained. It can be envisioned that with all these promising features, the Cu-Al films promise a great potential for use as highly reflective and flexible material for thin-film reflecting concentrators, solar energy devices and other optical mirrors.