

Chapter 2

Introduction

A review of the state of the art in the research and technology of silicon compounds for thermoelectric applications is presented in this book. It focuses on an aspect often forgotten in materials sciences: the chemical thermodynamics of materials. The silicides are interesting materials for wide applications such as sensors and thermoelectric generators and a lot of specific applications, magnetism for example, when the compounds contain magnetic properties.

Environmentally friendly materials and low cost will be the main attractivity for the mass dissemination of thermoelectric applications; mechanical strengthening and chemical stabilities versus temperature are interesting features in order to get a long-term use, and they are made with abundant elements in the Earth's crust; it is the case of silicides.

Besides the military and space applications and due to the characteristics we underlined, these materials can be applied in civil opportunities and participate to the fight against the global warming in the world.

It will become increasingly effective in encouraging consumers to purchase fuel-efficient vehicles and for manufacturers to invest in reducing CO₂ emissions and pollution. As a result, a disruptive technology step is required that will enable the manufactures of cars and marine engines and in general for all heavy industries to meet the forthcoming legislative standards. One very attractive way of achieving this objective is to generate power from the internal combustion engine (ICE) waste heat in vehicles. By doing this, the exhaust system created will offer greatly improved environmental performance due to enhancement of fuel efficiency and reduce the emissions at a cost that is affordable to the end user.

Secondly, and due to the fact that this technology is developed around automation and smart control of industrial processes in housing, the application concerning the building of autonomous power unit sources to aliment sensors in a wide range of temperature also needs to be achieved.

According to the published results, the silicides are materials with initial good thermoelectric properties. Consequently, it is reasonably permissible that these materials can get better properties by some changes in properties in a global

approach of the microstructures and structures and the research on the stabilities of compounds. These compounds present various conduction mechanisms and complex band structures. Moreover, as some of them are isotropic and some anisotropic, they could be successfully used in anisotropic thermoelectric devices. Many researches on thermoelectric properties of silicides have been fulfilled in the last 30 years, but one should remember that silicide research was initiated a long time ago by Academician Abram Fedorovich Ioffé in the former USSR [4].

This paper is divided into different chapters related to two themes: solid-state physics and chemistry of materials.

In a first section, we present a review of materials in which the physical properties are evidenced. Two kinds of materials have been widely studied: higher manganese silicides and magnesium silicides. Higher manganese silicides (HMS) MnSi_x exhibit interesting figures of merit at intermediate temperatures (573–873 K). Higher manganese silicides (HMS) with formula MnSi_x (x around 1.75) exhibit interesting figure of merit at intermediate temperatures (573–873 K) [7, 8]. These properties could also be improved by nanostructuration or doping with germanium [9, 10]. In such applications, the knowledge of the thermodynamic phase diagram is essential. This material is a p-type semiconductor. It should be associated with magnesium silicide material which is n-type. Then we describe some properties of magnesium silicide (Mg_2Si) which can be doped with heavy elements and possess potentially high performances. Compared to conventional thermoelectric materials, MnSi_x and Mg_2Si -based alloys have merit as they are non-toxic, sustainable, lightweight and low cost. We describe also some other silicides which were not deeply studied up to now, but we think that they show some interesting properties and they should be studied in the future.

The next section concerns the main originality of this paper. It is dedicated to the thermodynamic studies of multicomponent silicides. This section is divided into three subsections. Firstly, as the modern approach of phase diagrams is made presently by a global approach of phase diagrams, phase equilibria and phase stabilities, the philosophy of this approach is described. It is given by the study of thermodynamics of materials. Phase diagrams are well known as a description of a pressure, temperature and phase quantities (p , T , N_{ph}) of the phase relationships in a system, but usually they are described as pictures coming from assessed experimental results. Sometimes these descriptions are scarce or wrong. Nowadays, the thermodynamic properties must be described in a modern way. Consequently, one should add the necessary information in the system modelling. Then, the binary, ternary and multicomponent systems containing thermoelectric materials are described by using those modern tools associated in the general CALPHAD approach which include thermodynamic calculations. The phase diagrams which will be described are those presented in the section concerning properties. They are classified into two subsections.

Finally, this monograph describes some peculiarities of silicide-based materials for thermoelectric applications. These materials are promising for many applications; some of them show very high thermoelectric figure of merit, even higher than other compounds.

Multicomponent Silicides for Thermoelectric Materials
Phase Stabilities, Synthesis, and Device Tailoring

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