This dissertation has focused on the exploration of a few γ -brass types/related phases in the Ni–Zn–Ga, Ni–Zn–In, Mn–Ni-Zn, and Ir-Cd-Cu systems.

The γ -brass phases in the Ni-Zn-Ga and Ni-Zn-In system have been explored to understand how the replacement of trivalent Ga and In atoms for divalent Zn atoms could influence the structural stability and chemical compositions of γ -brass phases related to Ni₂Zn₁₁. In the Ni-Zn-Ga system, the existence, phase region, structural characteristics of the γ -brass type/related phases are detailed. In the Ni–Zn–In the ternary system, along with a pseudo binary phase, a new descendent of γ -brass related phase was discovered. Herein, the complete structure of the newly discovered phase was reported.

The current studies on the Mn_xNi₂Zn_{11-x} and Mn_xNi_{2-x}Zn₁₁ series provide an understanding between chemical composition and crystal structure. Insertion of Mn into the Zn site of Ni₂Zn₁₁ demonstrates how pseudo binary phases occur from the cubic γ -brass structure of Ni₂Zn₁₁. The samples loaded as Mn_xNi_{2-x}Zn₁₁ connect two phases: the γ -brass type pseudo binary phase (*I* $\overline{4}$ 3*m*; a \approx 9 Å), and a structurally complex phase (*F* $\overline{4}$ 3*m*; a \approx 18 Å) identified as the *T* phase in the phase diagram of the Mn-Ni-Zn system. The crystal structure of the *T* phase that was not known before this investigation. It has been characterized in this study. It is found that the *T* phase is very similar to the previous reports (Fe, Ni)Zn_{6.5}. Systematic insertion of Mn into Ni₂Zn₁₁ specifies that Mn prefers to substitute Zn site instead of Ni, possibly, due to the closer atomic size.

The influence of Cu on the crystal structure, chemical composition, and stability of the Ir₈Cd₄₁ system was also explored in this work. Interestingly, two very similar 2^3 - superstructure of γ -brass related phases were obtained in the Ir-Cd-Cu system. The crystal structures of the ternary phases at the Cu-poor region represent a new type of superstructure (2^3) of the γ -brass related phases in view of the combination of constituent cluster types positioned at the high symmetry points of the unit cell, whereas the structures at the Cu-rich region adopt the Rh₇Mg₄₄ type.

All the pseudo binary and ternary phases that occur with the variety of structures related to γ brasses are stabilized at VEC between 1.44-1.92 e/a. The range is expected for typical Hume-Rothery compounds. All these results advance our understanding of the role of the valence electron concentration (e/a) on the formation and stability of γ -brass type or related phases.

Keywords: Complex intermetallics; Gamma brass; Site preference; X-ray diffraction; Neutron diffraction.