NiMnGa Heusler alloys, functioning as either ferromagnetic shape memory alloys or magnetocaloric materials, have both practical applications and fundamental research value. Better understanding of the structure and property of NiMnGa alloys will help optimize the performance. In this study, for the first time, a combinatorial study by combining solid-to-solid diffusion couples and various characterization techniques was carried out to fundamentally investigate the NiMnGa ternary alloys. Phase equilibria, interdiffusion behavior, microstructural and crystallographic development, and mechanical properties in NiMnGa alloys were systematically examined. The microstructure and concentration profiles were examined by electron microscopy. The mechanical properties as a function of composition were assessed via nanoindentation.

Isothermal phase diagrams were examined and modified through the solubility values obtained for various phases. Both austenite and martensite were identified at room temperature in each diffusion couple with a clear interphase boundary. The compositions at the interfaces corresponded close to valence electron concentration (e/a) of 7.6, but decreased to lower values when Mn concentration increased to more than 35 at. %. Average effective interdiffusion coefficients for the β phase over various compositional ranges were determined and reported in the light of temperature-dependence. Ternary interdiffusion coefficients were determined, and indicated that Ni interdiffuses the fastest, followed by Mn then Ga. Interdiffusion flux of Ni also has strong influences on the interdiffusion of Mn and Ga with large and negative cross interdiffusion coefficients. The main ternary interdiffusion coefficients exhibited minimum values near 52 at. % Ni concentration.

Crystallographic variations in martensitic phase, including non-modulated (NM) martensite, modulated (5M or 7M) martensite, were found in the diffusion couples. The 5M and 7M martensites were only found near the interface between austenite and martensite, corresponding to compositions with lower e/a ratio. The NM martensites were found mostly away from the interface region, with high e/a ratios. The tetragonality ratio (c/a) for NM martensite generally increases with e/a ratio, but also depended on the composition. All martensitic microstructure consists of twinned variants with different orientations that were documented using electron diffraction. The twinning relationship along with the c/a ratio was correlated to martensitic transformation temperature. In addition, pre-martensitic state has been clearly observed in the cubic austenitic phase region, with distinctive tweed microstructure originating from the local lattice distortions.

Mechanical properties including reduced elastic modulus (Er) and hardness (H) as a function of composition were measured and analyzed by nanoindentation. A decrease of Er and H was observed with Mn or Ni substituting Ga, and Ni substituting Mn for the austenitic phase. However, an opposite trend was found for the martensitic phase. The softening of the elastic constants near the vicinity of martensitic transformation contributed to the sharp decrease in Er and H near the interface region. The measured Er and H had larger scatter for the martensitic phase than those for the austenitic phase, which was attributed to the martensitic variants with different orientations. Contribution from the variation in grain orientation or shape memory effect was determined to be small in this investigation.