Analysis of Interdiffusion in Multicomponent Alloys

Mohammad Afikuzzaman

M.Sc. (Applied Mathematics)

This thesis is submitted for the degree of

Doctor of Philosophy (Mechanical Engineering)

The University of Newcastle, Australia

Faculty of Engineering and Built Environment

School of Engineering

Centre for Mass and Thermal Transport in Engineering Materials

THE UNIVERSITY OF NEWCASTLE AUSTRALIA

November 2020

Statement of the Originality

I hereby certify that the work embodied in the thesis is my own work, conducted under normal supervision. The thesis contains no material which has been accepted, or is being examined, for the award of any other degree or diploma in any university or other tertiary institution and, to the best of my knowledge and belief, contains no material previously published or written by another person, except where due reference has been made. I give consent to the final version of my thesis being made available worldwide when deposited in the University's Digital Repository, subject to the provisions of the Copyright Act 1968 and any approved embargo.

Mohammad Afikuzzaman

Acknowledgement of Authorship

I hereby certify that the work embodied in this thesis contains published paper/s/scholarly work of which I am a joint author. I have included a written declaration below endorsed in writing by my supervisor, attesting to my contribution to the joint publications/scholarly work.

By signing below I confirm that Mohammad Afikuzzaman contributed (he performed calculation parts, prepared the drafts, figures and he was actively involved in the assessment of the outcomes) to the papers/ publications listed below:

Journal Article and Book Chapter

(i) **Mohammad Afikuzzaman**, Irina V. Belova, Graeme. E. Murch and John E. Morral, "Interdiffusion analysis in ternary systems to process composition profiles and obtain constant interdiffusion coefficients using one compact diffusion couple," *Journal of Phase Equilibria and Diffusion*, Vol 40, pp. 522-531, 2019.

(ii) **Mohammad Afikuzzaman**, Irina V. Belova and Graeme. E. Murch, "Investigation of interdiffusion in high entropy alloys: application of random alloy model," *Diffusion Foundations,* Vol 22, pp. 94-108, 2019.

Submitted Journal Article and Book Chapters

(i) Irina V. Belova, **Mohammad Afikuzzaman** and Graeme. E. Murch, "A new approach for analysing interdiffusion in multicomponent alloys," submitted to *Scripta Materialia*.

(ii) **Mohammad Afikuzzaman**, Irina V. Belova and Graeme. E. Murch, "Novel interdiffusion analysis in multicomponent alloys. Part 1: Application to ternary alloys," submitted to *Diffusion Foundations.*

(iii) Irina V. Belova, **Mohammad Afikuzzaman** and Graeme. E. Murch, "Novel interdiffusion analysis in multicomponent alloys. Part 2: Application to quaternary, quinary and higher alloys," submitted to *Diffusion Foundations.*

Conference Presentation (Poster)

(i) **M. Afikuzzaman,** Irina V. Belova and Graeme. E. Murch**,** "Modelling of interdiffusion in high entropy alloys," 3rd international conference on emerging advanced nanomaterials (ICEAN), Newcastle, Australia, October 30-November 2, 2018.

Conference Presentations (Oral)

(ii) Irina V. Belova, **M. Afikuzzaman** and Graeme. E. Murch**, "**Investigation of interdiffusion in high entropy alloys," TMS Annual Meeting & Exhibition, March 10-14, 2019, San Antonio, Texas, USA.

(iii) Irina V. Belova, **M. Afikuzzaman** and Graeme. E. Murch**, "**Diffusion and correlation effects in high entropy alloys," DSL, June 23-28, 2019, Athens, Greece.

(iv) **M Afikuzzaman,** Irina V. Belova and Graeme. E. Murch, **"**Computational modelling of interdiffusion in CoCrFeMnNi high entropy alloys," MRS fall meeting, December 1-6, 2019, Boston, Massachusetts, USA.

Dedication

To my father, my source of inspiration, encouragement, philosophy and understanding.

Acknowledgement

It is my great pleasure to express my deepest gratitude and appreciation to my supervisors A/Professor Thomas Fiedler, Professor Graeme E. Murch and Professor Irina V. Belova for their constant supervision, effective guidance, invaluable suggestions, endless support and kindness throughout the PhD program and preparing this thesis. I honestly appreciate their time and help what they have given to me in my research tenure. Without their help and co-operation, it would not be possible to perform my research in Computational Materials Science. In addition, I would like to express my special thanks to Professor Irina V. Belova, who trained me to explore many computation techniques to perform my modelling and simulations. She provided much critical advice in my simulations and suggested many notable additions and improvements.

I would like to thank members of the Centre for Mass and Thermal Transport Engineering Materials: Professor Rafal Kozubski (Jagiellonian University, Poland), Professor Zi-Kui Liu (The Pennsylvania State University, USA), Dr Leila Momenzadeh, Dr Mehdi Taherishargh, Dr Tumpa Rani Paul, Dr Tanvir Ahmed, Dr Ujjal Sarder, Mr Nima Movahedi and others for their helpful valuable suggestions during my research work. In addition, I would like to give thanks to Professor Yong Ho Sohn, University of Central Florida, Orlando, USA for providing the ternary experimental data which have helped to validate my study.

I would also like to offer my best regard to the faculty and staff members in the Faculty of Engineering and Built Environment, School of Engineering. Special thanks to Ms Lea Petrovic, Ms Katherine Harrison, Ms Jo Midwinter, Ms Kate Caves and Ms Sabrina Kesby for their help during my research period at the University of Newcastle. In addition, I would like to thank to the University of Newcastle for the financial support through a postgraduate research scholarship.

Last, but not least, I would like to express thank to my wife Kazi Shanchia for her sacrifice, love, encouragement and inspiring way of life. I also wish to thank my little prince Shahzaib Rizvin whose presence has brought a new motivation in my life. My special thanks to my lovely mom for her love and motivation in this period of research. Furthermore I would also like to convey my heartiest thanks to my elder brother Dr

Ashikuzzaman and sister Azmira Sultana for their love, support, motivation and kindness during this research period.

Table of Contents

List of Figures

List of Tables

List of Symbols

Arabic Symbols	
Symbol	**Description**
J_i	Flux of atoms i
L_{ij}	Onsager phenomenological coefficients
X_j	Driving force
n	Number of atomic species
N	Total number of atoms
$\Delta r_{i(j)}$	Collective displacement
V	Total volume of the system
k	Boltzmann constant
T	Absolute temperature
D_i^*	Self or Tracer diffusion coefficients
$\langle r^2 \rangle$	Mean square displacement
t	Time
$\widetilde{D}_{ij}$	Interdiffusion coefficients
D_0	Pre-exponential factor
C_i	Composition
x	Position
P	Matrix of thermodynamic factors
$\tilde{d}$	Matrix of the kinetic parts of the interdiffusion coefficients
f_i	Tracer correlation factor of species i
f_o	Geometrical correlation factor
f_{ii}	Diagonal collective correlation factors
$f_{ij}^{(i)}$	Off-diagonal collective correlation factor
r	Jump distance of an atom
N_i	Total number of jumps of atom i in time t
C_v	Vacancy fraction
w_i	Exchange frequency of an atom i
H_i	Vacancy escape frequency of atom i

$\tilde{d}_{ii}$	Diagonal interdiffusion coefficients
$\tilde{d}_{ij}$	Off-diagonal interdiffusion coefficients
x_M	Position of Matano plane
C_L	Composition of left end in Matano Plane
C_R	Composition of right end in Matano Plane
r_{ij}	Components of square root diffusivity
S_i	Total amount of i atoms
∇C^0	Composition gradient of component
ΔC_i	Difference of the composition vector
$[B]$	Eigenvectors
t_f	Diffusion time
M	Number of atoms per unit volume
M_0	Parameters directly related to the geometrical correlation factor

Greek Symbols	
Symbol	**Description**
φ	Thermodynamic factor
γ	Activity coefficients of species i
υ	Velocity of the laboratory reference frame
μ_i	Chemical potential of atoms i
$[\lambda]$	Eigenvalue

Abbreviations	
Symbol	**Description**
HM	Hall Method
EHM	Extended Hall Method
BM	Boltzmann-Matano
SF	Sauer-Freise
fcc	Face centred cubic
bcc	Body centred cubic
hcp	Hexagonal close packed
HE	Holdsworth and Elliot
MAA	Moleko, Allnatt and Allnatt
HEA	High Entropy Alloy
MPP	Matano Plane Position
EFDM	Explicit Finite Difference Method
SIMS	Secondary Ion Mass Spectrometry
MFP	Matlab Fitting Program
SQRD	Square Root Diffusivity
MSQRD	Modified Square Root Diffusivity
MSD	Mean Square Displacement
Zn	Zinc
Co	Cobalt
Cr	Chromium
Fe	Iron
Mn	Manganese
Ni	Nickle

Abstract

Theoretical, numerical and computational modelling of the interdiffusion analysis is the major part of the diffusion and diffusion-related fields of research. In this study, general approaches for the analysis of the interdiffusion composition profiles as well as interdiffusion coefficients are investigated for solid metallic alloy systems where the number of atomic components $n \geq 3$. The main focusing of the present dissertation is to analyse the interdiffusion phenomenon for ternary, quaternary and quinary (high entropy alloys) metallic systems making use of numerically and analytically for the closed form solutions. For the ternary and quaternary metallic system composition independent interdiffusion coefficients are used mainly for the detailed study of diffusion behaviour. In quinary metallic system composition dependent (as well as composition independent) interdiffusion matrices are used for the detailed study of CoCrFeMnNi HEAs. MATLAB programming language is used as the main tool for investigating the interdiffusion phenomenon in the different metallic system. Finally, the results obtained are compared with the available experimental data.