UNIVERSITI PUTRA MALAYSIA

PHYSICAL AND MECHANICAL PROPERTIES OF BILAYER CEMENTED TUNGSTEN CARBIDE AND STEEL FABRICATED THROUGH DIE COMPACTION PROCESS

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By

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November 2016

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In this study, a bimaterial of cemented tungsten carbide (WC) and steel was fabricated via die compaction process as it combines the hardness of WC and toughness of steel used for making machine tools. Major challenges related to this study is in two folds; firstly, the cobalt (Co) commonly used as WC binder has been reported to be scarce in supply and toxic making the International Agency for Research on Cancer (IARC) classify sintered WC–Co hard metals as carcinogenic and harmful to humans. Secondly, microstructural analysis has revealed the formation of detrimental phase (eta carbide) in co-sintered tungsten carbide and steel bilayer resulting in the deterioration of properties of this bilayer. Therefore, there is a need to replace cobalt with iron (Fe) as the binder, and also control the carbon (C) content in Fe as part of the composition in order to suppress eta carbide formation. WC–Fe–C and Fe–W–C bimaterial was fabricated with varying carbon content of Fe part composition (Fe–6W–xC, x = 0.2, 0.4, 0.6 and 0.8 wt.%). Sintering temperature was varied (1280^{o}C, 1290^{o}C &1295^{o}C) to control the sintering kinetics and limit mismatch between layers that commonly occur in bilayer compacts. Microstructural analysis revealed significant reduction of the eta carbide phase with increasing carbon content as the bilayer specimen, MC–0.8 with the highest carbon addition (0.8 wt.%) sintered at 1280^{o}C was observed to have vestigial trace of eta carbide phase when compared to other samples. An improved density results (6.1%) with increased carbon level resulting in stronger interfacial bond was observed in bilayer samples sintered at 1280^{o}C, while weak interfacial bond owing to shrinkage mismatch was observed in samples sintered at 1295^{o}C. Hardness values increased with increasing carbon addition at all sintering temperatures (At 1280^{o}C, MC–0.2 = 132.80 & 692.93 kgfmm^{-2} while MC–0.8 = 150.97 & 735.70 kgfmm^{-2} for Fe and WC parts respectively) which was attributed to the reduction of eta carbide formation. Through diametral compression test, bilayer samples sintered at 1280^{o}C were found to possess higher values of tensile strength which significantly increased from 45.10 MPa to 55.75 MPa with increase in carbon content.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Sarjana Sains

CIRI-CIRI FIZIKAL DAN MEKANIKAL BAGI DWILAPISAN TUNGSTEN KARBID TERGABUNG DAN KELULI YANG DIHASILKAN MELALUI PROSES PEMADATAN ACUAN

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Di dalam kajian ini, dwibahan tungsten karbid tergabung (WC) dan keluli telah dihasilkan melalui proses pemadatan acuan kerana ia menggabungkan kekerasan WC dan keliatan keluli yang digunakan untuk membuat alat-alat pemesinan. Cabaran utama ke arah ini terdapat dalam dua bahagian iaitu yang pertama, bekalan kobalt (Co) yang biasa digunakan sebagai pengikat WC adalah sukar didapati dan bertoksik menyebabkan Agensi Antarabangsa untuk Penyelidikan Kanser (IARC) mengelaskan logam keras WC–Co yang tersinter sebagai karsinogenik kepada manusia. Kedua, analisis mikrostruktur telah mendedahkan pembentukan fasa memudaratkan (eta karbid) di dalam dwilapisan tungsten karbid dan keluli tersinter, yang menyebabkan pengurangan sifat bahan dwilapisan tersebut. Oleh itu, terdapat keperluan untuk menggantikan pengikat Co dengan besi (Fe) serta mengawal kandungan karbon (C) dalam Fe di sebahagian komposisi untuk mengurangkan atau menghapuskan pembentukan eta karbid. WC–Fe–C dan Fe–W–C dwibahan telah dibentuk melalui kaedah metalurgi serbuk yang melibatkan pencampuran serbuk, pemadatan dan langkah pensinteran. Kandungan karbon di sebahagian komposisi Fe telah diubah (Fe–6W–XC, x = 0.2, 0.4, 0.6 dan 0.8 wt.%) untuk mengurangkan pembentukan eta karbid selepas pensinteran. Disebabkan pengecutan tidak sepadan kerana perbezaan ketumpatan adalah konsisten dan memberi kesan terhadap padatan dwilapisan tersinter, suhu pembakaran sampel dwilapisan telah diubah (1280, 1290 dan 1295^{o}C) untuk mengelakkan ketidaksepadanan dalam dwilapisan yang sering terjadi dalam padatan dwilapisan. Analisis mikrostruktur mendedahkan pengurangan ketara fasa eta karbid dengan peningkatan kandungan karbon sebagai spesimen dwilapisan. MC–0.8 dengan tambahan tertinggi karbon (0.8 wt.%) yang disinter pada suhu 1280^{o}C telah didapati mempunyai kesan sisa fasa eta karbid yang sedikit berbanding sampel yang lain. Kadar pemadatan yang dipertingkatkan sebanyak 6.1% menyebabkan ikatan antara muka yang kuat telah diperhatikan dalam sampel dwilapisan disinter pada 1280^{o}C, manakala keretakan dan ikatan antara muka yang lemah kerana ketidaksepadanan pengecutan yang besar dan pembentukan cecair berlebihan di antara muka telah diperhatikan dalam sampel yang disinter pada 1295^{o}C. Nilai kekerasan meningkat dengan peningkatan kandungan karbon pada semua suhu pensinteran (Pada

1280^{o}C, MC–0.2 = 132.80 & 692.93 kgfmm^{-2} manakala MC–0.8 = 150.97 & 735.70 kgfmm^{-2} bagi bahagian Fe dan WC, masing-masing) yang disebabkan oleh pengurangan pembentukan eta karbid. Melalui ujian mampatan lintang, spesimen yang disinter pada 1280^{o}C didapati mempunyai nilai kekuatan tegangan yang lebih tinggi yang meningkat dengan ketara daripada 45.10 Mpa kepada 55.75 MPa dengan peningkatan kandungan karbon.

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- the research conducted and the writing of this thesis was under our supervision;
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TABLE OF CONTENTS

		Page
ABSTRACT		i
ABSTRAK		ii
ACKNOWLEDGEMENT		iv
APPROVAL		v
DECLARATION		vii
LIST OF TABLES		xii
LIST OF FIGURES		xiii
LIST OF SYMBOLS AND ABBREVIATIONS		xvi
CHAPTER		
1	**INTRODUCTION**	1
	1.1 Background of Research	1
	1.2 Problem Statement	2
	1.3 Significance of Study	3
	1.4 Objectives	3
	1.5 Scope of Study	3
	1.6 Thesis Outline	4
2	**LITERATURE REVIEW**	5
	2.1 Introduction	5
	2.2 Powder Metallurgy	5
	2.2.1 Powder Metallurgy Process of Bilayer Components	6
	2.3 Reinforced Tungsten Carbide (WC–Co)	6
	2.4 Substituting Cobalt in Tungsten Carbide WC based Hardmetals	7
	2.5 Introduction to the Fe–W–C System	9
	2.5.1 Fe–C Binary System	10
	2.5.2 W–C Binary System	11
	2.5.3 Fe –W Binary System	11
	2.5.4 Fe–W–C Ternary Phase Diagram	12
	2.6 Isothermal Section of the Fe-W-C System	12
	2.7 Ball Milling of Powders	13
	2.8 Bilayer or Multilayer Compaction	14
	2.9 Bilayer or Multilayer Co-sintering	17
	2.9.1 Debinding of Powder Compacts	17
	2.9.2 Co-sintering	17
	2.10 Shrinkage in Sintered Bilayer Compacts	20
	2.11 Microstructural Characterisation and Eta Carbide Formation	21

	2.11.1 Control of Eta Carbide (η phase) in Cemented Carbide	23
	2.12 Hardness of Cemented Tungsten Carbide	25
	2.13 Diametral Compression Test	27
	2.14 Summary	30
3	**EXPERIMENTAL PROCEDURE**	31
	3.1 Introduction	31
	3.2 Materials and Method	31
	3.3 Fabrication of WC/Fe Bilayer Compacts	33
	3.3.1 Ball Milling of Raw Materials	33
	3.3.2 Cold Uniaxial Compaction of Milled Powders	33
	3.3.3 Sintering of Green Compacts	34
	3.4 Metallographic Sample Preparation	36
	3.4.1 Cold Mounting	36
	3.4.2 Grinding and Polishing Process	37
	3.5 Optical Microscopy Technique	38
	3.6 Field Emission Scanning Electron Microscopy Technique	39
	3.7 Physical Properties	40
	3.7.1 Density Measurement	40
	3.7.2 Shrinkage Analysis	41
	3.8 Mechanical Properties	42
	3.8.1 Hardness Measurement	42
	3.8.2 Diametral Compression Test	44
4	**RESULTS AND DISCUSSION**	46
	4.1 Introduction	46
	4.2 Evaluation of Physical Properties	46
	4.2.1 Green Density Distribution in Bilayer Compacts	46
	4.2.2 Density of Sintered Bilayer Compacts	48
	4.2.3 Microstructural Analysis of Bilayer Compacts	50
	4.2.4 Shrinkage in Bilayer Compacts	55
	4.3 Evaluation of Mechanical Properties	61
	4.3.1 Hardness of Bilayer Compacts	61
	4.3.2 Tensile Strength of Bilayer Compact	62
5	**CONCLUSION AND RECOMMENDATION**	65
	5.1 Conclusion	65
	5.2 Recommendation	65

REFERENCES	66
APPENDICES	73
BIODATA OF STUDENT	80
LIST OF PUBLICATIONS	81

LIST OF TABLES

Table		Page
3.1	Parameters for Sintering Bilayer Compacts	35
4.1	Density of green bilayer compact at different compaction pressure	47
4.2	Relative density of green monomaterial and bilayer compacts	48
4.3	Relative density of sintered bilayer compacts	48
4.4	Elemental composition of highlighted spectrum and atomic weight percent calculation of Fe_3W_3C (η phase)	53
4.5	Percentage values of Axial and Radial shrinkage of bilayer compacts	56

LIST OF FIGURES

Figure		Page
2.1	The results of TRS of WC–Fe–Cu with different Cu contents (Zhao et al., 2015)	9
2.2	Fe–C binary phase diagram (Thomazic, 2010)	10
2.3	W–C binary phase diagram (Thomazic, 2010)	11
2.4	Fe–W phase diagram (Thomazic, 2010)	12
2.5	Isothermal section of Fe–W–C system at 1280^{o}C (Thomazic, 2010)	13
2.6	The progression of motions for uniaxial die compaction (Sun et al., 2009)	15
2.7	Relative density of compacts vs applied compaction pressure (Poquillon et al., 2002)	16
2.8	Relative density against compaction pressure steel and Stellite 6. The *dashed line* corresponds to an additional point at 1020 MPa obtained for a 16-mm diameter compact at 498 kN (Pascal et al., 2012)	16
2.9	Phase diagram showing the compositions and temperature suitable to the LPS of the hard layer and TLPS of the tough layer (Pascal et al., 2009)	19
2.10	Different stages of chemistry of eta carbide, M_6C formation (Thomazic, 2011)	22
2.11	Back-scattered electrons micrograph of the phase formed at the interface. Black: pore, Dark gray: iron-rich matrix, Light gray: new phase, White: WC (Thomazic, 2010)	22
2.12	Vertical section of W – Fe – Mn – C phase diagram at (m%) 75.1 W –15.6 Fe – 4.4 Mn – C. 80:20 ratio refers to composite to metal ratio according to (Colmenares, 2011; Schubert et al., 2015)	24
2.13	Vertical section of Fe – W – C phase diagram calculated at 10 wt.% Fe (Guillermet, 1989; Fernandes et al., 2007)	25
2.14	The hardness results of WC–Fe–Cu with different Cu contents (Zhao et al., 2015)	27
2.15	Load-deformation relationship for the diametral disc test (Fahad, 1996)	28
2.16	Experimental set up for diametral compression test (Jonsén et al., 2007)	29
2.17	Force-displacement curve of diametral compression test (Jonsén et al., 2007)	29

2.18	Crack development during tests (Jonsén et al., 2007)	30
3.1	Flow chart for experimental procedure	32
3.2	(a) Planetary Monomill (b) Hahn rotary evaporator	33
3.3	(a) cylindrical die (b) Instron Universal testing machine (c) bilayer compacts (d) schematic representation of WC/Fe bilayer	34
3.4	(a) Vecstar tube furnace (b) flowmeter (c) sintered bilayer compacts	35
3.5	(a) longitudinal section of cut-off bilayer (b) front view of cut-off bilayer (c) schematic diagram of cuts made from samples (d) Lulus Cut-off machine	36
3.6	Cold mounting of samples	37
3.7	(a) Grinding and polishing machine (b) grinding process (c) samples after grinding	38
3.8	Olympus optical microscope	39
3.9	FESEM-EDX set up for surface morphology and elemental composition analysis	40
3.10	(a) Schematic diagram of sintered bilayer compacts and (b) Measurement of sintered bilayer dimensions using vernier caliper	41
3.11	Bilayer compacts sintered at (a) 1280^{o}C (b) 1290^{o}C and (c) 1295^{o}C	42
3.12	Schematic diagram of green and sintered bilayer compacts	42
3.13	Wilson Wolpert Micro-vickers hardness tester model:401MVD	43
3.14	Optical micrographs showing diamond indentations on a) Fe part b) WC part of sintered bilayer (Mag. × 5)	44
3.15	Experimental set-up of (a) sintered bilayer compact mounted on Instron Universal Testing Machine (b) fractured bilayer compact	45
4.1	Density against compaction pressure of WC/Fe bilayer compact at 30kN	47
4.2	Relative density against carbon content in WC/Fe bilayer at different sintering temperatures	50
4.3	FESEM images of samples sintered at 1280^{o}C with various C_{gr} contents: 0.2 wt.% (MC–0.2); 0.4 wt.% (MC–0.4); 0.6 wt.% (MC–0.6) and 0.8 wt.% (MC–0.8)	52
4.4	EDX analysis of spectrum 1	52
4.5	FESEM images of bilayer samples sintered at 1290^{o}C with various C_{gr} contents	54

4.6	FESEM images of bilayer samples sintered at 1295^{o}C with various C_{gr} contents	55
4.7	Optical images of microstructures at the interface of bilayer compacts with different levels of C_{gr} sintered at 1280^{o}C (Mag. × 5)	57
4.8	Optical images of microstructures at the interface of bilayer compacts with different levels of C_{gr} sintered at 1290^{o}C (Mag. × 5)	58
4.9	Optical images of microstructures at the interface of bilayer compacts with different levels of C_{gr} sintered at 1295^{o}C (Mag. × 5)	59
4.10	Axial shrinkage percentage values of bilayer samples sintered at 1280, 1290 and 1295^{o}C with different levels of carbon content	60
4.11	Radial shrinkage percentage values of bilayer samples sintered at 1280, 1290 and 1295^{o}C with different levels of carbon content	60
4.12	Relationship between hardness values and carbon addition to bilayer samples sintered at 1280^{o}C, 1290^{o}C and 1295^{o}C	62
4.13	Load-displacement relation for diametral compression test of WC/Fe bilayer compact	63
4.14	Relationship between compressive force results and composition of bilayer samples	63
4.15	Relationship between tensile strength results with various composition of bilayer compacts	64

LIST OF SYMBOLS AND ABBREVIATIONS

h	Hours
^oC	Degree Celcius
kN	Kilonewton
µm	Micrometer
mm	Milimeter
kgf/mm^2	Kilogram Force Per Milimeter Square
wt.%	Weight Percent
MPa	Mega Pascal
OM	Optical Microscopy
FESEM	Field Emission Scanning Electron Microscope
PM	Powder Metallurgy
EDX	Energy Dispersive X-ray
WC	Tungsten Carbide
WC_{in}	Interface of Tungsten Carbide Part
Fe	Iron
Fe_{in}	Interface of Iron Part
C	Carbon
PEG	Polyethylene Glycol
CTE	Coefficient of Thermal Expansion
IARC	International Agency for Research on Cancer
rpm	Revolution Per Minute
RA_m	Percentage Axial Shrinkage
RR_m	Percentage Radial Shrinkage
M_6C	Eta Carbide

CHAPTER ONE

INTRODUCTION

1.1 Background of Research

Global technological advancement has made multifunctional components a major constituent in many applications such as aeronautic, automotive parts, structural systems, cutting tools amongst others. These components are designed and assembled from a combination of different materials with complementary properties in order to develop specific functions such as thermal, mechanical, wear resistance that are not to be provided by a single material and to enhance structural efficiency or minimise weight (Pascal et al., 2009). This purpose may require the use of materials of different classes such as composite/metal (cemented carbide and steel) combining hardness and toughness properties required in machining and drilling tools (Pascal et al., 2005). In this regard, assembly techniques required to obtain these multimaterial components from single material parts often involve fragile and expensive operations (Largiller et al., 2012). Powder metallurgy (PM) is a suitable technique with limited processing steps involving powder mixing, compaction of mixed powder into a porous compact with green strength then sintering that transforms the green compact to bulk material with reduced porosity, increased density, desired mechanical and physical properties (Thomazic et al., 2010). Die compaction process, a step in PM technique is very efficient as it allows cost-effective fabrication of components, produces near net shape components. The quest to make technology economical and create excellent properties in components in numerous industries has channelled research efforts towards developing bimaterial components by conventional powder metallurgy to meet these demands. Several studies on multimaterial by PM has been carried out successfully and some with notable challenges of thermal expansion mismatch, sintering rate, shrinkage mismatch, delamination; Al_2O_3/ZrO_2 by tape-casting and lamination (Cai et al., 1997), 3Y–TZP/430L stainless steel (Dourandish & Simchi, 2009), 316L/17–4PH (Simchi et al., 2006) and W/AL_2O_3 (Boonyongmaneerat & Schuh, 2006). Boonyongmaneerat & Schuh (2006) observed a strain incompatibilty of about 4.7% due to sintering environment which induced biaxial stresses in W/Al_2O_3 bilayer while boron addition to the stainless-steel part composition minimized the shrinkage mismatch of about 9.7% observed in zirconia ceramic and stainless steel bilayer. It can now be concluded that powder compositions and sintering parameters must be explored and controlled for successful manufacturing of multifunctional components.

Tungsten carbide (WC) has attracted great interest from both engineers and academicians owing to its strength, hardness and wear resistance properties. Since its discovery in Germany in 1923 by Schroter, cobalt (Co) has been the common choice of binder for WC–based hardmetal as it provides it with adequate toughness (Penrice, 1987). The adhesion of cobalt to tungsten carbide and its capillary action or wettability provides a ductile bonding matrix for WC particles during sintering and allows the achievement of very dense components. However, some drawbacks are related with this system. The relatively high cost of cobalt and the toxicity concern when there is a possibility of high Co levels in the environment has prompted attempts by researchers to substitute cobalt binder for alternative materials (Gonzalez et al., 1995; Hanyaloglu et al., 2001).

Fe–W–C system and other iron (Fe) alloy binders have been studied in the past by researchers in an attempt to replace the Co binder. The Fe–W–C system is similar to the Co–W–C both in properties and thermodynamically, and when its composition is adjusted by the strict control of carbon addition to avoid free graphite or eta phase formation; mechanical properties such as hardness, transverse rupture strength, toughness show comparable or even superior values than those found in the WC–Co system (Fernandes et al., 2003). Fe–W–C is a steel of good toughness property with a low amount of carbon content that provides ductility property and also some tungsten to limit the tungsten gradient that will be generated by diffusion from the WC base part.

For the purpose of this study and in order to meet the growing demand for advanced components in industries, a bilayer component made up of WC–Fe–C and Fe–W–C parts with variable percentage weight of carbon in the steel part powder composition was fabricated through die compaction process to combine both hardness and toughness properties. Evaluation of mechanical properties was carried out on fabricated samples in conjunction with the physical properties after sintering at different temperatures to understand the interaction between the two layers as it affects the interfacial strength and bonding of co-sintered bilayer compacts.

1.2 Problem Statement

Technological advancement has been a great tool in global development which in turn has increased the demand for multifunctional components in structural systems, machine tools, mining and automobile industries. This development has stimulated researcher's pace towards generating specific properties such as hardness, toughness, wear-resistance by assembling different classes of materials in a bimaterial component that should not be provided in a single material to meet the increasing demand of these industries but not all of these researchers have been successful with this task due to some major challenges. In machining industries where drilling and cutting tools are largely required, the combined use of steel and cemented tungsten carbide provides toughness and hardness at the same time for this tools. Cobalt binder commonly used to provide toughness in cemented tungsten carbide for production of these tools is unfriendly to the environment and therefore must be substituted with iron (Penrice, 1987; Hanyaloglu et al., 2001; Trung et al., 2014).

In the development of cemented tungsten carbide and steel bilayer, i.e., WC and Fe base parts, Pascal et al. (2009) highlighted that the task of sintering different materials simultaneously with chemical compatibility comprising excellent mechanical properties in one component as well as suppressing formation of eta carbide at the interface of this bilayer due to depletion of carbon content in powder composition is challenging. Furthermore, it was reported that the selection of a suitable sintering temperature to form healthy and cohesive interface must be carefully considered as higher temperatures of 1300^{o}C and above have been discouraged due to the excess liquid formed during sintering at this temperature. This complex eta carbide is detrimental to the mechanical properties of the final product and therefore, it must be suppressed. In order to provide a possible solution to this challenge, die compaction process which is a step in PM technique was required to fabricate this multifunctional bilayer component made up of hard and tough parts with new powder formulations after which evaluation of physical and mechanical properties of the bilayer compacts was carried out under different sintering temperatures to determine its reliability and achieve excellent properties.

1.3 Significance of Study

The need for multifunctional components is fast growing in numerous industries. Powder metallurgy (PM) has provided a cost effective assembly technique of composite/metallic components. Fabrication of these components through PM majorly provides industries with better components having different material properties assembled together which could serve multiple functions. As a result of increasing application of tungsten carbide in areas such as machining industries, there arises the need to substitute the toxic and scarce cobalt binder for iron binder in WC composite. Interestingly, if some processing parameters such as carbon content and sintering temperature are well controlled, the proposed WC/Fe bilayer can provide superior properties of adequate strength and toughness to start a cut without chipping or destroying the tool's cutting edge and hardness or wear resistance to keep a cut going once it is started.

Therefore, this study examined some experimental analysis conducted to fabricate WC/Fe bilayer component with emphasis particularly on physical and mechanical properties which are key issues in determining components life's span and the possibility to enlarge the range of applications of tungsten carbide in drilling tools and other applications in different industries where they would be subjected to high efforts even in corrosive environments.

1.4 Objectives

1. To analyse the physical properties of sintered bilayer compacts at various sintering temperatures to achieve a defect-free interface.
2. To evaluate the mechanical properties of sintered bilayer compacts.

1.5 Scope of Study

Multifunctional components are made up of different materials that possess specific properties like mechanical, thermal, electrical, corrosion resistance. The scope of this work is focused on processing of WC–Fe–C and steel (Fe–W–C) bilayer through die compaction process of PM technique with the carbon content of the steel composition varied (0.2, 0.4, 0.6, 0.8 wt.% C_{gr}) to compensate for carbon loss during sintering and prevent eta carbide formation. The mechanism for bonding WC/Fe bilayer is through liquid phase sintering (LPS) where the liquid required for interlayer diffusion is formed around 1138°C (Pascal et al., 2009) and sintering above 1300°C for 1 hr is also reported to deform bilayer. Therefore, sintering temperature was varied (1280°C, 1290°C

&1295^{o}C) to control the sintering behaviour of bilayer compacts and maintain sound interface between layers.

The physical and mechanical properties of the samples were analysed and comparisons were made after sintering at different temperatures for 60 minutes. Shrinkage mismatch were also observed for all samples at different sintering temperatures through geometrical means as material parts in bilayers have been reported not to exhibit similar behaviour during sintering.

1.6 Thesis Outline

The first chapter which is the introductory part of this thesis gives an insight into the study as a whole. It has the background, problem statement, significance of study and research objectives. Also inclusive is the scope which highlights the areas which this study will cover.

Chapter two presents a critical survey of relevant investigations in the line of this study. Overviews of the processing technique, backgrounds, effectiveness of WC-based hardmetals as well as concept of multifunctional components are presented to give basis to the procedural steps embarked upon in this study.

An experimental documentation of the steps and procedures employed for this work are described in chapter three. Here, discussions on the steps involved in the processing technique of bilayer samples, equipments utilized for analysis and microscopic techniques are presented.

Chapter four presents detailed results obtained in the course of this study and the discussion of the data in relation with the research objectives and the existing findings related to this field of study.

The conclusion and summary of results of this study are presented in chapter five followed by recommendations for future research studies.

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LIST OF PUBLICATIONS

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Ojo-Kupoluyi, O. J., Tahir, S. M., Azmah Hanim, M. A., Baharudin, B. T. H. T., Khamirul, M. A., & Anuar, M. S. Role of carbon addition on the microstructure and mechanical properties of cemented tungsten carbide and steel bilayer. *International Journal of Advanced Manufacturing Technology*, *(Submitted)*

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