



Faculty of Manufacturing Engineering



Nor Syazana Adilah Binti Zulkifli

**Master of Manufacturing Engineering
(Advanced Materials and Processing)**

2021

**GRAPHENE NANOPATELETS MODIFIED CHEMLOK
ADHESIVE SYSTEM FOR NATURAL RUBBER -
ALUMINIUM BONDING**

NOR SYAZANA ADILAH BINTI ZULKIFLI



**A thesis submitted
in fulfillment of the requirements for the degree of Master of
Manufacturing Engineering in Advanced Materials and
Processing**

Faculty of Manufacturing Engineering

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

2021

DECLARATION

I declare this thesis entitled “Graphene Nanoplatelets Modified Chemlok Adhesive System for Natural Rubber-Aluminium Bonding” is the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

Signature :

Name : NOR SYAZANA ADILAH BINTI ZULKIFLI

Date : 31th JANUARY 2021

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

APPROVAL

I hereby declare that I have read this thesis and in my opinion, this thesis is sufficient in terms of scope and quality for the award Master of Manufacturing Engineering in Advanced Materials and Processing.


Signature :  **PROF. MADYA DR. NORAIHAM BINTI MOHAMAD**
Profesor Madya
Fakulti Kejuruteraan Pembuatan
Universiti Teknikal Malaysia Melaka

Supervisor Name : ASSOC. PROF. DR. NORAIHAM BINTI MOHAMAD

Date : 31th JANUARY 2021

اونيورسيتي تيكنيكل مليسيا ملاك
UNIVERSITI TEKNIKAL MALAYSIA MELAKA

DEDICATION

To you all

my beloved father, Zulkifli Bin Othman

my appreciated mother, Rohayati Binti Ahmad

my adored sisters and brothers,

all related lecturers, technicians and fellow friends

for moral support, cooperation, encouragement and understandings

Thank You So Much & Love You All Forever



ABSTRACT

Nowadays, the engine mount in automotive undergoes various efforts for properties improvement due to the highest demand for better performance, more safety, and greater coziness. Generally, it is made from a combination of rubber and mild steel with a Chemlok adhesive system but the same adhesive system gives insufficient bonding strength between natural rubber and aluminum alloy. Therefore, this study is aiming to study the potential of a nano-manipulated adhesive system by modifying the existing Chemlok adhesive system in promoting better bonding strength between natural rubber-aluminum alloy (NR-Al). The GNPs modified Chemlok adhesives were prepared via two steps ultrasonic-assisted stirring process using an ultrasonic bath and hot plate. There are two main parts of this research project, Part A on the preparation and characterization of GNPs modified Chemlok 220 adhesive system and Part B on the properties and characteristics of the NR-Al bonded with the unmodified Chemlok 220 (control) and GNPs modified Chemlok 220 systems. In Part A, the modified Chemlok adhesive system was analyzed for gel time, XRD, Raman, FTIR, and DSC analyses. The analyses proved that the modification was successful with safe flash and gel time for utilization, and XRD, Raman, and FTIR showed that the modified system consists of GNPs characteristics peak that dispersed with certain intercalation with Chemlok matrix, presence of active functional groups, and showing the reinforcing effects and thermal stability. In Part B, the suitable substrates' surface conditions (Al surface roughness and NR condition), the process parameters (sweeping time and primer presence), and the GNPs loading at 0wt%, 0.5wt%, 3wt%, and 7wt% for both unmodified and GNPs modified Chemlok adhesive were investigated. Three layers of the Chemlok adhesives were applied on the pre-treated aluminum alloy (Al) substrate. Then, the NR-Al substrates were bonded using a hot press machine under the pressure of 100 kgf/cm² for 20 minutes at the temperature of 140°C. Then, the samples were subjected to a 90-degree peel test based on ASTM D429 by UTM machine to evaluate the bonding strength and the peel-fractures surfaces were evaluated both physically and under SEM. The Al-surface prepared by sandblasting, uncured NR compound, sweeping time of 2 minutes with the presence of Chemlok 205 primer were selected as the suitable parameters for NR-Al bond preparation. The adhesive strength was observed to increase with the increment of GNPs in the GNPs modified Chemlok 220 adhesives and reached the percolation threshold at 3 wt% for the system without primer and at 0.5 wt% for the system with primer. The improvement in peel strength for the effect of suitable GNPs loading with primer in modified adhesive system is 30%. The GNPs modified Chemlok system introduced the combination of cohesive and adhesive failures that was further supported by the morphological characteristics at higher magnifications scale. The peel strength of the adhesive systems was highly correlated with the extent of the mode of failures experienced by the system. Therefore, this finding is significantly vital to improving the existing adhesive bonding system for natural rubber-aluminum alloy bonding in automotive components like the engine mounting.

ABSTRAK

Kini, pencagak enjin automotif melalui pelbagai usaha peningkatan sifat kerana permintaan tinggi untuk prestasi yang lebih baik, lebih banyak ciri keselamatan, dan keselesaan yang lebih baik. Secara amnya, ia diperbuat dari gabungan getah dan keluli biasa menggunakan sistem perekat Chemlok namun sistem perekat ini memberikan kekuatan ikatan yang tidak mencukupi antara getah asli dan aloi aluminium. Maka, kajian ini bertujuan untuk mengkaji potensi sistem perekat yang dimanipulasi-nano dengan mengubahsuai sistem perekat Chemlok sedia ada dalam menguatkan kekuatan ikatan yang lebih baik antara getah semula jadi - aloi aluminium (NR-Al). Perekat Chemlok yang diubahsuai GNP disediakan melalui proses pengadukan berbantu ultrasonik dua langkah menggunakan mandian ultrasonik dan plat panas. Terdapat dua bahagian utama projek penyelidikan ini, Bahagian A mengenai penyediaan dan pencirian sistem perekat Chemlok 220 yang diubahsuai GNP dan Bahagian B mengenai sifat dan ciri NR-Al yang terikat dengan sistem Chemlok 220 yang tidak terubahsuai (kawalan) dan Chemlok yang diubahsuai GNP 220. Pada Bahagian A, sistem perekat Chemlok yang diubahsuai dianalisis untuk masa gel, analisis XRD, Raman, FTIR, dan DSC. Analisis membuktikan bahawa pengubahsuaian berjaya dengan masa kilat dan masa gel yang selamat untuk digunakan, dan analisa XRD, Raman, dan FTIR menunjukkan bahawa sistem yang diubahsuai terdiri daripada puncak ciri GNP yang tersebar dengan interkalasi tertentu dengan matriks Chemlok, kehadiran kumpulan fungsi aktif, dan menunjukkan kesan pengukuhan dan kestabilan terma. Pada Bahagian B, keadaan permukaan substrat yang sesuai (kekasaran permukaan Al dan keadaan pematangan NR), parameter proses (masa sapuan dan kehadiran primer), dan pemuatan GNP pada 0wt%, 0.5wt%, 3wt%, dan 7wt% untuk kedua-dua sistem yang tidak diubahsuai dan pelekter Chemlok yang diubahsuai GNP disiasat. Tiga lapisan perekat Chemlok 220 disalutkan pada substrat aluminium (Al) yang diprarawat. Kemudian, substrat NR-Al diikat menggunakan alat penekan panas di bawah tekanan 100 kgf/cm² selama 20 minit pada suhu 140 °C. Kemudian, sampel menjalani ujian pengelupasan 90-darjah berdasarkan ASTM D429 menggunakan mesin UTM bagi menilai kekuatan ikatan dan permukaan patah-terkelupas kemudiannya dinilai secara fizikal dan di bawah SEM. Permukaan Al yang disediakan oleh peledakan pasir, sebatian NR yang belum matang, masa sapuan 2 minit dengan kehadiran primer Chemlok 205 dipilih sebagai parameter yang sesuai untuk penyediaan ikatan NR-Al. Kekuatan perekat diperhatikan meningkat dengan kenaikan GNP dalam perekat Chemlok 220 yang diubahsuai GNP dan mencapai titik ambang perkolasi pada 3 wt% untuk sistem tanpa primer dan pada 0.5% berat untuk sistem dengan primer. Peningkatan kekuatan pengelupasan diperhatikan meningkat hingga sekitar 30% dengan adanya primer di hampir semua nilai pemuatan GNP. Sistem Chemlok yang diubahsuai GNP menunjukkan kombinasi kegagalan kohesif dan rekatan yang disokong oleh ciri-ciri morfologi pada skala pembesaran yang lebih tinggi. Kekuatan pengelupasan sistem perekat sangat berkaitan dengan tahap mod kegagalan yang dialami oleh sistem. Oleh itu, penemuan ini sangat penting untuk meningkatkan sistem ikatan perekat Chemlok sedia ada untuk ikatan getah semula jadi-aloi aluminium bagi komponen-komponen automotif seperti pencagak enjin.

ACKNOWLEDGEMENT

In the name of ALLAH, the most gracious, the most merciful, with the highest praise to Allah that I can manage to complete this master project successfully.

My respected supervisor, Associate Profesor Dr. Noraiham Binti Mohamad for the great mentoring that was given to me throughout the project. Besides that, I would like to express my gratitude to her for the kind supervision, advice, and guidance as well as for exposing me to meaningful experiences throughout the study.

Last but not least, I would like to give special thanks to Fakulti Kejuruteraan Pembuatan (FKP) and Universiti Teknikal Malaysia Melaka (UTeM), all lecturers, teaching engineers, technicians, and friends, who gave me knowledge, motivation, and cooperation mentally in completing this report. They had given their critical suggestion and comments throughout my research. Not to forget, my appreciation towards the contributions of the raw materials by HML Auto Industries (M) Sdn. Bhd. (HML). A special thank you to Mr. Teng Ming Ming, the Managing Director, and Mr. Muhammad Afiq Ani for the supports.

Finally, I would like to thank everybody in FKP, UTeM, and HML who was vital in this Master Project (MP) report, as well as expressing the apology that I could not personally mention each one of you in this report.

TABLE OF CONTENTS

	PAGE
DECLARATION	
APPROVAL	
DEDICATION	
ABSTRACT	i
ABSTRAK	ii
ACKNOWLEDGMENT	iii
TABLE OF CONTENTS	iv
LIST OF TABLES	viii
LIST OF FIGURES	ix
LIST OF ABBREVIATIONS	xi
CHAPTER	
1. INTRODUCTION	
1.1 Background	1
1.2 Problem Statement	3
1.3 Objectives	6
1.4 Scope of Study	6
1.5 Significant of Study	7
1.6 Thesis Overview	8
2. LITERATURE REVIEW	
2.1 GNPs as Filler	9
2.1.1 Carbon Nanomaterials	9
2.1.2 GNPs Properties	10
2.1.3 Agglomeration of GNPs	12
2.2 Chemlok as Matrix	14
2.2.1 Thermoset Adhesive	14
2.2.2 Chemlok Properties	16
2.2.3 Approach to Apply Chemlok	18

2.3	Rubber Metal Bonding	19
2.3.1	Natural Rubber Properties	19
2.3.2	Properties of Aluminium	21
2.3.3	Adhesion Behaviour	22
2.4	Adhesion Strength and Mechanism	27
2.4.1	Factor of Adhesion Strength	27
2.4.2	Primer Application in Adhesive	29
2.4.3	Surface Treatment of Substrate	30
2.5	GNPs /Chemlok as Nano-Adhesive	32
2.5.1	Modified Adhesive System	32
2.5.2	Mechanism of Adhesive Bonding	33
2.5.3	Effects of GNPs in Adhesive	35
2.6	Summary	39

3. METHODOLOGY

3.1	Overview	40
3.2	Raw Material	42
3.2.1	Natural Rubber	42
3.2.2	Aluminium Alloy	42
3.2.3	Graphene Nano Platelets	43
3.2.4	Adhesive and Primer	43
3.2.5	Solvents	44
3.3	Preparation of Graphene Modified Chemlok Adhesive System	44
3.3.1	Preparation and Design Matrix of Modified Chemlok Adhesive System via Try and Error Experiment	44
3.4	Determination of Properties and Characteristics of NR-Al bonded with unmodified Chemlok 220 and GNPs modified Chemlok 220	46
3.4.1	General Preparation of NR-Al Bonding	46
3.4.2	Stage 1: Determination of Suitable Substrates' Surface Condition and Process Parameters by Unmodified Chemlok 220	48
3.4.2.1	The Effect of Al Substrate Surface Roughness	48
3.4.2.2	The Effect of NR Substrate Curing Condition	50
3.4.2.3	The Effect of Adhesive Sweeping Time	51

3.4.3	Stage 2: Determination of Properties and Characteristics of GNPs Modified Chemlok 220 Adhesive System	51
3.4.3.1	The Effect of Curing Temperature	51
3.4.3.2	The Effect of GNPs Loading and the Presence of Primer in Unmodified and GNPs Modified Chemlok	53
3.4.4	Stage 3: Failure Mode Analysis and Morphological Characteristics of NR-Al Peel-Fractured Surfaces	54
3.5	Testing/ Analysis of Adhesive System	54
3.5.1	90° Peel Test to Determine the Peel Strength of NR-Al Bond	54
3.5.2	Structural Analysis by XRD	55
3.5.3	Structural Analysis by Raman	56
3.5.4	Compositional Analysis by FTIR	56
3.5.5	Thermal Analysis by DSC	56
3.5.6	Morphological Analysis by SEM	57
4.	RESULT AND DISCUSSION	
4.0	Overview	58
4.1	Part A: Characteristics of Prepared GNPs Modified Chemlok 220 Adhesives via Try and Error Experiment	59
4.1.1	Gel Time of GNPs modified Chemlok 220 Adhesives	60
4.1.2	XRD Analysis of GNPs modified Chemlok 220 Adhesives	63
4.1.3	Raman Analysis of GNPs modified Chemlok 220 Adhesives	65
4.1.4	FTIR Analysis of GNPs modified Chemlok 220 Adhesives	67
4.1.5	DSC Analysis of GNPs modified Chemlok 220 Adhesives	69
4.2	Part B: Properties and Characteristics of NR-Al bonded with the unmodified Chemlok 220 and GNPs modified Chemlok 220 systems	71
4.2.1	Stage 1: Properties and Characteristics of Control Chemlok 220 System for the Determination of Suitable Substrates' Surface Condition and Sweeping Technique	71
4.2.1.1	The Effect of Al Substrate' Surface Roughness	71
4.2.1.2	The Effect of NR Substrate' Surface Condition	75
4.2.1.3	The Effect of Adhesive Sweeping Time	77

4.2.2 Stage 2: Properties and Characteristics of GNPs Modified Chemlok 220 Adhesive System	80
4.2.2.1 The Effect of Curing Condition on Peel Strength of Modified Chemlok 220 Adhesive System	80
4.2.2.2 The Peel Strength of Modified Chemlok 220 Adhesive System Cured at T_{cure}	82
4.2.3 Stage 3: Failure Analysis of NR-Al Peel-Fractured Surfaces	86
4.2.3.1 Failure Mode Identification	86
4.2.3.1 SEM Observation	89
5. CONCLUSION	
5.1 Conclusion	95
5.2 Recommendation	98
REFERENCES	99
APPENDICES	
A Gantt Chart I and II	
B Result Turnitin	



اونيورسيتي تيكنيكل مليسيا ملاك
UNIVERSITI TEKNIKAL MALAYSIA MELAKA

LIST OF TABLES

TABLE	TITLE	PAGE
2.1	Techniques for dispersion in polymer composite	12
2.2	The Chemlok adhesive selection based on type of rubber	17
3.1	Properties of natural rubber	42
3.2	Properties of aluminium alloy	42
3.3	Properties of GNPs	43
3.4	Properties of Chemlok 205/220	43
3.5	Properties of ethanol and acetone	44
3.6	Formulation of GNPs modified Chemlok adhesive system (old)	45
3.7	Formulation of GNPs modified Chemlok adhesive system (new)	46
3.8	DOE for the function of Al substrate surface roughness	49
3.9	DOE for the function of NR substrate curing condition	50
3.10	DOE for the function of adhesive sweeping time	51
3.11	DOE for the function of adhesive curing temperature	52
3.12	DOE for the function of adhesive formulation	53
4.1	Raw data result of D peak and G peak	66
4.2	Comparisons of T _g and T _m for unmodified and modified Chemlok	70
4.3	Metal substrate condition with Ra value	72
4.4	Solvent vapour pressure	78
4.5	Raw data results from peel test for sample without primer	83
4.6	Raw data results from peel test for sample with primer	85
4.7	Failure mode analysis of fractured surface after peel testing	87

LIST OF FIGURES

FIGURE	TITLE	PAGE
2.1	Graphene sheets with covalent bonded atoms by Van der Waals forces	13
2.2	Chemical structure of general rubber	19
2.3	The bonding layer	23
2.4	Rubber-to-metal adhesion mechanism with bonding agent in phase (top) (b) Film formation principle with aqueous-based bonding agents (below)	25
2.5	Adhesive and cohesive force	26
2.6	Mechanical adhesion	27
2.7	Electrostatic adhesion theory at polymer	28
2.8	Inter diffusion theory	28
2.9	Adhesion mechanism	29
2.10	Oxide layer morphology	30
2.11	Rubber metal bonding flowchart	34
2.12	GNPs production	37
3.1	Flowchart of overall research methodology	41
3.2	Weighted GNPs in bottle treated and GNPs dispersed in ethanol	45
3.3	The coagulated mixture of Chemlok 220 with ethanol treated GNPs	45
3.4	The surface roughness testing	47
3.5	Mold cavity and hot press machine	48
3.6	Al surfaces: (a) original, (b) sand blasted, (c) chemically treat	46
3.7	The GNPs modified Chemlok 220 adhesive systems	52
3.8	Peeling test piece to gain the rubber-metal bond strength (90°)	55
3.9	Adhesion pattern and Scanning Electron Microscope	58

4.1	GNPs powder, well-dispersed ethanol treated GNPs and coagulation in ethanol treated GNPs-Chemlok 220 system	59
4.2	Flash time and Gel time test	60
4.3	Gel time and flash time at various GNPs content in Chemlok 220	61
4.4	XRD spectra of modified Chemlok/GNPs at (0, 0.5, 3 and 7) wt%	63
4.5	Raman spectra of unmodified and GNPs modified Chemlok 220 systems	65
4.6	FTIR spectra for polyol and pre-polymer of polyurethane resins	67
4.7	FTIR spectra of modified Chemlok at varies GNP wt%	68
4.8	DSC thermogram curve of unmodified Chemlok and GNPs modified Chemlok adhesive 7wt% GNPs	69
4.9	Illustration of GNPs create thermal conductive channel	70
4.10	Effect of surface preparation to adhesive strength	73
4.11	Illustration of surface roughness effect on the rubber-aluminum bonding and the wetting characteristics based on the surface roughness	74
4.12	Effect of uncured and cured natural rubber to peel strength	76
4.13	Sweeping adhesive on metal substrate	77
4.14	Effect of sweeping time to peel strength	79
4.15	Mechanical adhesion based on the surface roughness	79
4.16	Effect of sample curing mechanism to peel strength	81
4.17	Peel strength (maximum stress) of modified Chemlok/GNPs at	85
4.18	Two types of failure mode mechanism	86
4.19	SEM micrographs of NR-Al peel-fractured surfaces bonded with neat Chemlok adhesive at 0wt% GNPs	89
4.20	SEM micrographs of NR-Al peel-fractured surfaces bonded with modified Chemlok adhesive at 0.5wt% GNPs	91
4.21	SEM micrograph of neat Chemlok and Chemlok/3wt% GNPs	92
4.22	SEM micrograph of NR-Al peel-fractured surfaces bonded with modified Chemlok adhesive at 0.5wt% GNPs	93
4.23	SEM micrograph of modified Chemlok 220 adhesives at 0.5 wt%, 3.0 wt% and 7.0 wt% GNPs loading (1000x)	93
4.24	SEM micrographs of void formation on NR substrates at 100x and 1000x magnifications for the modified Chemlok adhesive system	94

LIST OF ABBREVIATIONS

ASTM	-	American Society for Testing and Materials
SEM	-	Scanning Electron Microscope
SOP	-	Standard Operation Procedure
XRD	-	X-Ray Diffraction
PSA	-	Particle Size Analysis
DSC	-	Differential Spectroscopy Calorimeter
FTIR	-	Fourier Transform Infrared Spectroscopy
VOC	-	Volatile Organic Compound
GNPs	-	Graphene Nanoplatelets
CLD	-	Costrained Layer Damping
ELVs	-	End of Live Vehicles
SBR	-	Styrene Butadiene Rubber
CR	-	Chloroprene Rubber
NBR	-	Nitrile Rubber
EPDM	-	Ethylene Propylene Diene Rubber
NR	-	Natural Rubber
SBR	-	Styrene Butadiene Copolymers
ISO	-	International Standard Organization
GHG	-	Greenhouse Gas
WBL	-	Weak Boundary Layer
CNT	-	Carbon Nanotubes
POSS	-	Polyhedral Oligometric Silsesquioxanes
IMB	-	In-mold Bonding

CHAPTER 1

INTRODUCTION

1.1 Background

For decades, rubber-metal combination creates many automotive parts like engine mount, suspension, and silent blocks. The interaction of metal and rubber transmit forces, enable defined movements, attenuate and dampen vibrations by isolating vibration energy. So, vibrations and noise caused by engine transmission are hardly noticeable (Wen et. al., 2017). The passive engine mount uses a rubber-metal bond to deal with vibration attenuation by isolating vibration sources and optimize the systems (Ooi et. al., 2010). Despite form coziness, automotive is under pressure for better performance like better fuel efficiency and carbon emissions drop (Fentahun and Savas, 2018). Modern automotive demands led to the advancement of rubber-metal generation with enhancement of wear, impact and heat.

Lightweight usage to fulfill automotive needs by rubber-metal bond ranging in size from motors mounts to suspension parts. The combination of rubber flexibility and metal stability makes them special. Weight reduction is vital since vehicle trade remains market better performance, better fuel efficiency as load minimize. But it is challenging to link different materials. Structural bonding is not preferable due to the reliability of long-term bonding capability and performance (Guadagno et. al., 2015). Previously, the automotive industries had implemented bonding of rubber with steel but, aluminum replaces steel due to better features like light, great strength-to-weight ratio, and superior mechanical properties (Miller et. al., 2000). For instance, the engine mount facing incremental demand for lower fuel intake, and good energy efficiency which requires new material selection.

Commonly, rubber-metal bonding is used for products that need both combinations of rubber flexibility and metal stiffness. Due to great importance in the car industry, the effort on ensuring better adhesion between both always becomes a concern. By part design, substrate choice must consider load and desired durability. The engine mount is made of rubber and steel to secure dynamic forces transmission in a vehicle and absorb road shocks and engine vibrations, so engine movement is absent. The steel is used to absorb impact energy in an accident but with great weight pressure (Shashank, 2016). The aluminum replaces steel as lightweight material but current bonding systems are insufficient to offer reliable bonding strength for the rubber to aluminum alloy.

Automakers prefer adhesive joining with the primer coating method as it meets the lightweight principle as it is suited to current demand. Chemlok adhesive is highly utilized by industries to bond rubber to metal for many vehicle parts. Although it is proven to create significant bonding with steel, however, it still fails to provide sufficient bonding strength for bonding rubber to aluminum alloy. So, one way to improve adhesive strength is creating nano-adhesive by reinforcing nanoparticles into the adhesive matrix. So, modified Chemlok adhesive was studied to improve the bonding strength of rubber-metal bond by introducing nanofiller in the adhesive blends together from the surface preparation. The adhesive and adherent should have strong interfacial interaction when they are in contact resulting in a great adhere effect as the surface molecules of solid interact causing inter-diffusion to occur. Since materials differ, elements like interfacial interaction and cross-linkage in adhere mechanism are concerning (Iraj et. al., 2012) to avoid structural and compositional alterations that later giving a negative impact on bonding performance.

1.2 Problem Statement

Moving forward with technology on rubber engine mount as demand in using light material, technology on joining system is still lack. Automaker prefers adhesive bonding as it meets the lightweight principle. But, there is limited study regarding ways to promote better adhesion between rubber and metal being implemented. Common way to foster high strength bonding is making substrate surface to interlock rubber and substrates. Sand-blasting is applied for improving surface energy and surface roughness followed by cleaning it with alcohols to dissolve impurities (Iraj et. al., 2012).

Industries like HML Auto Industries Sdn. Bhd that uses primer and adhesive is intended to meet the standard set by the government. They have used the Chemlok 205/220 adhesive system to bond rubber-metal components in an engine mount. It is a two-coat bonding system of adhesive and primer (Ismail et. al., 2015). Chemlok 220 is the bonding agent meanwhile Chemlok 205 is used as the primer coat for surface protection and corrosion resistance. Both of its used solvent-based vulcanizing adhesives agents for curing. Chemlok 220 uses xylene and Chemlok 205 has a mixture of chlorinated rubber and phenolic resin dissolved in methyl ethyl ketone. The phenolic resins aim for strong chemical interactions on a metal surface (Ismail and Harun, 2017). In general, the Chemlok system has provided excellent rubber to steel bonding and good design freedom. It produces high-quality reliable products for a longer lifespan. But, xylene and chlorinated solvent agents release volatile organic compound (VOC) bringing negative side effects to human health and environmental quality hence the evaporation rate needs to be controlled (Zhang et. al., 2018). Furthermore, it is well reported that the use of current adhesive materials Chemlok 205/220 is insufficient to provide enough strength to bonded rubber on aluminum sheets. Therefore, a study on new adhesive modification by introducing nanofiller is vital to compensate for the issue.

The introduction of nanofillers in the current adhesive system may improve the bonding strength, require smaller amounts for might eliminates the necessity to use the primer. Graphene is a promising candidate to be used as filler due to ultrahigh strength $\sim 1060\text{GPa}$ (Ma et. al., 2018), higher surface area property $\sim 2630\text{m}^2/\text{g}$ (Papageorgiou et. al., 2017) that giving higher surface contact for interaction. It has good thermal conductivity of $5 \times 10^3 \text{ W/mK}$ and capable to dissipate heat by creating conducting pathways and avoid static charge growth. High surface area causes graphene to easily agglomerates and need to be dispersed well in an organic solvent for surface treatment before it could be added into a polymeric matrix. But, graphene has low compatibility with polar solvents. So, incorporating graphene nanoplatelets (GNPs) seems like the best approach to impart the superior properties of graphene into the existing Chemlok adhesive system. It has an appreciable degree of polarity to be dispersed in an organic solvent with OH- pole from hydroxyl groups. High dispersity GNPs in an adhesive system could boost aging resistance and prolong bonding life via effective heat transport as engine mount life depends on thermal dissipation capacity.

Previously papers state that the effect of formulation plays a vital role in any adhesive system. Almost the majority of them showed that filler percentage in adhesive boosts bonding property due to good material characteristics, function, and performance (Quan et. al., 2018). Adhesive bonding is a bonding mechanism with two main forces, which are adhesive force between adhesive and adherent, while cohesive force in between bonding agent. Both forces need to be balanced as there are different surface energy despite the bonding agent. The fracture was likely to occur in the middle or center of the adhesive area that can be seen from the peel test. The strong contribution of the effect of nanofillers onto the adhesive strength makes it one of the important factors to be studied.

Chemlok is a thermoset adhesive that necessary to undergone curing to function. The curing mechanism influences the level of crosslinks which dictates the mechanical properties of the adhesive. However, the bonding strength induced by the adhesive to rubber-metal bonding is affected by various factors 1) the substrate conditions, 2) adhesive formulation, and 3) processing parameters. To foster durable, high strength bonding could be achieved by preparing substrate surface to include an active interface for linkage. The substrate surface energy must greater than liquid adhesive to enable spread throughout the substrate. The engine mount manufacturer uses a primer coat for surface protection and corrosion resistance before applying adhesive. It aims in adjusting surface free energy for a better wetting effect, triggers chemical reactions of adhesive and adherend, inhibits substrate corrosion, and as an intermediate film to boost bond strength. Gel time is crucial as bonding strength will reduce when the adhesive pre-coated sample area pre-exposes to excessive energy sources that may induce the curing process. Therefore study on substrate conditions, presence of primer, and processing parameter to control the gel time is vital. This is because the adhesive polymeric chains change their molecular structure from separated chains to 3D networks during curing. It could be disturbed by the presence of other functional groups that may be introduced during the substrate preparation and throughout the bonding process.

So, this project is investigating the potential of GNPs modified existing Chemlok adhesive systems for the effect of different system formulations (at different GNPs loadings and the presence of with or without primer) towards the increment of peel strength values. The effects of Al and NR substrates' surface condition, adhesive sweeping time and adhesive formulation for unmodified and modified adhesive systems were also carried out.

1.3 Objective

The objectives of this research are:

- i. To analyze the properties of GNPs modified Chemlok 220 adhesive system at different weight percentages of GNPs loadings (0, 0.5, 3, 7 wt%).
- ii. To evaluate the effect of suitable substrate conditions, process parameters and adhesive formulation on the peel strength of natural rubber-aluminum bonded by unmodified and GNPs modified Chemlok 220 adhesive systems.
- iii. To correlate the peel strength of NR-Al bonded with GNPs modified Chemlok 220 adhesive system with the peel-fractured surface morphologies by SEM observation.

1.4 Scope of Study

The scopes of this research are:

- i. To prepare graphene nanoplatelets, GNPs modified Chemlok 220 adhesive system with the incorporation of different GNPs loadings, which are 0 %, 0.5 wt%, 3 wt%, and 7 wt% using a direct stirring and mixing via ultrasonication by try and error experiment. The modified adhesive systems were analyzed using XRD, Raman Spectroscopy, FTIR, and DSC to measure the efficiency of the preparation.
- ii. The NR-Al sheets were bonded with an unmodified Chemlok 220 adhesive system to determine the suitable substrate conditions (surface roughness of Al sheet and NR curing condition), process parameters (sweeping time), and adhesive formulation (presence of Chemlok 205 primer). The suitable parameters were determined from the peel strength of the natural rubber-aluminum bond subjected to a 90-degree peel test according to ASTM D429-Method B standard by using a 20kN UTM machine.

- iii. Then, the NR-AI were bonded with GNPs modified Chemlok 220 adhesive system using the suitable substrates' surface condition, sweeping time, and curing condition. The contributions of GNPs loadings with and without the presence of Chemlok 205 primer were measured by the peel strength of the NR-AI bond subjected to a 90-degree peel test according to ASTM D429-Method B standard by using a 20 kN UTM machine. The fracture surfaces from this peel test were observed by SEM and the failure mechanism was deduced from the morphological characteristics.

1.5 Significant of Study

The significance of this research can be summarized as follows:

- i. The filler existence can potentially replace the current adhesive system limitation due to insufficient strength bonding. The new system introduces Graphene in Chemlok adhesive system to provide a more efficient bonding between rubber and metal.
- ii. The natural rubber-based blend is material for technology-related products that are natural, safe, durable, and sustainable compared to the synthetic rubbers made from a petroleum source.
- iii. Lastly, the modified adhesive system is simple and easily adaptable with the existing manufacturing processing line. It provides an easy solution at a relatively lower manufacturing cost. It would be beneficial to manufacturing, automotive, electronics, and other industries that utilized Chemlok systems in their production.

1.6 Thesis Overview

This report has been divided into five chapters. Chapter 1 provides this project introduction consist of the background study, problem statement, objectives, scopes, significances of the study, and thesis overview. Chapter 2 contains the literature review of research discussing related theoretical knowledge, fundamental theory, existing findings reported by other researchers on the relevant topics on NR-Al adhesive system and Chemlok adhesive system. Chapter 3 explains the methodology used in the development of GNPs modified Chemlok adhesive systems to bond natural rubber and aluminum. Then, it covers the peel testing and characterization analysis techniques such as XRD, Raman Spectroscopy, FTIR, DSC, and SEM for structural, compositional, thermal, and morphological observation. Chapter 4 addresses the result and findings from the testing and analyses performed on samples produced to investigate several factors throughout the research works. It discusses the obtained peel strength of the NR-Al bonded by the Chemlok adhesive systems, correlates the findings with analyses, and evaluates the contributing factors towards the achieved bonding strength. Chapter 5 concludes the objectives of the study, highlighting the vital elements and provides recommendations for further study.