

**FEASIBILITY STUDY OF REPROCESSED
ACRYLONITRILE BUTADIENE STYRENE (ABS) AND
OPTIMIZATION OF INJECTION MOULDING
PARAMETERS VIA TAGUCHI METHOD**

NG CHIN FEI

UNIVERSITI SAINS MALAYSIA

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MOULDING PARAMETERS VIA TAGUCHI METHOD**

by

NG CHIN FEI

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for the degree of
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TABLE OF CONTENTS

Acknowledgement.....	ii
Table of Contents.....	iii
List of Tables.....	vi
List of Figures.....	viii
List of Abbreviations.....	ix
Abstrak.....	xi
Abstract.....	xiii

CHAPTER 1: INTRODUCTION

1.0	Research Background	1
1.1	Impacts of Plastic Production	2
1.2	Plastic Recycling	3
1.3	Quality Enhancement of Recycled Plastics	5
1.4	Problem Statement	6
1.5	Research Objectives	8
1.6	Research Scope	8
1.7	Thesis Outlines	10

CHAPTER 2: LITERATURE REVIEW

2.0	Introduction	11
2.1	Plastic Overview	12
2.2	Classifications of Plastic Recycling	14
2.3	Constraints on the Use of Recycled Plastics	17
2.4	Improvement of Recycled Plastics	19
2.5	Reprocessing of Acrylonitrile Butadiene Styrene (ABS)	22
2.6	Plastic Processing Methods	26
2.6.1	Injection Moulding	28
2.6.2	Factors Affecting the Part Quality	30
2.7	Optimizing the Factors Affecting Part Quality	36
2.7.1	Taguchi Method	37

2.8	A Review on the Previous Works of Taguchi Method	40
2.8.1	Standalone Taguchi Method	42
2.8.2	Integration of Taguchi Method with Various Approaches	44
2.9	Findings from the Literature	59

CHAPTER 3: METHODOLOGY

3.0	Introduction	64
3.1	Feasibility Study	65
3.1.1	Part Description	67
3.1.2	Material	68
3.1.3	Experimental Procedures	69
3.1.4	Quality Testing	71
3.2	Computer-Aided Engineering (CAE) Simulation	74
3.2.1	Model Preparation	76
3.2.2	Material	78
3.2.3	Determination of Quality Characteristics	78
3.2.4	Selection of Processing Parameters	79
3.2.5	Selection of Orthogonal Array (OA)	81
3.2.6	Analysis of the Simulation Results	82
3.3	Processing Optimization	87
3.3.1	Determination of Quality Characteristics	89
3.3.2	Selection of Processing Parameters	89
3.3.3	Selection of Orthogonal Array (OA)	90
3.3.4	Quality Testing	91
3.3.5	Analysis of the Optimization Results	93
3.3.6	Confirmation Test	95
3.4	Summary	95

CHAPTER 4: RESULTS AND DISCUSSION

4.0	Introduction	97
4.1	Results of the Feasibility Study	98
4.1.1	Melt Flow Index (MFI)	98

4.1.2	Hoop Tensile Strength	99
4.1.3	Elongation at Break	101
4.1.4	Scanning Electron Microscopy (SEM)	103
4.2	Results of the Simulation Experiment	109
4.2.1	S/N Ratio and Data Normalization	111
4.2.2	Analysis of Variance (ANOVA)	112
4.3	Results of the Optimization Experiment	116
4.3.1	S/N Ratio and Data Normalization	117
4.3.2	Main Effects Analysis	119
4.3.3	Analysis of Variance (ANOVA)	124
4.3.4	Prediction of Performance at Optimal Process Conditions	128
4.3.5	Confirmation Test	129
4.4	Summary	131
CHAPTER 5: CONCLUSIONS AND FUTURE WORKS		
5.0	Introduction	133
5.1	Concluding Remarks	133
5.2	Recommendations for Future Work	137
REFERENCES		140
APPENDICES		
Table $F_{.01}(f_1, f_2)$, 99% Confidence		A-1
Calculations		A-3

LIST OF TABLES

		Page
Table 2.1	The advantages and limitations of plastic recycling processes	16
Table 3.1	General properties of ABS Novodur P2MT	68
Table 3.2	Blending composition of virgin and recycled ABS	69
Table 3.3	Material properties of ABS Novodur P2MT (LANXESS)	78
Table 3.4	Processing parameters and their levels in the simulation experiment	80
Table 3.5	Design of experiments using L_{27} orthogonal array	83
Table 3.6	Processing parameters and their levels in the optimization experiment	90
Table 3.7	Design of experiments using L_9 orthogonal array	91
Table 4.1	Simulation results for the 3D model	110
Table 4.2	S/N ratios and data normalization of simulation results	111
Table 4.3	ANOVA table for the simulation results before pooling	113
Table 4.4	ANOVA table for the simulation results after pooling	114
Table 4.5	Optimization experimental results for the ring stopper	117
Table 4.6	S/N ratios and data normalization of optimization results	118
Table 4.7	Main effects analysis for hoop tensile strength	120
Table 4.8	Main effects analysis for elongation at break	121
Table 4.9	Main effects analysis for shrinkage	122
Table 4.10	Main effects analysis for multiple quality characteristics	124
Table 4.11	ANOVA table for hoop tensile strength in optimization experiment	126
Table 4.12	ANOVA table for elongation at break in optimization experiment	127

Table 4.13	ANOVA table for shrinkage in optimization experiment	127
Table 4.14	ANOVA table for multiple quality characteristics in optimization experiment	128
Table 4.15	Results of confirmation test for multiple quality characteristics	130

LIST OF FIGURES

	Page
Figure 2.1 Ishikawa diagram of processing parameters in injection moulding	34
Figure 3.1 Flow chart of the feasibility study	66
Figure 3.2 The ring stopper	67
Figure 3.3 (a) Split disk test fixture; (b) Setup of test fixture	73
Figure 3.4 Flow chart of simulation experiment	75
Figure 3.5 3D model of the ring stopper	76
Figure 3.6 Mould design with four cavities of ring stopper	77
Figure 3.7 Simulation model with the runner system	77
Figure 3.8 Flow chart of processing optimization experiment	88
Figure 4.1 Relative MFI of recycled ABS as a function of the number of reprocessing cycles	98
Figure 4.2 Effect of reprocessing cycle on hoop tensile strength	100
Figure 4.3 Effect of reprocessing cycle on elongation at break	102
Figure 4.4 Micrograph of tensile fracture surface of: (a) virgin ABS; (b) 20% of recycled ABS subjected to 1 reprocessing cycle; (c); 20% of recycled ABS subjected to 5 reprocessing cycles; (d) 20% of recycled ABS subjected to 10 reprocessing cycles	104
Figure 4.5 Micrograph of tensile fracture surface of virgin ABS with: (a) 10% of recycled ABS; (b) 20% of recycled ABS; (c) 30% of recycled ABS; (d) 40% of recycled ABS; (e) 50% of recycled ABS	106
Figure 4.6 Main effects plot for hoop tensile strength	120
Figure 4.7 Main effects plot for elongation at break	121
Figure 4.8 Main effects plot for shrinkage	122
Figure 4.9 Main effects plot for multiple quality characteristics	124

LIST OF ABBREVIATIONS

ISO	International Standards Organization
ABS	Acrylonitrile Butadiene Styrene
CAE	Computer Aided Engineering
ANOVA	Analysis of Variance
MSW	Municipal Solid Waste
PET	Polyethylene terephthalate
PC	Polycarbonate
MFI	Melt Flow Index
CRTM	Compression Resin Transfer Moulding
CD	Compact Disc
N6	Nylon 6
N66	Nylon 6.6
MTN66	Moderately Toughened Nylon 6.6
RTN66	Rubber Toughened Nylon 6.6
CTE	Coefficient of Thermal Expansion
DOE	Design of Experiments
OA	Orthogonal Array
S/N	Signal-to-Noise
POM	Polyoxymethylene
PA66	Polyamide 66
HDPE	High Density Polyethylene
GPS	General Purpose Styrene

GRA	Grey Relational Analysis
LCD	Liquid Crystal Display
PP	Polypropylene
PS	Polystyrene
PCA	Principal Component Analysis
ANN	Artificial Neural Network
GRNN	Generalized Regression Neural Network
BPNN	Backpropagation Neural Network
RMSE	Root Mean Square Error
GA	Genetic Algorithms

**KAJIAN KEBOLEHLAKSANAAN AKRILONITRIL BUTADIENE STIRENA
(ABS) DIPROSES SEMULA DAN PENGOPTIMUMAN PARAMETER
SUNTIKAN ACUAN MELALUI KAEDAH TAGUCHI**

ABSTRAK

Sehingga kini, pengeluaran plastik di seluruh dunia dibebani dengan isu-isu alam sekitar dan pengurusan sisa pelupusan cekap sekeliling plastik sisa pasca industri. Usaha besar untuk memudahkan kitar semula plastik ke tahap yang lebih tinggi sementara mengikuti peraturan ISO 14001 yang ketat. Dalam kajian ini, kebolegunaan akrilonitril butadiene stirena (ABS) diproses semula sebagai ganti kepada ABS murni dikaji dengan menilai kesan komposisi campuran ABS dikitar semula dan pemprosesan semula pada sifat-sifat tegangan, rheologi dan morfologi. Satu keseimbangan yang baik antara ciri-ciri dan keupayaan pemprosesan plastik kitar semula amat diperlukan untuk memastikan pengeluaran yang berterusan. Pengoptimuman parameter suntikan acuan bagi penutup gelang ABS diproses semula melalui integrasi simulasi berangka dan kaedah Taguchi dibentangkan. Memandangkan fungsi dan penggunaan produk, kekuatan tegangan gelang, pemanjangan semasa patah dan pengecutan dipilih sebagai penilaian prestasi bagi penutup gelang. Dalam kajian ini, eksperimen simulasi awal telah dijalankan dengan menggunakan L_{27} susunan orthogonal untuk menyaring parameter bererti dan kombinasi optimum parameter pemprosesan ditentukan oleh penggunaan L_9 susunan orthogonal, analisis pengaruh utama dan analisis varians dalam eksperimen pengoptimuman. Seperti yang ditunjukkan dalam keputusan, kekuatan tegangan gelang, pemanjangan semasa patah dan pengecutan bagi penutup gelang ABS

diproses semula pada keadaan proses yang optimum dipertingkatkan sehingga ke tahap yang hampir dengan ABS murni. Penemuan eksperimen menandakan bahawa rekabentuk eksperimen berdasarkan integrasi simulasi berangka dan kaedah Taguchi mampu meningkatkan prestasi plastik diproses semula dengan bilangan minimum ujian eksperimen di mana masa persediaan yang singkat dan kos pemprosesan yang lebih rendah diperlukan.

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ABSTRACT

To date, plastic manufacturers all over the world are burdened with environmental and waste management issues revolving around the efficient disposal of the post-industrial plastic waste. Tremendous efforts are aimed at facilitating the plastic recycling to a greater extent in accordance to the strict regulation of ISO 14001. In this research, usability of reprocessed acrylonitrile butadiene styrene (ABS) as substitute for virgin ABS was investigated by evaluating the effect of blending compositions of recycled ABS and reprocessing on tensile, rheological and morphological properties. A good balance between properties and reprocess-ability of recycled plastic which ensures its reuse over long production runs is absolutely necessary. Optimization design of the injection moulding parameters for reprocessed ABS ring stopper via the integration of numerical simulation and Taguchi method is presented. Considering the functionality and application of the product, hoop tensile strength, elongation at break, and shrinkage are selected as the performance measure of the ring stopper. In this research, a preliminary simulation experiment was conducted with an L_{27} orthogonal array (OA) to screen the significant parameters and the optimal combination of the processing parameters is identified with the use of L_9 OA, main effects analysis and ANOVA in the optimization experiment. As results showed the hoop tensile strength, elongation at break, and shrinkage of the reprocessed ABS ring stopper at optimal process conditions were improved up to the

level close to that of virgin ABS. The experimental findings implied that an experimental design based on the integration of numerical simulation and Taguchi method is capable to enhance the performance of reprocessed plastics with a minimum number of experiment trials where shorter setup time and lower processing cost is needed.

CHAPTER 1

INTRODUCTION

1.0 Research Background

The term of plastic has attracted more attention in the literature for the past 100 years since the introduction of the first industrial plastic at the latter part of the 19th century. John Wesley Hyatt, an American, finally came upon the solution in year 1869 with celluloid which makes its debut in plastic industry (McCord, 1964). Ever since after, there have been several milestones in the history of material science as the invention of plastic has, arguably, touched more lives than any other technological breakthrough.

Plastics play a significant role across the environmental, societal, and economic dimensions of sustainable development. Our modern lifestyle would not be possible without plastics. Plastics have proliferated so readily throughout the modern world because of their inherent properties such as lightweight, versatility, and durability (Fortelný et. al., 2004). By possessing these advantageous characteristics, plastics has become a good candidate for replacement of other materials that range from simple plastic parts such as household storage containers, to sophisticated devices such as heart replacement valves. It is this range of properties together with their low cost that has driven the annual worldwide demand for plastics to reach at least 308 million tonnes by 2010 (Andrady and Neal, 2009).

1.1 Impacts of Plastic Production

Although plastics have had a remarkable impact on our culture and lifestyle, the production and use of plastics pose increasing threat to environment. Most plastics are made from fossil fuels and thus plastics production has an impact on oil consumption, both as a raw material and to deliver energy for the manufacturing process (van der Voet et al., 2003). The process takes a large number of decompositions and recombination that a commercial plastic is produced. According to Statistical Review of World Energy (2010), the total oil consumption of the world in 2008 was 81.73 million barrels per day and the global proved oil reserves at the end of 2008 are estimated to have been 1.258 trillion barrels. At the current rate of consumption, it would last 42 years before the oil reserves are completely depleted. It is estimated that 4% of the world's annual oil production is used as a feedstock for plastic production and an additional 3-4% during manufacture (Nemerow et al., 2009). Considering the massive amount of oil usage in the world even 7% is a very large quantity of oil to be used in the plastic production.

On the other hand, high plastic production inevitably leads to an increasing amount of plastics ending up in the waste stream which results in serious pollution problems. A significant portion of the waste generated in Malaysia comes from plastics. The amount of plastic waste continues to accumulate as the population is growing along with a great expansion of plastic industry in our country. Therefore, various options of plastic waste management, such as landfill, incineration, and recycling are introduced, in order to dispose the plastic waste (Lazarevic et al., 2010). Owing to the plastic waste is nonbiodegradable, the landfill disposal method is inefficient as the plastic waste takes long time to fully decompose and takes up a

large amount of valuable land filling space due to its low density. Another alternative for disposing plastic waste is incineration. The primary benefit of incineration is able to reduce the waste mass by 70% and volume by up to 90% (Lam et al., 2010). However, it is difficult to be implemented due to issues such as emission of gaseous pollutants (Khajuria et al., 2010). The dioxins produced from the incineration could cause acid rain which destroys vegetation, wildlife, rivers, soils, and even architecture. It could be harmful to the environment and human health. Thus, considering the drawbacks of landfill and incineration, plastic recycling is more favourable with the advantage in pollution abatement as well as efficient saving of fossil energy to produce virgin plastic resins.

1.2 Plastic Recycling

Plastic recycling seems to be the only alternative to handle the plastic waste effectively compared to the landfills and incineration waste disposal methods. Not to mention the indispensable role of plastic recycling in solving the petroleum depletion issue in which petroleum is one of the nonrenewable raw materials to produce plastic parts but also is a finite resource that will someday be depleted. Recycling of plastic is gaining significance in order to reduce the consumption of petroleum and conserve the limited resource. In addition, the potential economic benefits of using recycled plastic attracted the interest as 70% less energy is consumed to recycle plastic products compared to the production of virgin resins (Santos and Pezzin, 2003). When all these factors are taken into account, the overall impact of waste plastic recycling would be significant.

To date, plastic manufacturers all over the world are burdened with environmental and waste management issues revolving around the efficient disposal of the post-industrial plastic waste such as product rejects, plastic scrap, sprues and runners. Obviously, plastic waste creates a significant problem to the manufacturers as the manufacturers need to comply with the environmental regulations to prevent pollution. Manufacturers have to shift to green production where efforts are aimed at facilitating the plastic recycling to a greater extent in accordance to the ISO 14001. Thus, plastic recycling has drawn attention along with growing environmental concerns, as well as the potential of economic benefits by establishing a waste reduction and recycling policy.

Unfortunately, large scale plastic recycling is still hindered by a wide range of barriers in the industry. One of the problems is recycled plastics has been doubted to be a reliable replacement of virgin plastics. This is due to the belief that changes in mechanical and rheological properties have created a perception that recycled plastics have a low economical value. Hence, the manufacturers avoid using recycled materials that they believe would affect the product quality (Balart et al., 2005). In fact, the properties of plastics tend to decrease after several injection cycles according to the nature of the polymer subjected to multiple reprocessing. But still, improvements could be made from both mechanical and material perspectives concurrently to find the optimal point in producing recycled plastic products with satisfactory quality.

1.3 Quality Enhancement of Recycled Plastics

A good balance between properties and reprocessability of recycled plastic which ensures its reuse over long production runs is absolutely necessary. This subject has been the focus of many papers and part of the literature on recycled plastic especially for the multiple reprocessing is of academic rather than of commercial interest. Various approaches are proposed by the researchers to improve performance and properties of recycled plastic from the material perspective, such as blending with other plastics and the addition of filler and stabilizers (Vilaplana and Karlsson, 2008). Nevertheless, high processing costs has become an obstacle for implementing the aforementioned approaches from a laboratory to an industrial scale compared to the price of using virgin resin. As a consequence, an interest existed in the industrial sector for looking into the possibility of optimizing the injection moulding process from the manufacturing perspective. It could be an appealing approach to improve the recycled plastic products considering no extra processing cost is required.

It is vital to understand the basic mechanisms of the injection moulding process prior the process optimization. The quality of the plastic products, either it is produced of virgin material or recycled material, depends on a large number of process variables. Factors that influencing the part quality can be classified into four categories: part design, mould design, machine performance, and processing conditions (Bharti and Khan, 2010). In a currently running production process, the part design, mould design, and machine performance are commonly assumed as established and fixed. Any modification made would lead to higher cost of production. Optimization of processing parameters can be regarded as a virtue

approach to offer significant property improvements to the recycled plastic products without making any major alteration in the production process.

The settings of processing parameters in injection moulding process can be categorized into four groups: temperature, pressure, time, and distance. Each parameters adjustment will have either a positive or a negative effect on the physical and aesthetic properties of the moulded product. Nevertheless, the interaction effect between the processing parameters cannot be neglected because all processing parameters are mutually dependent and changing one will affect the others. Therefore, with the prior knowledge of the relationship between processing parameters and the part properties, the processing parameters can be manipulated in order to improve the quality of the recycled plastics to a great extent.

1.4 Problem Statement

A diligent effort is needed to prove the recycled plastics subjected to multiple reprocessing cycles to be a continuous and reliable replacement for virgin plastics. Polymers are exposed to thermo-mechanical degradation during reprocessing (Żenkiewicz et al., 2009). That is the reason why recycled plastics will not exhibit similar mechanical and rheological properties as they are in virgin form. The mechanical and rheological properties of polymers are changed and the functional quality of reprocessed polymers is deteriorated (Vilaplana and Karlsson, 2008). Nevertheless, improvement can be done via the blending technique incorporated with the optimization of processing parameters in producing recycled plastics to upgrade the properties of recycled plastics considering both mechanical and material viewpoint.

Owing to the recyclates degrade to a varying extent depending on the number of reprocessing cycles, blending recycled plastics with virgin materials is one of the most common and straightforward procedures for upgrading the properties of recycled plastic products. Various compositions of recyclates mixed with the virgin resins have diverse effect in processing behaviour, mechanical, and rheological performance of the blends. The main problem related to the blending approach is determining the appropriate amount of recycled polymers to be mixed with the virgin resins, in order to obtain the optimal blending composition to produce recycled plastic products that do not show significant variation from the virgin ones.

Determining the optimal process parameter settings critically influence productivity, quality, and cost of production in the plastic injection moulding industry. However, it raises another question of how the optimization of processing parameters could be done effectively. Previously, the setting of the injection moulding process parameters relies heavily on the experience and intuition of experts and involves a trial-and-error process (Shi et al., 2003). This tuning exercise is repeated until the quality of the moulded parts is found satisfactory. However, trial-and-error method is time consuming and is difficult to obtain an optimal parameter setting for injection moulding process which has numerous processing parameters to be controlled (Shie, 2008). Thus, one of the main aspects of this work is to propose an effective approach for the optimization of the processing parameters with the aim of enhancing the product quality of recycled plastics.

1.5 Research Objectives

The objectives of this thesis can be enumerated as follows:

1. To study the relationship of the blending composition of virgin and recycled plastic resins towards the mechanical and material perspectives and to obtain the optimal blending composition to produce acceptable product quality with lower raw material cost.
2. To analyze the changes of mechanical, rheological and morphological properties of recycled plastic due to thermo-mechanical degradation during multiple reprocessing and to determine the appropriate reprocessing cycles to manufacture products with satisfactory quality.
3. To study the effectiveness and importance of the integrated approach using the Taguchi method and numerical simulation in determining the significant processing parameters out of the numerous injection moulding process variables.
4. To improve the tensile properties and dimensional stability of recycled plastic products by optimizing the processing parameters via Taguchi method.

1.6 Research Scope

Plastic wastes can be categorized into two types based on disposal classification: post-consumer products and post-industrial scrap. In this research, post-industrial scrap is given the momentous priority due to its viability to be widely used in the industry. It makes great economic sense as both plastic waste and raw material cost are reduced by feeding the granulated plastic waste directly back into the moulding machine as a replacement of virgin resin. Furthermore, the post

industrial scrap comprises clean plastic waste with similar type of material from the production process and thus the incompatibility issue will never exist.

This study is restricted to the evaluation of thermoplastics due to the infeasibility of thermosets in recycling process. Out of all types of thermoplastics, acrylonitrile butadiene styrene (ABS) is utilized for its property-price profile wherein both properties and price are intermediate between the lower priced commodity thermoplastics and the more expensive high performance engineering plastics. ABS is one of the most frequently used polymers in various applications such as the automotive industry, the electrical and electronic equipment, communication instruments, and other commodities. The widespread applications of ABS make them to be an interesting plastic material for recycling.

The research work was collaborated with a company that produces plastic products in Penang. This research is target towards implementing closed-loop recycling in the production to effectively handle the post-industrial plastic waste. It was performed in industrial settings to yield recycled plastic products with satisfactory quality which are commercially viable in the market. Nevertheless, there are some limitations in this research where no modification is allowed to be done on the designs of product and mould as it will affect the productivity.

Aiming at enhancing the part quality for recycled plastics, the highlight in this research is to investigate the effectiveness of Taguchi method as a design of experiment to optimize the processing conditions in injection moulding. Computer-aided engineering (CAE) simulation was integrated with Taguchi method to make better analysis in regards to parameters selection. The hoop tensile properties and

shrinkage will represent the performance measures for the virgin and recycled ABS blend.

1.7 Thesis Outlines

The thesis is presented in the following manner: Chapter 1 of this thesis introduces the research and covers the rest of works in context. The background of plastics and plastic production is introduced and the problems associated with it are explained. A solution has been sought by introducing plastic recycling. The problem statements and the objectives are also stated in this chapter. The literature review of the related subjects has been outlined in Chapter 2. The main objectives of this chapter are to present the latest knowledge and studies from other researchers relating to the feasibility of performing reprocessing operations in the plastic industry particularly in recycling of plastic and the implementation of Taguchi method in optimizing the injection moulding processing parameters to enhance the mechanical properties of the recycled plastic products. Chapter 3 delineates the methodology used in this research wherein every procedure of the experiments is explained in detail. Chapter 4 analyzes and discusses the results obtained from the experiments based on the Taguchi method and analysis of variance (ANOVA). Chapter 5 highlights the conclusion on this research work and ends with the future work continuation.

CHAPTER 2

LITERATURE REVIEW

2.0 Introduction

This chapter starts with a brief discussion of plastic using the classification based on the basis of its properties and its processibility. The following section explores the feasibility of four plastic recycling methods to overcome the environmental threat caused by plastic wastes. The constraints of the usage of recycled plastics are highlighted, followed by a discussion on improving the properties of recycled plastics. An introduction of various plastic processing techniques is presented and in this research injection moulding will be the focus of discussion due to its wide-range applications in the engineering field. The effects of material selection, part and mould designs and processing parameters are analyzed in detail to investigate their significance in affecting the occurrence of defects and quality of final products. Previous works relating with standalone Taguchi method and integration of Taguchi method with various approaches including numerical simulation, grey relational analysis, artificial neural network and genetic algorithm will be reviewed thoroughly in the study. Finally, the findings from the literature review will be discussed at the end of this chapter.

2.1 Plastic Overview

The term “plastics” encompasses materials composed of various elements such as carbon, hydrogen, oxygen, nitrogen, chlorine, and sulphur. Typically, plastics are produced by the conversion of natural products or by the synthesis from primary chemicals generally coming from oil and natural gas. Large molecules of hydrocarbon are heated under controlled conditions and broken down into monomers. These monomers are chemically bonded into chains called polymers. The structure of the polymers from different combinations of monomers can yield different properties and characteristics, such as strength and moulding capability (Halden, 2010). The versatility and variability of plastics allow them to be tailored to meet very specific technical requirements best suited for particular applications.

Plastics have entrenched themselves within the spectrum of modern materials. Approximately 16% of applications are designed with plastics and cannot be feasibly substituted (Pilz et al., 2010). It would be hard to imagine the world without plastics in view of the fact that plastic products have infiltrated every corner of our daily life, from the morning toothbrush to the garbage bag that is carried out at the end of the day (Spokas, 2008). With the intrinsic properties including lightweight, durable, strong and cost competitive, plastics are very attractive to be used by the consumers as substitutes for natural materials such as paper, metals, wood and glass (Andrady and Neal, 2009). In relation to the divergent properties and versatility exhibited by plastics, plastics can be used in many applications such as food packaging, clothes, shelter, communication, transportation, construction, health care and leisure industries (Sudesh and Iwata, 2008).

Nowadays, plastic products from all around the world are in high demand and continue to grow at a very fast pace. The popularity of plastic products has spurred a spectacular increase in the world's annual production of plastic materials from 1.5 million tonnes in 1950 to 230 million tonnes in 2009 (PlasticsEurope, 2010). As with most materials, the annual growth of plastics was hit hard by the economic crisis where global plastics production has fallen from 245 million tonnes in 2008 to 230 million tonnes in 2009 (PlasticsEurope, 2010). However, taking into account the trend of increased consumption of plastic in the world it can be expected that the plastic manufacturing industry will recover from the economic slowdown to reach its peak in several years. Unfortunately, high plastic consumption inexorably leads to the increasing amount of plastic ending up in the waste stream. Approximately 30 million tonnes of plastic waste were generated in the United States in 2009, representing 12.3 percent of total municipal solid waste (MSW) (United States Environmental Protection Agency, 2009). In relation to the population growth along with a great influx of foreign workforce to cities, the problem tends to be more severe in developing countries, such as Malaysia. According to the Ninth Malaysia Plan, plastic materials constitute the second largest component of MSW next to food waste, which accounts for 24% of total MSW (Manaf et al., 2009). The existence of plastics though makes our lives smooth and comfortable, but it is essential to consider the problem of plastic waste management.

Plastics have become a major threat due to their nonbiodegradability and high visibility in the waste stream. Plastic waste can take decades and probably centuries to decompose. In Malaysia, the most common and easiest method to handle a significant portion of plastic waste is landfill. Nevertheless, with available landfill

capacity decreasing and public opposition to authorities for making new facilities, many of the landfills in Malaysia are reaching their critical levels. There is an urgent need for a sustainable plastic waste management strategy to manage the huge amount of plastic waste generated. Hence, recycling is the best alternative and should be introduced into the plastic waste management concept to manage the plastic wastes effectively.

2.2 Classifications of Plastic Recycling Process

Plastic recycling is viewed as one of the most important actions currently available to counteract the issue of excessive plastic waste and solve the high demand for virgin resins. According to the Malaysian Plastics Forum (2007), plastic wastes are categorized as post-consumer wastes and post-industrial wastes. Post-consumer wastes are referred to the discarded plastic materials generated from residential and consumer waste after their lifecycles are over. Typically, the post-consumer wastes consist of a mixture of plastics which are usually contaminated. Recycling of post-consumer plastic is complicated due to the necessity of materials separation as some studies showed that up to 10.5% foreign materials contained in the post-consumer wastes (Navarro et al., 2008). On the other hand, post-industrial wastes are defined as the process scrap parts such as sprues, runners and defective parts that are not contributing into production. In view of the fact that the post-industrial wastes are relatively clean and segregated by the type of plastic, they are currently recycled more than post-consumer wastes as they do not face the polymer incompatibility issue. Therefore, attention should be focused on the homogeneous

waste streams such as post-industrial wastes to maximise the potential of plastic recycling.

In principle, recycling processes for plastic wastes are classified into four categories: primary, secondary, tertiary, and quaternary (Curlee, 1987):

Primary Recycling

Primary recycling is often referred to as closed-loop recycling in which polymer from a single product or product type is collected and reprocessed into either its original use or comparable products with equivalent properties. The plastic wastes have to be segregated based on their types prior to reprocessing as the primary recycling is limited to clean resins.

Secondary Recycling

Secondary recycling is an open loop recycling in which it allows for a higher mixture of different kinds of plastic wastes. Generally, most of the post-consumer plastic wastes are unsorted and they contain significant level of impurities and contaminants (Mancini et al., 2010). Typically, secondary recycling is known as a downgrading recycling process as the reprocessing of mixed contaminated plastics results in inferior mechanical properties compared with those produced from virgin polymers due to the incompatibility of most polymer pairs (Tselios et al., 1998; Qin et al., 2008).

Tertiary Recycling

Tertiary recycling, which is known as chemical or feedstock recycling, degrades the polymers to its monomers or other low molecular weight products, usually liquids or gases, which are suitable for use as a feedstock in refinery (Ali and Siddiqui, 2005).

Nevertheless, tertiary recycling is more costly than other recycling approaches as it involves depolymerization and repolymerization reactions (Hopewell et al., 2009). From the industry's perspective, the tertiary recycling is not viable economic approach and cannot be commercialized.

Quaternary Recycling

Quaternary recycling is energy recovery which generates heat and electricity by the incineration of plastic waste (Karlsson, 2004). This approach would be considered as the last option of plastic waste treatment due to the emission of various gaseous pollutants such as polychlorinated dibenzo-p-dioxins and dibenzofurans, which are detrimental both to human health and environment (Lisk, 1988).

For better qualitative comparison, the advantages and limitations of different recycling approaches are listed in Table 2.1.

Table 2.1: The advantages and limitations of plastic recycling processes

Type of recycling process	Advantages	Limitations
Primary recycling	Recovers the recycled plastics to products having equivalent quality to the virgin resins.	Limited to uncontaminated and clean resins.
Secondary recycling	Allows for high level of mixture of plastic wastes.	Inferior properties due to impurities.
Tertiary recycling	High product yield and minimum waste.	Costly due to complicated chemical reactions.
Quaternary recycling	Results in a volume reduction of 90–99%.	Insufficient use of resources and dreadful gas emissions.

(Source: Aurrekoetxea et al., 2001; Mihut et al., 2001; Al-Salem et al., 2009)

2.3 Constraints on the Use of Recycled Plastics

Large-scale plastic recycling can be carried out on many products, ranging from consumer commodities such as soft drink bottles (Chilton et al., 2010) to industry applications such as automobile components (BMW Group Recycling, 2003). Nevertheless, plastic recycling is still hindered by a wide range of barriers, including restrictions imposed by standards and specifications, as well as the mistrust on the quality assurance of recycled plastics. Most of the plastic manufacturers and consumers are reluctant to use recycled plastics due to the perception that recycled plastics are inferior to virgin resin and the usage of recyclates will result in mediocre performance in their products (Butler, 1991).

The presence of impurities in recycled plastics is a significant aspect limiting the feasibility of recycling. Polymeric materials may be contaminated with impurities during recycling operations due to their permeable nature to absorb the low molecular weight compounds (Karlsson, 2004). The impurities encompass various forms such as carbon residue, dirt, printing inks, product label papers, metals, additives, as well as mixed polymeric fractions. Polymeric impurities and nonpolymeric impurities act as stress risers and affect the performance of the recycled resins (Howell, 1992). According to Liang and Gupta (2000), an addition of only 1% of polymeric impurity would be enough to significantly decrease several mechanical properties. The existence of different types of thermoplastics is also treated as polymeric impurities because they tend to be incompatible with each other and hardly form one-phase system. The mixed polymeric fractions are mutually insoluble and structurally heterogeneous (Selke, 1988). The incompatible blend will

lead to poor adhesion properties in the polymeric mixture interface and hence deterioration in overall macroscopic properties (Mathew and Thomas, 2003).

Recycled products of primary recycling possess the same functional properties as the virgin material as the recyclates are homogeneous and contaminant-free. Nevertheless, the recyclates still undergo the thermo-mechanical degradation when they are subjected to multiple reprocessing. During multiple reprocessing, the polymeric materials are submitted to successive cycles of high temperature and intensive shear stress, leading to deterioration in their physical properties and functional quality (Perez et al., 2010). The thermo-mechanical degradation of plastic materials can be regarded as the simultaneous occurrence of two main mechanisms during multiple reprocessing: thermo-oxidative and mechano-oxidative (Strömberg and Karlsson, 2009).

The thermal degradation takes place when the polymer is subjected to high temperatures in an oxygen-ubiquitous atmosphere during melting and reprocessing operations, whereas the mechanical degradation occurs due to the shear forces applied during the grinding and processing. The thermo-mechanical degradation induces chain scissions of the polymeric chains due to the effect of radical chain reaction, causing a series of modifications in the structural and macroscopic properties of the polymers (Karahaliou and Tarantili, 2009). As a result of chain scissions, short-chained molecules tend to reduce the molecular weight of the polymeric chains and lead to a decrease in viscosity. The number of reprocessing operations has significant effect on the extent of degradation, influencing the mechanical and rheological properties of recycled products (Lozano-González et al., 2000; Russo et al., 2007). Therefore, plastic waste cannot be recycled endlessly as it

undergoes degradation due to repeated processing and this limits the number of times plastic can be recycled.

The quality of the plastic wastes intended for reprocessing is determined by the history of the recyclate synthesis. The previous use and the number of reprocessing have significant effect on the mechanical and rheological properties of recyclates. Many studies have been carried out in overcoming those impediments to improve the recycled plastics to a comparable standard with the virgin materials. Most of the works emphasised the development of a suitable technology for the recycling and improvement of the properties of the recyclates.

2.4 Improvement of Recycled Plastics

High market acceptance of recyclates is a decisive condition for continuing development of recycling industries. The reason which precludes the recyclates to be commercially viable is because of their inferior mechanical properties and unpredictable rheological properties due to the presence of impurities and thermo-mechanical degradation during reprocessing. Therefore, improvement of the material and mechanical properties of recyclates is essential to maximize the potential of recyclates in high-value applications. To transform the recyclates into a reliable resource for plastic products, two strategies for effective recyclates improvement are introduced.

In the first strategy, additives including stabilizers, impact modifiers, fillers, compatibilizers and reactive repair molecules are added to recycled resins and each type of additive demonstrates different functional use in enhancing the properties of recyclates (La Mantia, 2002). The stabilizers do not effectively recover the recyclates

but they are used to protect the recyclates from thermo-mechanical degradation during reprocessing and to extend the material's longevity (Pospíšil et al., 1995). The introduction of impact modifiers can significantly improve the elongation at break and impact strength of recyclates. On the other hand, the modulus, impact, and tensile strength of recyclates can be reinforced by the use of fillers. However, there is a critical weakness of fillers in which the processability and elongation at break are simultaneously decreased with the filler reinforcement. The inclusion of compatibilizers commonly bases their function on physical or chemical effects; reactive compatibilizers introduce reactive moieties that form covalent bonds to link the blending polymers which are chemically identical, whereas nonreactive compatibilizers improve the interfacial adhesion due to good miscibility with both polymers of the blend (Karlsson, 2004). The use of reactive repair molecules, such as radical generators or compounds with reactive functional groups, is to increase the molecular weight of the polymeric chain and improve the mechanical and rheological properties of recyclates (Vilaplana and Karlsson, 2008). The chemical structure and molecular architecture of the recyclates can be rebuilt by inducing branching or cross-linking reaction to extend the polymeric chains (Scaffaro et al., 2007).

The other strategy to help maintain the properties of the final product is to blend optimal quantity of recycled plastics with the virgin plastics. Thus, increasing the percentage of recycled material in the blend without significantly affecting its performance is one of the main issues in recycling plastic. Even though the virgin and recycled polymers are of similar polymer type, they may have different molecular weight distribution, molecular structures (branching, cross-linking, and

presence of oxidative moieties), degree of degradation and semicrystalline morphology (Vilaplana and Karlsson, 2008).

Several studies on the influence of recyclates content on the quality characteristics of the blend show different views. Phuong et al. (2008) suggested only small and limited amount of recyclates (under 5%) can be blended with virgin resins in order to achieve comparable properties as virgin plastics. According to Llorin et al. (2003), majority of the PET manufacturers recycles about 10% of their industrial waste. On the other hand, Fermeglia et al. (2006) pointed out that less than 15% of the post-industrial rejects can be added to the virgin materials to produce good mouldings.

The argument regarding the appropriate amount of recyclates to be mixed with virgin resins was concluded by the study of Liang and Gupta (2001) in which up to 15% recycled polymer can be added to the virgin polymer without significantly altering processing and mechanical properties of the virgin resin when purity level is 99%. Nevertheless, this concept is only valid for recycled PC/virgin PC and recycled ABS/virgin ABS. Furthermore, Liang and Gupta (2002) extended their works in the copolymer PC/ABS blends by increasing recycled PC content and found that a PC/ABS blend comprising 40% recycled PC, 10% low molecular weight virgin PC, and 50% virgin ABS is able to produce acceptable quality of processing and mechanical properties. In the study of Al-Mulla and Gupta (2006), they discovered that up to 25% of recycled PC can be added to virgin PC without significantly altering its mechanical properties by adding 15% of short glass fibers. These significant findings showed that the properties of recyclates can be upgraded by an

appropriate blending with virgin resins to mask the batch-to-batch property variations of the recyclates.

The different functionalities of additives to tailor the properties of recyclates allow for more versatile uses with many applications. Nevertheless, the cost of the additives is a major consideration of the plastic manufacturers because they need to bear an extra manufacturing cost regardless of the effectiveness of additives in improving the recyclates. On the contrary, the blending approach offers a feasible alternative to recycling industry for improving the properties of recycled plastics with nearly no extra processing cost required. With the effective strategies to improve the properties of recyclates, plastic recycling can be performed to any great extent. As acrylonitrile butadiene styrene (ABS) is the main focus of this research, a review done on the degradation of ABS materials subjected to multiple reprocessing will be further explored in the next section.

2.5 Reprocessing of Acrylonitrile Butadiene Styrene (ABS)

The acrylonitrile butadiene styrene (ABS) terpolymer is a widely used engineering thermoplastic based on three monomers: acrylonitrile, butadiene, and styrene. A typical ABS usually consists of 15 to 35% acrylonitrile, 5 to 30% butadiene, and 40 to 60% styrene, but these proportions may vary in a relatively wide range (Żenkiewicz et al., 2009). Therefore, the ABS three monomers system can be tailored to yield desired properties by adjusting the composition of each monomer, given that acrylonitrile imparts chemical resistance and hardness, butadiene enhances impact strength, and styrene contributes ease of processing characteristics (Giles et al., 2005). ABS is utilized for its property-price profile wherein both properties and

price are intermediate between the lower priced commodity thermoplastics and the more expensive high performance engineering plastics (Karahaliou and Tarantili, 2009). For that reason, ABS has a very wide range of applications including electrical and electronic equipment, automobiles, communication instruments, and other commodities (Boronat et al., 2009).

The consumption and disposal rate of ABS in plastic manufacturing companies is increasing at a frenetic pace in Malaysia. According to Wahab et al. (2007), the ABS resins achieve the highest consumption rate of 28%. Their studies involved 20 plastic manufacturers registered with the Malaysian Plastics Manufacturers Association; most of the survey respondents were from the electrical and electronic sector. Electrical and electronic sector is the second largest industrial segment for plastic products next to packaging sector in Malaysia (Lim, 2010). Statistics showed that ABS makes up a significant proportion of the plastic fraction in electrical and electronic equipment, consisting of 26% of ABS alone and 16% of polymeric blends comprising ABS (American Plastic Council, 1999). This is particularly imperative when using ABS for producing recycled plastic products especially multiple processing due to the high resin price of ABS which costs between RM 5400 and 6600 for 1 metric tonne (Malaysian Petrochemical Association, 2006).

Extensive scientific literatures show that different types of degradation reactions may occur when the polymeric materials are subjected to successive high temperatures or shear cycles during reprocessing (Sarrionandia et al., 2009). Nevertheless, the processing conditions and processing cycles are found to have significant influence on the properties of reprocessed ABS. Bai et al. (2007)

evaluated the recyclability of ABS plastics from waste computer housings under various reprocessing conditions of temperature and shear rate. They also studied the effect of multiple reprocessing. They found that, within the range of reprocessing parameters studied, temperature had a more significant effect than shear rate on mechanical properties. On the other hand, the modulus of elasticity did not change significantly with increasing number of reprocessing cycle, but tensile strength increased slightly. In another study, the ABS resins with different viscosity grades were subjected to two consecutive cycles of moulding and it was found that the ABS materials with low viscosity grade showed polymer degradation upon increasing the number of processing cycles whereas the ABS materials with high viscosity grade showed an increase of melt viscosity as the number of injection moulding cycles is increased, indicating a rheological upgrade (Boronat et al., 2009).

All the previous works used 100% recycled ABS and all of them are subjected to not more than four times reprocessing cycles. The results showed insignificant variation of mechanical and rheological properties. However, the limited information obtained on a few reprocessing cycles is insufficient for the plastic manufacturers and consumers to look into the feasibility of implementing closed-loop production over long production runs. Therefore, further investigations were carried out on ABS recyclates subjected to more reprocessing operations.

In the research by Karahaliou and Tarantili (2009), the ABS material was subjected to five subsequent cycles in a twin screw extruder. The results showed that repeated processing does not essentially affect either the melt flow index or the tensile strength of ABS. However, the ductility value of reprocessed ABS fluctuated with increasing number of reprocessing cycle where an increase of elongation at