



**CLAY REINFORCED RECYCLED BIAXIALLY-ORIENTED
POLYPROPYLENE COMPOSITES THROUGH WATER-
ASSISTED MELT COMPOUNDING FOR CONCRETE FINE
AGGREGATES**



MASTER OF SCIENCE IN MANUFACTURING ENGINEERING

2024



**Faculty of Industrial and Manufacturing Technology and
Engineering**



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Anis Aqilah binti Abd Ghani

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DEDICATION

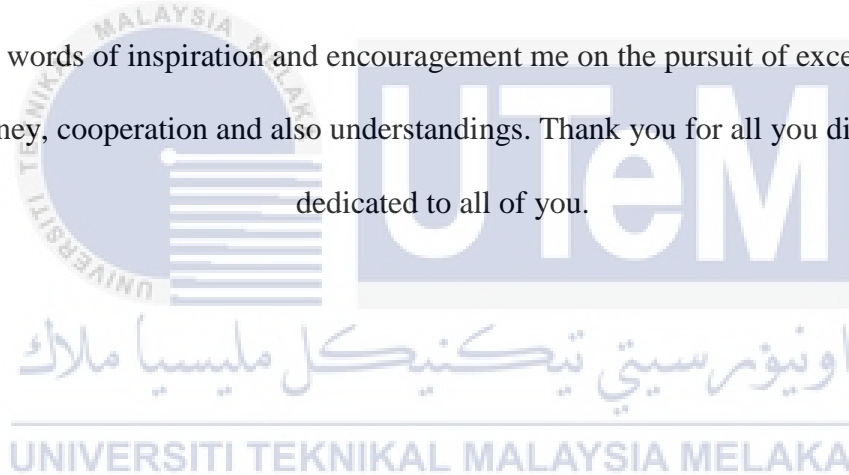
To my beloved husband, Ammar Fakhrullah bin Arifin

My caring mother, Noorfizam binti Omar

My appreciated father, Abd Ghani bin Sha'ari

My adored sister, Intan Hanani binti Abd Ghani

For giving words of inspiration and encouragement me on the pursuit of excellence, moral support, money, cooperation and also understandings. Thank you for all you did. This work is dedicated to all of you.



ABSTRACT

Natural aggregate depletion has becoming a global problem despite increased structural construction demands. Besides, accumulating plastic waste is a big challenge people face worldwide. Transforming waste plastics into construction aggregates appeared to be a sensible solution to both problems. However, 1) the low interaction between plastic aggregates with organic cement lowers the strength of concretes, and 2) the high temperature of the conventional melt compounding process of plastic aggregates becomes the ultimate concern that needs to be addressed further. This study used recycled biaxially-oriented polypropylene (rBOPP) waste provided by San Miguel Plastic Film Sdn. Bhd. In Stage 1, the rBOPP was compounded with kaolin clay using a water-assisted compounding process. The process parameters (temperature: 130 to 180 °C; time: 5 to 10 minutes) and formulation (% clay: 0 to 10 wt%; % water: 0 to 10 wt%) were optimized with the help of the Response Surface Methodology (RSM) using a two-level factorial design. The optimum parameters to produce plastic composite aggregates (PCA) were 1 wt% kaolin clay, 10 wt% water at a temperature of 180°C and a time of 5 minutes with a tensile strength of ~32MPa. Then, the PCA and rBOPP without clay (PWA) were bulk produced in a plastic factory and further validated for physical and mechanical properties. The PCA and PWA were tested for physical and mechanical properties per standards ASTM D792, ASTM D1895, ASTM D2240 and ASTM D638. The PCA had enhanced tensile strength and tensile modulus with an increment of 1.2 and 8 % compared to PWA. The properties were supported with morphological analysis through scanning electron microscopy (SEM), compositional analysis through Fourier-transform infrared spectroscopy (FTIR) and structural analysis through X-ray diffractometry (XRD). In Stage 2, the optimum formulation of PCA at different ratios (10 wt%, 15 wt% and 20 wt%) was tested for the workability and compressive strength of M20 concrete mixtures against the natural aggregates (NA) and PWA. The M20 mixed concretes were produced by hand and tested using a slump test and compression test according to the BS standard of EN 2390-3: 2019 and BS EN 12390: 3: 2000. It was found that PCA concrete using 10 wt% showed a slump value of 27 mm and compressive strength of 29 MPa. The data were supported by morphological characteristics and stability of concrete structures through camera images, optical microscopy (OM) and SEM. The optimum amount of PCA is proven to produce concrete with good workability and significant compressive strength. It is also proven that PCA aggregates with clay particles can strengthen the concrete by 30% compared to PWA. The finding of this study is an alternative to solve both issues of natural aggregate depletion and plastic pollution. It benefits construction and plastic manufacturers to adopt green materials and greener waste management.

KOMPOSIT POLIPROPILENA BERORIENTASI-DWIPAKSI BERTETULANG TANAH LIAT DIKITAR SEMULA MELALUI PENYEBATIAN LEBUR TERBANTU AIR UNTUK AGREGAT HALUS KONKRIT

ABSTRAK

Penyusutan agregat semulajadi menjadi masalah global walaupun permintaan pembinaan struktur meningkat. Selain itu, pengumpulan sisa plastik merupakan cabaran besar yang dihadapi oleh manusia di seluruh dunia. Mengubah sisa plastik kepada agregat pembinaan nampaknya merupakan penyelesaian yang wajar untuk kedua-dua masalah. Walau bagaimanapun, 1) interaksi rendah antara agregat plastik dengan simen organik merendahkan kekuatan konkrit dan 2) suhu tinggi proses pengkompaunan cair konvensional agregat plastik menjadi kebimbangan utama yang perlu ditangani lebih lanjut lagi. Kajian ini menggunakan bahan buangan polipropilena berorientasi-dwipaksi kitar semula (rBOPP) yang disediakan oleh San Miguel Plastic Film Sdn. Bhd. Pada Peringkat 1, rBOPP telah disebatikan dengan tanah liat kaolin menggunakan proses pengkompaunan berbantuan air. Parameter proses (suhu: 130 hingga 180 °C; masa: 5 hingga 10 minit) dan perumusan (% tanah liat: 0 hingga 10 % berat; % air: 0 hingga 10 % berat) telah dioptimumkan dengan bantuan Metodologi Permukaan Sambutan (RSM) menggunakan reka bentuk faktorial dua peringkat. Parameter optimum untuk menghasilkan agregat komposit plastik (PCA) ialah 1% berat tanah liat kaolin, 10% berat air pada suhu 180 °C dan masa 5 minit dengan kekuatan tegangan ~ 32 MPa. Kemudian, PCA dan rBOPP tanpa tanah liat (PWA) dihasilkan secara pukal di kilang plastik dan seterusnya disahkan untuk sifat fizikal dan mekanikal. PCA dan PWA telah diuji untuk sifat fizikal dan mekanikal mengikut piawaian ASTM D792, ASTM D1895, ASTM D2240 dan ASTM D638. PCA telah meningkatkan kekuatan tegangan dan modulus tegangan dengan peningkatan sebanyak 1.2 dan 8 % berbanding PWA. Ciri-ciri tersebut disokong dengan analisis morfologi melalui pengimbasan mikroskop elektron (SEM), analisis komposisi melalui fourier mengubah inframerah spektroskopi (FTIR) dan analisis struktur melalui difraktometri sinar-X (XRD). Pada peringkat 2, formulasi optimum PCA pada nisbah yang berbeza (10% berat, 15% berat dan 20% berat) telah diuji untuk keboleherjaan dan kekuatan mampatan campuran konkrit M20 terhadap agregat semulajadi (NA) dan PWA. Konkrit campuran M20 dihasilkan dengan tangan dan diuji menggunakan ujian jatuhan dan ujian mampatan mengikut piawaian BS EN 2390-3: 2019 dan BS EN 12390: 3: 2000. Didapati konkrit PCA menggunakan 10% berat menunjukkan nilai kemerosotan 27 mm dan kekuatan mampatan 29 MPa. Data tersebut disokong oleh ciri morfologi dan kestabilan struktur konkrit melalui imej kamera mikroskop cahaya (OM) dan SEM. Jumlah optimum PCA terbukti menghasilkan konkrit dengan keboleherjaan yang baik dan kekuatan mampatan yang ketara. Ia membuktikan bahawa agregat PCA dengan zarah tanah liat boleh mengukuhkan konkrit sebanyak kenaikan 30% berbanding dengan PWA. Dapatan kajian ini merupakan alternatif untuk menyelesaikan kedua-dua isu penyusutan agregat semulajadi dan pencemaran plastik. Ia memberi manfaat kepada pengeluar pembinaan dan plastik untuk menggunakan bahan hijau dan pengurusan sisa yang lebih hijau.

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LIST OF SYMBOLS

| | | |
|-------------|---|-----------------------------|
| $CuK\alpha$ | - | Copper K-alpha Radiation |
| E | - | Young's Modulus |
| F_c | - | Compressive Strength |
| k | - | Conductivity |
| T_c | - | Crystallization Temperature |
| w/c | - | Water-to Cement Ratio |



LIST OF ABBREVIATIONS

| | | |
|-------|---|--|
| ABS | - | Acrylonitrile Butadiene Styrene |
| A/C | - | Aggregate-to-Cement |
| ANOVA | - | Analysis of Variance |
| ASTM | - | American Society for Testing and Materials |
| BOPP | - | Biaxially-Oriented Polypropylene |
| BS | - | British Standard |
| CA | - | Coarse Aggregate |
| CASOS | - | Center for Computational Analysis of Social and Organizational Systems |
| CPWA | - | Concrete Plastic Waste Aggregates |
| DOE | - | Design of Experiment |
| EDX | - | Energy Dispersion X-ray |
| EPS | - | Expanded Polystyrene |
| EPW | - | Electronic Plastic Aggregate |
| EVA | - | Ethylene-Vinyl Acetate |
| FA | - | Fine Aggregate |
| FTIR | - | Fourier-Transform Infrared Spectroscopy |
| HDPE | - | High-Density Polyethylene |
| HPC | - | High-Performance Concrete |
| IPW | - | Irradiated Plastic Waste |
| ITZ | - | Interfacial Transition Zone |
| LDPE | - | Low-Density Polyethylene |
| M | - | Mix Ratio |
| MMA | - | Methyl Methacrylate |
| MMT | - | Montmorillonite |
| MPW | - | Metalized Plastic Waste |
| MSW | - | Municipal Solid Waste |
| Mt | - | Million Tonnes |
| MW | - | Molecular Weight |
| Na | - | Sodium |
| NA | - | Natural Aggregates |
| NCA | - | Natural Crushed Aggregate |
| OM | - | Optical Microscopy |
| OMMT | - | Organically Modified Montmorillonite |
| OPC | - | Ordinary Portland Cement |
| PA | - | Plastic Aggregates |
| PA-6 | - | Polyamide-6 |

| | | |
|-------|---|--|
| PC | - | Polycarbonate |
| PCA | - | Plastic Composite Aggregates |
| PCS | - | Post-Cracking Strength |
| PE | - | Polyethylene |
| PES | - | Polyester |
| PET | - | Polyethylene Terephthalate |
| PF | - | Plastic Fibre |
| PFA | - | Plastic Fine Aggregates |
| PFA | - | Poly(tetrafluoroethylene-co-perfluoropropylvinylether) |
| PMMA | - | Polymethyl Methacrylate |
| PP | - | Polypropylene |
| PS | - | Polystyrene |
| PU | - | Polyurethane |
| PVC | - | Polyvinyl Chloride |
| PW | - | Plastic Waste |
| PWA | - | Plastic Waste Aggregates |
| rBOPP | - | recycled Biaxially-Oriented Polypropylene |
| RCA | - | Recycled Concrete Aggregates |
| RCA | - | Recycled Crushed Aggregates |
| RCG | - | Recycled Crushed Glass |
| RPW | - | Regular Plastic Waste |
| RSM | - | Response Surface Methodology |
| SEM | - | Scanning Electron Microscopy |
| SLA | - | Smart-Lightweight Aggregates |
| SPA | - | Substituted Plastic Aggregates |
| SS | - | Sum of Squares |
| TPS | - | Thermoplastic Starch |
| UNEP | - | United Nations Environment Programme |
| UTM | - | Universal Testing Machine |
| WA | - | Water-Assisted |
| XRD | - | X-Ray Diffractometers |
| XRF | - | X-Ray Fluorescence |

LIST OF PUBLICATIONS

1. Mohamad, N., Abd Ghani, A.A., Anen, M.A., Abd Razak, J., Raja Abdullah, R.I., Mohd Ali, M.A., Ab Maulod, H.E., and Meng, S.S., 2023. Optimization of Recycled Polypropylene Using Water-Assisted Melt Compounding via Response Surface Methodology. *Springer Proceedings in Physics*, 289 (July 2023), pp.47 – 54.
2. Zulkifli, N.S.A., Mohamad, N., Abd Ghani, A.A., Chang, S.Y., Abd Razak, J., Ab Maulod, H.E., Hassan, M.F., Abu Bakar, M.A.Q., Ani, M.A., Teng, M.M., and Ahsan, Q., 2022. Graphene Nanoplatelets Modified Chemlok® Adhesive System for Natural Rubber – Aluminium Bonded Component in Engine Mount. *International Journal of Automotive and Mechanical Engineering*, 19 (1), pp.9530 – 9542.
3. Mohamad, N., Abd Ghani, A.A., Amran, M.A.A., Abd Razak, J., Raja Abdullah, R.I., Mohd Ali, M.A., Ab Maulod, H.E., and Meng, S.S., 2022. Brief Review on Potential Production of Plastic Waste Concrete Aggregates Using Water-Assisted Melt Compounding. *Springer Nature Singapore Pte Ltd*, pp.523 – 532.
4. Abd Ghani, A.A., Mohamad, N., Amran, M.A.A, Abd Razak, J., Raja Abdullah, R.I., Mohd Ali, M.A., Mahamood, M.A., and Meng, S.S., 2021. Recycled Thermoplastic Concrete Aggregates from Water-Assisted Compounding: A Short Review. *Proceedings of Malaysian Technical Universities Conference on Engineering and Technology*, pp.108 – 109.

5. Abd Ghani, A.A., Mohamad, N., Abd Razak, J., Ahsan, Q., Yee, C.S., Ani, M.A., Teng, M.M., and Ul-Hamid, A., 2020. Optimization of Hot Press Parameters to Maximize the Physical and Mechanical Properties of Natural Rubber Composites for Elastomeric Mount. *Malaysian Journal on Composite Science and Manufacturing*, 1(1), pp.27 – 37.



CHAPTER 1

INTRODUCTION

1.1 Research Background

Concrete is the world's second most-used construction material after water. Instead of being a distinct entity, concrete is a composite of numerous constituents. The fundamental components are water, sand, cement, gravel or broken stones. Gravel or broken stones are examples of coarse aggregate, while sand is an example of fine aggregate. The cement coats and binds the fine and coarse particles when thoroughly combined. Shortly after the components are joined, the hydration reaction takes place, resulting in rock-solid concrete. However, environmental difficulties are already emerging as one of the greatest threats to the production of natural concrete aggregates. Natural concrete aggregate, which constitutes three-quarters of the composition of concrete, is a key concern as the primary component material. In 2007, Malaysia produced 77.7 million tonnes of natural aggregates on its own (Ismail and Ramli, 2013). Because the rising demand for concrete aggregates entails the substantial use of natural stone materials, the continued abuse of aggregates will eventually deplete the available supplies (Rahman et al., 2009).

On the other hand, worldwide plastic consumption has skyrocketed and plastic products have become an indispensable part of our everyday lives (Gu and Ozbakkaloglu, 2016). The name plastic is derived from the Greek word *plastikos*, which means mouldable. Because of its flexibility or malleability, this term refers to a material's ability to be cast, extruded or pressed into various shapes (Plastics Europe, 2019). Among the several forms of recycling management

systems, repurposing plastic waste in the building industry is regarded as a good alternative for plastic waste disposal. Recycled plastics may be utilized without losing quality throughout the service cycle and can even be used instead of virgin building materials. Plastics were widely used in concrete as plastic aggregates (PA), which substituted natural aggregates, and several researchers investigated the characteristics of concrete, including plastic components (Almeshal et al., 2020).

The United Nations Environment Programme (UNEP) estimates that more than 400 million tonnes of plastic are produced annually worldwide. Up till 2015, approximately 6300 million tonnes of plastic waste has been produced. About 9% are recycled, 12% are burned and 79% are placed in landfills or the environment. If current development and waste disposal trends continue, by 2050, landfills and the natural environment will contain more than 12 billion tonnes of plastic waste (Geyer et al., 2017). Reprocessing waste plastic material into concrete is an environmentally viable alternative to plastic waste disposal due to its ecological and economic benefits. In addition, this reduces the quantity of plastic waste that is burned and the amount of plastic waste that is produced. Polypropylene (PP) is chosen for this study because it is widely used as a variety of packaging materials, especially in the form of bottles and has high strength and hardness.

This research project investigates the feasibility of generating plastic waste aggregate by water-assisted melt compounding of plasticized thermoplastic-clay composite (PCA). Water-assisted (WA), also known as liquid-mediated melt compounding of composites. It is fundamentally a solution-mixing technique. It allows for greater material dispersion than conventional melting, but is restricted to soluble chemicals. It is an increasing strategy for overcoming the disadvantages of individual melt compounding and solution mixing. When mixed with additives, water or aqueous liquids act as more than just temporary carriers for