An Acad Bras Cienc (2022) 94(3): e20201601 DOI 10.1590/0001-3765202220201601
Anais da Academia Brasileira de Ciências | *Annals of the Brazilian Academy of Sciences*
Printed ISSN 0001-3765 I Online ISSN 1678-2690
www.scielo.br/aabc | www.fb.com/aabcjournal

CHEMICAL SCIENCES

Synthesis of N-acyl glycine surfactant from palm oil as green repellent and toxicant to termite (*Microcerotermes diversus*)

SYAMIMI N.F. SAZALEE, NUR A.A.A. RUSLAN, NURHAMIZAH NORDIN, WAHIZATUL A. AZMI, VICIT R.E. SUK, MISNI MISRAN, TAN S. YONG, KOAY K. TEIK & POH W. CHIA

Abstract: This study described for the first time, the synthesis of a greener, safer, and more effective termiticide using a bio-based surfactant, *N*-acyl glycine derived from palm oil for the control of *Microcerotermes diversus*. Laboratory findings showed that the highest repellent activity was observed in *N*-acyl glycine surfactant (83.33%) at 50 ppm. In addition, *N*-acyl glycine surfactant also exhibited substantial time and concentration-dependent anti-termiticidal activity in which the highest termite mortality was observed after 3 days of exposure at 50 ppm of the surfactant (100%). Furthermore, 32.49 ppm concentration of *N*-acyl glycine surfactant (LC_{50} = 32.49 ppm) attained 50% of termite lethality. The current innovated termiticide with the use of *N*-acyl glycine surfactant offers a better efficacy, lower cost, and prevents the use of dangerous termiticides that are critical in creating a more sustainable environment, and controls *Microcerotermes diversus* at the same time.

Key words: Green termiticide, bio-based surfactant, *N*-acyl glycine surfactant, *Microcerotermes diversus*, repellency, toxicity.

INTRODUCTION

Termites are the most common and destructive pests of crops, forests, and other cellulose products, such as papers, houses, and plastics (Su & Scheffrahn 2000). They feed on a wide range of organic detritus, such as decaying leaves, humus, dry grass, and living or dead wood, and serve as an essential ecological role by converting dead materials into organic matter (Kuswanto et al. 2015). Termites' natural activities will potentially help increase pH, organic carbon content, water content, and soil porosity by recycling dead organics in their natural environment (Ghaly & Edwards 2011). Nevertheless, as termites continue to function as an ecological engineer, they cause many serious problems in which many objects become highly destructive, resulting in major losses in many parts of the world (Oludairo et al. 2016). Since termites do not leave any obvious shreds of evidence to their presence, their existence is typically found after severe damage has been discovered, which makes it difficult to control because of the cost needed to treat the damaged buildings and other wood products (Ugbomeh & Diboyesuku 2019). It is therefore necessary to use termiticides to control termites and also to prevent these damaging pests from continually causing loss to the world.

Traditionally, chemical methods are the most common method of termite control and are likely to remain a significant component of control strategies in the future (Toughan et al.

2017). The repellence and toxicity of the chemical barrier avoid tunnelling and penetration of termites into soil and structure, and thus preventing infestation. Bifenthrin, disodium octaborate tetrahydrate (DOT), calcium arsenate, and chlorpyrifos (Kuswanto et al. 2015) which are either contact or systemic poisons are the active ingredients contained in termiticides (Acda 2018). Chlorpyrifos or also known as organophosphate is one of the examples of chemical termiticides that has been commonly used to combat termites by causing paralysis, convulsions, and finally death through the inhibition of acetylcholinesterase enzyme, a neurotransmitter that results in overstimulation of termites' nervous system (Dar et al. 2019). Organochlorine termiticides, like heptachlor, have also been used for termite control in some countries such as China, where it is mainly combined with seeds such as grain and corn to kill termites in soil (Qiu et al. 2018). In addition, bifenthrin, a synthetic pyrethroid with the advantages of high efficiency, low toxicity, high spectrum, and long effective time (Hua et al. 2015) was a common treatment for repelling and killing termites, particularly for the preservation of wood products (Kotlarewski et al. 2019). Non-repellent termiticides such as fipronil (Acda 2018) and imidacloprid (Henderson et al. 2016) are another preferable approach used in controlling termites especially for colony management in which they act as toxicants to termites (Henderson et al. 2016). However, due to the environmental concerns over toxic and non-green compounds in the formulation of chemical termiticides, termiticides have been banned in certain countries (Chand et al. 2018).

In recent years, the use of greener pesticides or insecticides has received great interest among scientists (Abid et al. 2020, Attaullah et al. 2020, Boris et al. 2016, Khanikor et al. 2018, Oliveira et al. 2017, Rasib & Aihetasham 2016, Roszaini et al. 2013). As part of our continuous effort in the field of green and sustainable chemistry and education research (Abdullah et al. 2021, Asseri et al. 2017, Chia et al. 2019, Ruslan et al. 2020, Yap et al. 2020), we would like to report the synthesis of the bio-based surfactant as a termiticide or repellent against *Microcerotermes diversus*, which is the most destructive termite that causes serious economic damage for wooden products in buildings (Habibpour 2010). In this research, a bio-based surfactant, namely the *N*-acyl glycine amino acid surfactant was synthesised from the palm oil to act as a natural repellent and toxicant to termite (*Microcerotermes diversus*). In particular, the *N*-acyl glycine amino acid surfactant has been widely used in various areas (Reznik et al. 2010), such as personal care products, household products, cosmetics, and detergents due to their physicochemical properties and antimicrobial activities (Xia et al. 1995). Due to the enormous use of hazardous chemicals as pesticides for termite control, there is a growing concern about these chemicals whereby they can potentially harm the environment and give negative impacts on living things. As such, the use of green bio-based surfactant, *N*-acyl glycine as a repellent and toxicant to termites is an alternative to the conventional one. According to the literature, *N*-acyl glycine amino acid surfactant is generally considered low in toxicity, non-irritating, low allergenic potential, good biodegradability, and harmless to the marine environment (Yea et al. 2018). Therefore, this research aims in reducing the use of hazardous termiticides in which the concept of green chemistry for toxic waste and pollution prevention can be empowered. Besides, the environment and humans can also benefit from this approach.

MATERIALS AND METHODS

Materials and equipment

In this study, all chemicals and reagents were obtained from Sigma-Aldrich (Malaysia), including sodium hydroxide, hydrochloric acid, glycine sodium salt hydrate, and thionyl chloride. These chemicals were directly used in the current experiment. The characterization of *N*-acyl glycine surfactant was performed using the Fourier-Transform-Infrared (FT-IR). The Gas Chromatography-Mass Spectrometry (GC-MS) was utilised to analyse the molecular mass, and the values were given in the unit of mass divided by charge ration (m/z).

Termites collection

The population of subterranean termites, *Microcerotermes diversus* was collected from a termitarium at Universiti Malaysia Terengganu, Kuala Terengganu. The termite mound was dug up using a shovel and the collected termites were then deposited in plastic containers. Termites inside the container were fed with dry wood and the container was covered with a muslin cloth. The container carrying termites was placed in a cool dark area inside the laboratory until needed.

Synthesis of *N*-acyl glycine surfactant

The *N*-acyl glycine surfactant was prepared following the previous literature with slight modification on it (Zhang et al. 2016). Initially, a 250 mL three-necked round bottom flask suspended with glycine sodium salt hydrate (3 mmol) was added with a catalytic amount of sodium methoxide and palm oil (1 mmol). The reaction mixture was heated at 160°C and left for stirring for 5 h. After cooling to 60°C, the reaction mixture was added with water (30 mL) and acidified with aqueous HCl (2 M). The *N*-acyl glycine was collected in the form of white precipitate after filtration.

Characterisation of sodium N-acyl glycine surfactant

Fourier-transform infrared (FTIR) spectroscopy

About 100 µg of sodium *N*-acyl glycine surfactant was placed on a clean stainless-steel plate of Fourier Transform Infra-Red (FTIR) Spectrum 400 (Perkin Elmer, USA) equipped with GladiATR™ Attenuated Total Reflectance (ATR) (Pike Technologies, USA). The spectra were measured at a resolution of 4 cm^{-1} within the range of 4000-450 cm^{-1}, with 32 scans per sample. The wavenumber of the FT-IR spectroscopy peaks was coordinated to the respected functional group (Lucarini et al. 2018, Suk et al. 2019). The *N*-acyl glycine surfactant derived from palm oil: yield 96%. IR (mmax, cm^{-1}): 3450 (N-H), 1702 (C=O), 1640 (amide band I), and 1580 cm^{-1}(amide band II).

Gas-Chromatography-Mass spectroscopy (GC-MS)

The mass spectroscopy analysis of the *N*-acyl glycine surfactant derived from palm oil was characterised over the GC-MS Shimadzu model (QP2010SE), mounted with a silica capillary column. The electrospray ionization mass spectroscopy (ESI-MS) was m/z= 284 (myristoyl glycine), 312 (palmityl glycine), 336 (linoleoyl glycine), 338 (oleoyl glycine), and 340 (stearoyl glycine), respectively.

Differential scanning calorimetry (DSC)

About 5 mg of *N*-acyl glycine surfactant was transferred into 40 µL Tzero aluminium pans and was carefully sealed with Tzero Hermetic Lid using a Tzero sample press. The pan and lid were transferred to the sample holder of DSC Q20 Difference Scanning Calorimetry (TA

Instruments, USA). An empty Tzero pan, and lid was used as a standard. The DSC was set to ramp at 10 °C min^{-1} to 250 °C in the presence of nitrogen gas at flow rates of 30 mL min^{-1} (Jin et al. 2014). The melting temperature of the sample was evaluated from the sharp peak in the thermogram.

Determination of critical micelles concentration (CMC)

Critical micelles concentration (CMC) of the *N*-acyl glycine surfactant was investigated using surface tension, turbidity, and DLS techniques (Saxena et al. 2017, Teo et al. 2011). The *N*-acyl glycine surfactants of 0.005, 0.010, 0.015, 0.020, 0.025, 0.030, 0.035, 0.040, 0.045, and 0.050 g mL^{-1} in deionised water were prepared and left to homogenise overnight at room temperature. The surface tension, turbidity, and DLS of each sample were measured in triplicates at room temperature. The results were plotted against the *N*-acyl glycine surfactant concentration.

Morphological observation

The morphology, size, and distribution of *N*-acyl glycine surfactant micelles were observed using computer-controlled High-Resolution Transmission Electron Microscopy (HRTEM) JEM2100F (JEOL, USA) equipped with Gatan software (Gatan Inc, USA). A day old sample was spiked on to a 400 mesh copper-coated carbon grid, followed with a drop of 3% phosphotungstic acid as a negative staining agent (Suk & Misran 2017). The excess liquid was carefully removed using a clean filter paper. The grid was incubated in the desiccator overnight. The micrograph was taken immediately after the satisfactory replica was observed to prevent micelles from disintegrating due to exposure at high kV of energy source.

Average hydrodynamic radius and surface charge

The average hydrodynamic radius and surface charge of the micelles prepared from the *N*-acyl glycine surfactant was measured using Zetasizer in the Centre for Fundamental and Frontier Sciences in Nanostructure Self-Assembly (FSSA), UM. The sample was carefully transferred into a clean gold plated U-shaped capillary cell. The average hydrodynamic radius and surface charge of the micelles were measured in triplicates at 30°C (Suk et al. 2020).

Toxicity determination

This method was done based on Roszaini et al. (2013) study (Roszaini et al. 2013) with some modifications. Solutions of 30 ppm, 40 ppm, and 50 ppm were prepared by dissolving *N*-acyl glycine surfactant in distilled water. The solutions were then applied to Whatman No. 1 filter paper samples with a 9 cm diameter and dried for 1 h. Filter papers treated with distilled water were used as control and three replicates were used for each test. Ten termites were introduced onto each petri dish and they were kept in the dark at room temperature. Termite mortality was counted and recorded every 24 h for 3 days. The percentage of mortality was determined using a formula based on a current literature (Cynthia et al. 2016):

$$Percentage\ mortality = \frac{No\ of\ dead\ termites}{Total\ no\ of\ termites} \times 100$$

Repellence test

This method was done based on Roszaini et al.'s (2013) study (Roszaini et al. 2013) with some modifications. Solutions of 30 ppm, 40 ppm, and 50 ppm were prepared by dissolving *N*-acyl glycine surfactant in distilled water. One mL of different concentrations of *N*-acyl glycine surfactant solutions were applied to half of the

filter paper measuring 9 cm in diameter and another half of the filter was treated with 1 mL of distilled water. Both parts of the filter paper were then dried for 1 hour and were immediately re-joined with adhesive tape. Next, each re-joined filter paper was placed in a petri dish and ten termites were introduced at the centre of the petri dish. The experiment was replicated three times with ten termites in each replicate. The number of termites was counted on both treated and untreated filter papers after 1 h. The percentage of repellent was determined using the following formula (Roszaini et al. 2013):

$$Repellent\ percentage = \frac{Nc - Nt}{Nc + Nt} \times 100$$

Where,

Nc = Number of termites present in the control

Nt = Number of termites present in the treated half

Data analysis

The repellency percentage of the *N*-acyl glycine surfactant was analysed using a one-way analysis of variance (ANOVA), followed by Tukey post-hoc test from the IBM statistical package for the social science (SPSS) software Version 20.0. The percentage mortality rate for each concentration was also determined. For toxicity, the value of lethal concentration 50 (LC_{50}) of the *N*-acyl glycine surfactant was determined by plotting a graph of percentage cumulative mortality (%) against the *N*-acyl glycine surfactant concentration (ppm) using probit analysis.

RESULTS AND DISCUSSION

Characterization of N-acyl glycine surfactant

DSC thermograms of surfactant

Figure 1 shows the thermogram of *N*-acyl glycine surfactant. The sharp peak, indicating the melting point was observed at 147.8°C. The obtained melting point value was in agreement with the previous reported melting points of amino acid-based surfactants (El-Sayed et al. 2018). The glass transition temperature, which is the temperature range where the surfactant changes from a rigid to soft material, appeared within 25°C to 35°C. Degradation peak was

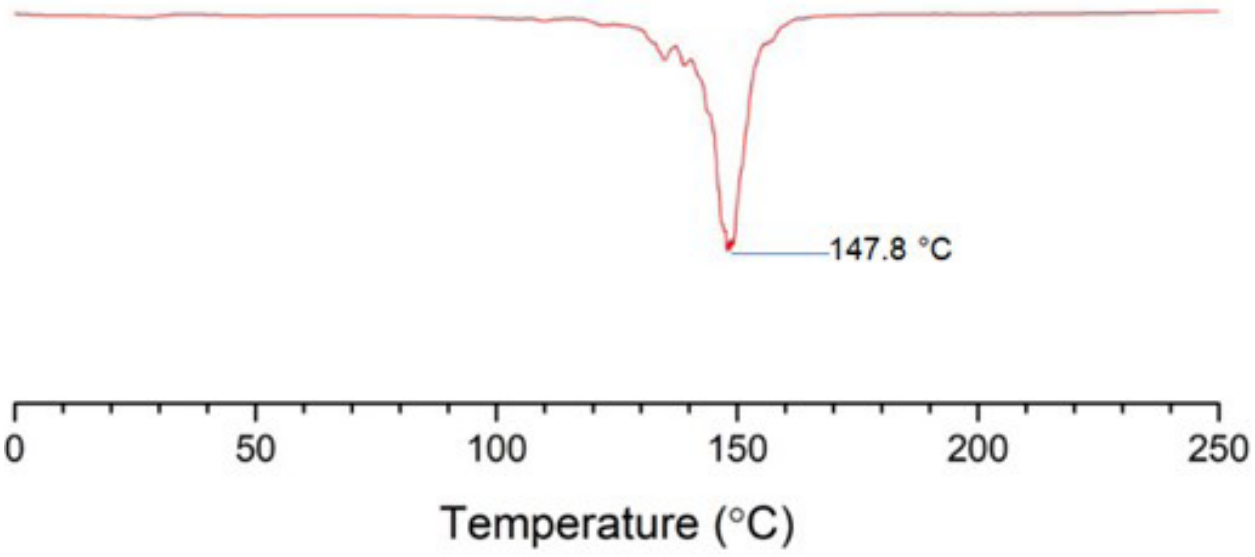

Figure 1. Differential Scanning Calorimetry (DSC) thermogram of *N*-acyl glycine surfactant.

not observed in the thermogram-tested temperature, indicating that the *N*-acyl glycine surfactant was thermally stable up to 250° C.

Determination of critical micelles concentration (CMC)

The CMC was determined at the inflection point of surface tension, turbidity, and DLS curves. The inflection points occurred due to the spontaneous formation of large micelle particles. Table I shows that the minimum concentration needed to form the *N*-acyl glycine surfactant micelles was less than 0.1 g ml^{-1}. The average CMC was 0.0174±0.0021 g ml^{-1}. This is compatible with the average hydrodynamic size, as depicted in Table II. All data are presented as means ± standard deviation of the mean (SD).

Surfactant is known to adsorb at the interface of the two immiscible systems and reduce their interfacial free energy (Rosen & Kunjappu 2012). At a low concentration of surfactant in the solution, there were a few monomers present in the solution. Therefore, the solution appeared as a clear colourless solution. The molecules were diffused from the bulk, concentrated, and re-orientated so that their hydrophobic groups were directed away from the solution at the liquid-gas interface, and thus affecting their interfacial free energy. The interfacial free energy, γ, is a minimum work needed to create a boundary at the interface per unit area and is usually called an interfacial tension for the liquid-liquid interface and the surface tension for the liquid-gas interface. The interfacial free energy of deionised water was measured as 71.2 mNm^{-1}. As the amount of *N*-acyl glycine surfactant in the solution was increased gradually, the presence of its monomers at the liquid-gas interface was proportionally increased, and the interfacial free energy was measured to be 33.6 mN m^{-1}. Further addition of *N*-acyl glycine surfactant will cause its monomer saturating at the liquid-gas interface and spontaneously forming the micelles in order to reduce the free energy of the system by removing hydrophobic groups from contacts with the water (Tan & Misran 2013). The solution has gradually become cloudy and larger particle was detected. This phenomenon can be detected by the inflection point of the graph. The concentration where the monomers were saturated and spontaneously formed a particle is called CMC and this phenomena were also can be observed by turbidity and DLS plots.

The equilibrium interfacial tension profiles of the liquid-gas interface were constructed by plotting interfacial pressure, Π against the natural logarithm concentration of *N*-acyl glycine surfactant. Interfacial pressure, Π is the difference in γ of the given liquid-gas interface in the absence ($\gamma_{l/g}$) and in the presence of *N*-acyl glycine surfactant, ($\gamma_{l+s/g}$) which is related as $\Pi = \Delta\gamma = \gamma_{l/g} - \gamma_{l+s/g}$. The equilibrium interfacial profile presented that interfacial pressure, Π was affected by bulk *N*-acyl glycine surfactant concentration, which indicated that

Table I. Critical Micelles Concentration (CMC) (gml^{-1}), maximum adsorption density, Γ, and minimum surface area per surfactant molecule, A_{min}, of *N*-acyl glycine surfactant at room temperature.

Critical Micelles Concentration (CMC) (gml^{-1})				Maximum adsorption density, Γ ($mol\ m^{-2}$)	Minimum surface area per surfactant molecule, A_{min} ($Å^2$)
Turbidity technique	DLS technique	Surface tension technique	Average		
0.0185	0.0150	0.0186	0.0174±0.0021	4.06×10^{-6}	40.86

it was adsorbed efficiently at the given liquid-gas interface. The data were then fitted into a linear equation and yielded a straight line with the linear equation of y = -10.24x - 1.70. The regression coefficient was 0.94, proving that *N*-acyl glycine surfactant was considered pure, probably because of the absence of surface-active impurities (Lunkenheimer et al. 1995).

The interfacial pressure, Π, and bulk surfactant concentration were related to the maximum adsorption density, Γ, which can be determined through Gibbs adsorption equation as $d\Pi/d\ ln\ C = -nRT\Gamma$, where R is the universal gas constant, 8.314 Nm mol^{-1} K^{-1}, *T* is the absolute thermodynamic temperature, 303.15 K, n is the number of molecular species involved, and $d\Pi/dlnC$ was the slope of equilibrium interfacial. For a single surfactant component, n=1 (Matuura et al. 1959, Tadros 2013). Next, the value of Γ can be related to the demand for minimum surface area per surfactant molecule, A_{min}, by using the equation $A_{min} = 1/(N_A\Gamma)$, where N_A stands for Avogadro number, 6.02×10^{23}. The A_{min} value indicated the area occupied by one surfactant molecule at the saturated surfactant-air interface (El Feky et al. 2016). The Γ and A_{min} of *N*-acyl glycine surfactant were calculated and presented in Table I, showing that *N*-acyl glycine surfactant is comparable to other amino acid surfactants and has the highest tendency to be packed more orderly and tightly (Sreenu et al. 2014). Altering the active groups, chain length, conformation, and degree of unsaturation of the hydrocarbon chains will affect the area per surfactant molecule as well as the orientation of surfactant at the interface (Petelska & Figaszewski 2011).

Morphological observation

High-Resolution Transmission Electron Microscopy (HRTEM) was employed to study the morphology of the micelles due to its ability to deliver the energetic electrons to the surface of the specimen and provide the information on the morphological, compositional, and crystallographic arrangement of the sample up to atomic-scale. Figure 2 shows the *N*-acyl glycine micelles and its HRTEM micrograph, where the formation of light or dark halos around the structures was due to the negative staining methods using phosphotungstic acid. The micrograph was corrected by aberration mode in order to avoid the presence of fringe lattices, improved the contrast as well as visibility of micelles (House et al. 2016). The micrographs showed the presence of spherical micelles particles less than 100 nm dispersedly distributed in the micrograph (Sreenu et al. 2014). However, aggregation of the particles was observed which may due to the intermolecular association of micelles as well as the drying of the process of the samples. The aggregation of the particles raised the average hydrodynamic radius of the *N*-acyl glycine surfactant micelles as displays in Table II.

Average hydrodynamic radius and surface charge

The average hydrodynamic radius of *N*-acyl glycine surfactant micelles was measured using

Table II. Melting point, average hydrodynamic radius, zeta potential, conductivity, and mobility of *N*-acyl glycine surfactant at 30˚C.

Melting point (°C)	Average Hydrodynamic Radius (nm)	Zeta Potential (mV)	Conductivity (mS/cm)	Mobility (μmcm/Vs)
147.8	207.60±12.17	-87.40±5.38	0.163±0.002	-6.85±0.42

dynamic light scattering (Anderson et al. 2013, Storey & Ymén 2011). Table II shows the average hydrodynamic size of *N*-acyl glycine surfactant micelles, where the average size was 207.60±12.17 nm. The large hydrodynamic size of micelles was mainly due to the micelles growth and aggregation of particles (Sreenu et al. 2014).

Evaluating the electrical charge of the particles provides important information in defining the properties of a colloidal suspension, such as flotation, flocculation, and stability of suspended particles (Hunter 2013). Zeta potential of *N*-acyl glycine surfactant micelles was found to be negative, which was -87.40±5.38 mV. This was due to the distribution of ionised fatty acid (Fameau et al. 2014) surrounding the particles that increased the counter ions close to the surface and formed the electrical double layer around the particle. The less negative zeta potential values that were observed explained that the micelles were less forced to contact to each other, adhered, or aggregated due to the attractive van der Waals forces that overcame the electrical double layer repulsive forces (Crooke 2007) as observed in the HRTEM micrograph.

Repellent activity of N-acyl glycine surfactant

Subsequently, the repellent activity of *N*-acyl glycine surfactant was studied on termites at different concentrations, 30, 40, and 50 ppm, respectively, as shown in Figure 4. The result in Figure 3 shows a significant increase in repellence activity as the concentration of *N*-acyl glycine surfactant increased from 30 ppm to 50 ppm. The 50 ppm of *N*-acyl glycine surfactant showed the highest repellency activity towards termites compared to the other concentration with 83.33 ± 8.82% (p = 0.01) which was quite similar to the repellency activity reported in previous literature using an essential oil (*Dipterocarpus* sp.) against termites which was 84% (Roszaini et al. 2013). Meanwhile, the lowest repellency was noted at

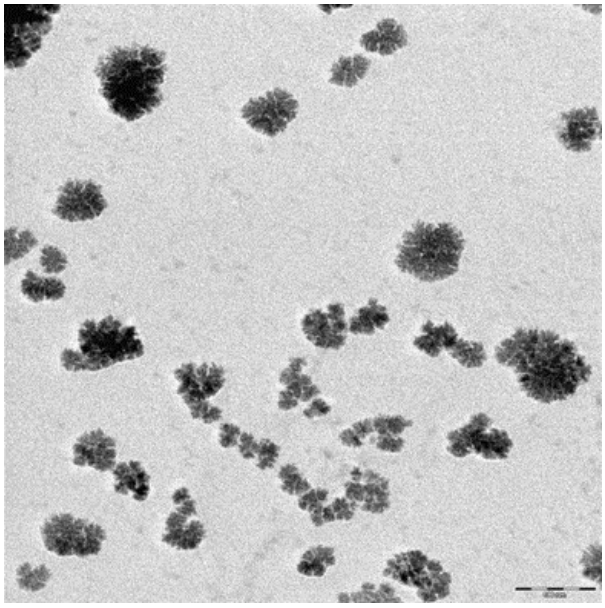

Figure 2. High-Resolution Transmission Electron Microscopy micrographs of *N*-acyl glycine surfactant micelles observed at room temperature. The concentration was 0.20 g ml^{-1} and the scale is 100 nm.

30 ppm of the surfactant with 23.33 ± 8.82% (p = 0.01). The 40 ppm concentration showed no significant difference, either from 30 ppm or 50 ppm. Mainly, the *N*-acyl glycine surfactant is made up of several fatty acid components which can serve as a potential repellent towards termites, as described by many researchers in the previous studies. This was supported by a research carried out by Rasib & Aihetasham (2016) in which wood extract, *Morus alba* shows 100% repellency activity at a concentration of 30% w/v when treated on termites, *Coptotermes heimi* due to the presence of fatty acids, a major compound found in *Morus alba.*

Toxicity activity of N-acyl glycine surfactant

As *N*-acyl glycine surfactant shows good repellency properties towards termites, the toxicity activity of the surfactant was also studied in this research. In particular, fatty acids are known for being biologically active against pest anthropods in which the toxicity of these compounds increases with the length of the

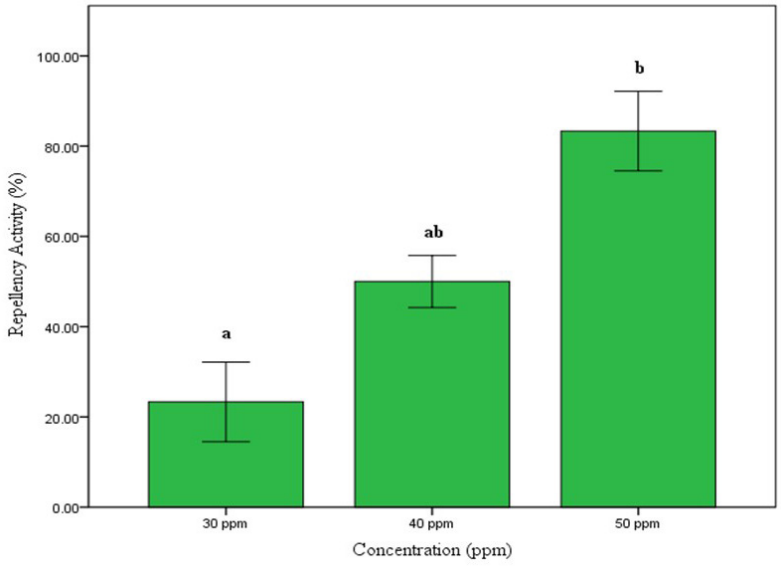

Figure 3. Repellency activity of *Microcerotermes diversus* to different concentrations of *N*-acyl glycine surfactant. Different letters above bars indicate a statistically significant difference at $p < 0.05$.

carbon chain (Sims et al. 2014) along with the existence of chemical bonds, saturated and unsaturated (Baldwin et al. 2008). These have been proven by the earlier research carried out by Oliveira et al. (2017) (Oliveira et al. 2017) in which vegetable oil containing fatty acids composition was able to kill 50% of mites even at low concentrations. In the GC-MS analysis, the fatty acids that were detected in *N*-acyl glycine surfactant were linoleic acid (C18:2), linolenic acid (C18:3), myristic acid (C14:0), palmitic acid (C16:0) as well as oleic acid (C18:1) and stearic acid (C18:0) resulted a higher termiticidal ability against termites. Figure 4 shows that *N*-acyl glycine surfactant had a major impact on the mortality of the subterranean termite in which all of the concentration tested, 30 ppm, 40 ppm, and 50 ppm showed significant toxicity. From the result obtained, the highest termite mortality was noted at 50 ppm of *N*-acyl glycine surfactant which indicates the highest toxicity, while the percentage of mortality gradually increased with an increase in exposure time between 30 ppm and 40 ppm, respectively.

Figure 4 demonstrates that 50 ppm of *N*-acyl glycine surfactant is more effective than the other concentrations in terms of termite mortality in which the complete lethality of termite (100%) was obtained in day 3 of exposure time. Meanwhile, 30 ppm of *N*-acyl glycine surfactant reported the least mortality rate, 55%. When comparing the current study with the previous one (Khanikor et al. 2018), *N*-acyl glycine surfactant is more likely to act as a potential termiticide since it only requires a maximum of 50 ppm concentration to cause complete mortality (100%) of termites. Besides, a study conducted by Roszaini et al. (2013) revealed that the analysis took 25 days to reach 100% of termite mortality, whereas N-acyl glycine surfactant only took 3 days to result in complete lethality. As shown in Table III, *N*-acyl glycine surfactant only requires a concentration of 32.49 ppm to cause 50% of termites' lethality (LC_{50}). The results were comparable with commercially available termiticides that were tested on subterranean termite such as Bifenthrin (up to 98% mortality after 1 day treatment), Fipronil (up to 91% mortality after 1 day treatment), Chlorfenapyl (up to 42% after 1 day treatment), and Indoxacarb (up to 27% after 1 day treatment). This show that *N*-acyl glycine surfactant is possible to be developed as a termiticides.

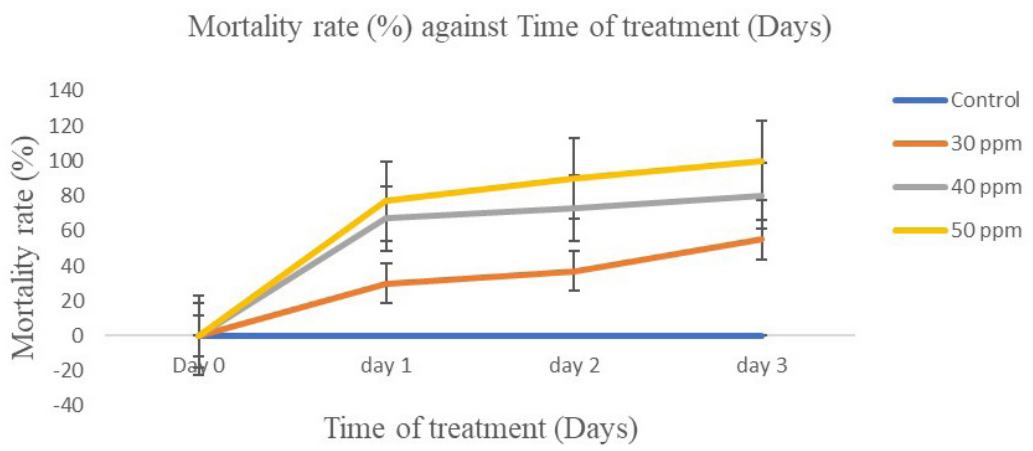

Figure 4. Graphs showing the mortality rate of subterranean termite after treatment of *N*-acyl glycine at different concentrations.

Table III. The values of LC_{50} of *N*-acyl glycine surfactant against subterranean termite, *Microcerotermes Diversus* obtained from probit analysis.

Bio-based surfactant	LC_{50} value (ppm)	Regression equation	R^2 (2df)
N-acyl glycine surfactant	32.49	y = 94.78ln(x) – 279.93	0.99

CONCLUSION

The result of this study clearly shows that the bio-based surfactant, *N*-acyl glycine surfactant can be developed into a new repellent or toxicant against termites. In this study, *N*-acyl glycine surfactant was a strong termiticide against subterranean termite causing complete mortality in the laboratory at the concentration of 50 ppm. Besides, the value of LC_{50} of *N*-acyl glycine surfactant obtained in this study was 32.49 ppm. Several advantages were identified using the *N*-acyl glycine surfactant as termiticide including the elimination of the use of hazardous termiticides, inexpensive, and biodegradable. Additionally, the desired product was synthesised using a natural product, palm oil which is considered environmentally friendly and does not cause harmful impacts to humans and the environment. Biosynthesis and electrochemical methoxylation could possibly be the alternatives in preparing the surfactant with minimum involvement of HCl and heat. Above and beyond, this research also helps to empower the concept of green chemistry for toxic waste and pollution prevention which begins with the collaboration among academics, industry, and government. Finally, the researchers anticipated that the current innovation of termiticide with better efficacy and low cost will be an attractive method for termite control in the future.

Acknowledgments

The authors would like to acknowledge The Ministry of Education Malaysia for Fundamental Research Grant Scheme (FRGS) (Vot. No. 59499), UMT/RMIC/FRGS/1/2018/59499(1) and Universiti Malaysia Terengganu for research facilities.

REFERENCES

ABDULLAH HSTSH, ASSERI SNARM, MOHAMAD WNKW, KAN SY, AZMI AA, JULIUS FSY & CHIA PW. 2021. Green synthesis, characterization and applications of silver nanoparticle mediated by the aqueous extract of red onion peel. Environ Pollut 271: 116295. DOI 10.1016/j.envpol.2020.116295.

ABID AD, SAEED S, ZAKA SM, ALI M, SHAHZAD MS, KHAN KA & IQBAL N. 2020. Manifold passages in an assorted infection in a host could improve virulence of Helicoverpa armigera Nucleopolyhedrovirus (HaNPV). Saudi J Biol Sci 27: 1419-1422. DOI 10.1016/j.sjbs.2020.02.023.

ACDA MN. 2018. Sustainable termite management using physical barriers. Termit Sustain Manag 219-232. DOI 10.1007/978-3-319-68726-1_10.

ANDERSON W, KOZAK D, COLEMAN VA, JAMTING AK & TARU M. 2013. A comparative study of submicron particle sizing platforms: Accuracy, precision and resolution analysis of polydisperse particle size distributions. J Colloid Interface Sci 405: 322-330. DOI 10.1016/j.jcis.2013.02.030.

ASSERI SNARM, TAN SH, MOHAMAD WNKW, POH SC, CHIA PW, KAN SY & CHUAH TS. 2017. MgCl2 as efficient and inexpensive catalyst for the synthesis of 1, 4-dihydropyridine derivatives. Malays J Anal Sci 2: 13-19. DOI 10.17576/mjas-2017-2101-02.

ATTAULLAH ET AL. 2020. Insecticidal, biological and biochemical response of Musca domestica (Diptera: Muscidae) to some indigenous weed plant extracts. Saudi J Biol Sci 27: 106-116. DOI 10.1016/j.sjbs.2019.05.009.

BALDWIN RW, KOEHLER PG & PEREIRA RM. 2008. Toxicity of fatty acid salts to German and American cockroaches. J Econ Entomol 101: 1384-1388. DOI 10.1603/0022-0493(2008)101[1384:TOFAST]2.0.CO;2.

BORIS DK, WOLALI SN, ABLA DM, GUILLAUME KK & ISABELLE AG. 2016. Termiticidal activities of few plant extracts against Macrotermes subhyalinus smeathman and Trinervitermes geminatus wasmann (Isoptera: Termitidae) survival. African J Agric Res 11: 2475-2480. DOI 10.5897/ajar2016.11146.

CHAND RR, JOKHAN AD, CHARAN H, RAJ K & SINGH P. 2018. Threats posed by Asian subterranean termites in the Fiji Islands and their potential controls: A review. New Zeal Plant Prot 71: 129-139. DOI 10.30843/nzpp.2018.71.111.

CHIA PW, POH SC, AZIZ MH, RADZI SAM, YONG JFS & KAN SY. 2019. Water extract of onion peel ash: an efficient green catalytic system for the synthesis of isoindoline-1, 3-dione derivatives. Malay J Anal Sci 23: 23-30. DOI 10.17576/mjas-2019-2301-03.

CROOKE ST. 2007. Antisense drug technology: Principles, strategies, and applications. 2nd ed, CRC Press: Florida, 848 p.

CYNTHIA OC, PRECIOUS O & LYNDA OS. 2016. The toxicity and repellency of some plant extracts applied as individual and mixed extracts against termites (Macrotermes bellicosus). J Entomol Zool Stud JEZS 4: 406-418.

DAR MA, KAUSHIK G & VILLARREAL-CHIU JF. 2019. Pollution status and bioremediation of chlorpyrifos in environmental matrices by the application of bacterial communities: A review. J Environ Manage 239: 124-136. DOI 10.1016/j.jenvman.2019.03.048.

KUSWANTO E, AHMAD I & DUNGANI R. 2015. Threat of subterranean termites attack in the asian countries and their control: A Review. Asian J Appl Sci 8: 227-239. DOI 10.3923/ajaps.2015.227.239.

El FEKY AA, SHALABY M, El-SHAMY OA & SUZY A. 2016. Surface activity and adsorption of some surfactants at aqueous/air interface at different temperatures. Int J Sci Techno Res 5: 179-184.

EL-SAYED R, ALOTAIBI HH & ELHADY HA. 2018. Synthesis, surface parameters, and biodegradability of water-soluble surfactants for various applications. J Oleo Sci 67: 551-569. DOI 10.5650/jos.ess17214.

FAMEAU AL, ARNOULD A & SAINT-JALMES A. 2014. Responsive self-assemblies based on fatty acids. Curr Opin Colloid Interface Sci 19: 471-479. DOI 10.1016/j.cocis.2014.08.005.

GHALY A & EDWARDS S. 2011. Termite damage to buildings: Nature of attacks and preventive construction methods. Am J Eng Appl Sci 4: 187-200. DOI 10.3844/ajeassp.2011.187.200.

HABIBPOUR B. 2010. Laboratory evaluation of Flurox, a chitin synthesis inhibitor, on the termite, *Microcerotermes diversus*. J Insect Sci 10: 1-8. DOI 10.1673/031.010.0201.

HENDERSON G, GAUTAM BK & WANG C. 2016. Impact of ground-applied termiticides on the above-ground foraging behavior of the formosan subterranean termite. Insects 7: 1-10. DOI 10.3390/insects7030043.

HOUSE SD, BONIFACIO CS, GRIESHABER RV, LI L, ZHANG Z, CISTON J, STACH EA & YANG JC. 2016. Statistical analysis of support thickness and particle size effects in HRTEM imaging of metal nanoparticles. Ultramicroscopy 169: 22-29. DOI 10.1016/j.ultramic.2016.06.007.

HUA X ET AL. 2015. A sensitive monoclonal antibody-based enzyme-linked immunosorbent assay for the detection of bifenthrin in a chemical soil barrier. Sci Total Environ 502: 246-251. DOI 10.1016/j.scitotenv.2014.09.032.

HUNTER RJ. 2013. Zeta Potential in colloid science: Principles and applications volume 2 of colloid science, revised. Academic Press, London, 386 p.

JIN X, XU X, ZHANG X & YIN Y. 2014. Determination of the PCM melting temperature range using DSC. Thermochim Acta 595: 17-21. DOI 10.1016/j.tca.2014.09.004.

KHANIKOR B, BARMAN J, SARMA R, MAHANTA S & ADHIKARI K. 2018. Evaluation of efficacy of three essential oils against Odontotermes feae (Isoptera: Termitidae). J Environ Pollut Hum Heal 6: 68-76. DOI 10.12691/jephh-6-2-4.

KOTLAREWSKI NJ, DERIKVAND M, LEE M, NOLAN G & HAGUE JRB. 2019. Bifenthrin treatment for balsa: Susceptibility

of Papua New Guinea-grown Ochroma pyramidale to attack by Coptotermes acinaciformis (Blattodea: Rhinotermitidae) in an Australian context. Int Biodeterior Biodegrad 137: 153-157. DOI 10.1016/j.ibiod.2019.01.003.

LUCARINI M, DURAZZO A, SÁNCHEZ DEL PULGAR J, GABRIELLI P & LOMBARDI-BOCCIA G. 2018. Determination of fatty acid content in meat and meat products: The FTIR-ATR approach. Food Chem 267: 223-230. DOI 10.1016/j.foodchem.2017.11.042.

LUNKENHEIMER K, CZICHOCKI G, HIRTE R & BARZYK W. 1995. Novel results on the adsorption of ionic surfactants at the air/water interface - sodium-n-alkyl sulphates. Colloids Surfaces A Physicochem Eng Asp 101: 187-197. DOI 10.1016/0927-7757(95)03157-9.

MATUURA R, KIMIZUKA H & YATSUNAMI K. 1959 The study of the adsorption of detergents at a solution-air interface by radiotracer method. III. The effect of inorganic electrolytes on the adsorption of sodium dodecyl sulfate. Bull Chem Soc Jpn 32: 646-650. DOI 10.1246/bcsj.32.646

OLIVEIRA NNFC, GALVÃO AS, AMARAL EA, SANTOS AWO, SENA-FILHO JG, OLIVEIRA EE & TEODORO AV. 2017. Toxicity of vegetable oils to the coconut mite Aceria guerreronis and selectivity against the predator Neoseiulus baraki. Exp Appl Acarol 72: 23-34. DOI 10.1007/s10493-017-0134-x.

OLUDAIRO OO, AIYEDUN JO, ADEYI AJ, AYENI AMJ & AUNGBIAS AA. 2016. Control and prevention of biodeteriation caused by termites (Isoptera). J Res For Wildl Environ 8: 44-50.

PETELSKA AD & FIGASZEWSKI ZA. 2011. The equilibria of phosphatidylcholine-fatty acid and phosphatidylcholine-amine in monolayers at the air/water interface. Colloids Surf B 82: 340-344.DOI 10.1016/j.colsurfb.2010.09.006.

QIU L, WANG H & WANG X. 2018. Heptachlor degradation characteristics of a novel strain and its application. Water Sci Technol 77: 2113-2122. DOI 10.2166/wst.2018.127.

RASIB KZ & AIHETASHAM A. 2016. Constituents and termiticide potential of some wood extracts against Coptotermes heimi (Wasmann) (Isoptera: Rhinotermitidae). Turkish J Entomol 40: 165-174. DOI 10.16970/ted.94447.

REZNIK GO ET AL. 2010. Use of sustainable chemistry to produce an acyl amino acid surfactant. Appl Microbiol Biotechnol 86: 1387-1397. DOI 10.1007/s00253-009-2431-8.

ROSEN MJ & KUNJAPPU JT. 2012. Surfactants and Interfacial Phenomena, 4th ed, John Wiley & Sons Ltd., New York, 946 p.

ROSZAINI K, AZAH MAN, MAILINA J, ZAINI S & FARIDZ ZM. 2013. Toxicity and antitermite activity of the essential oils from Cinnamomum camphora, Cymbopogon nardus, Melaleuca cajuputi and Dipterocarpus sp. against Coptotermes curvignathus. Wood Sci Technol 47: 1273-1284. DOI 10.1007/s00226-013-0576-1.

RUSLAN NAAA, SUK VRE, MISRAN M & CHIA PW. 2020. Highly efficient and green approach of synthesizing carboxylic acids from aldehydes using sodium hexametaphosphate. Sustain Chem Pharm 16: 100246. DOI 10.1016/j.scp.2020.100246.

SAXENA N, PAL N, DEY S & MANDAL A. 2017. Characterizations of surfactant synthesized from palm oil and its application in enhanced oil recovery. J Taiwan Inst Chem Eng 81: 343-355. DOI 10.1016/j.jtice.2017.09.014.

SIMS SR, BALUSU RR, NGUMBI EN & APPEL AG. 2014. Topical and vapor toxicity of saturated fatty acids to the German Cockroach (Dictyoptera: Blattellidae). J Econ Entomol 107: 758-763. DOI 10.1603/EC12515.

SREENU M, NAYAK RR, PRASAD RBN & SREEDHAR B. 2014. Synthesis, surface and micellar properties of sodium N-oleoyl amino acids. Colloids Surfaces A Physicochem Eng Asp 449: 74-81. DOI 10.1016/j.colsurfa.2014.02.037.

STOREY RA & YMÉN I. 2011. Solid State Characterization of Pharmaceuticals, illustrate. John Wiley & Sons Ltd., New York, 528 p.

SU NY & SCHEFFRAHN RH. 2000. Termites as Pests of Buildings. Termit Evol Soc Symbio Ecol 437-453. DOI 10.1007/978-94-017-3223-9_20.

SUK VRE, KHALID K & MISRAN M. 2019. Preparation and characterization of ylang-ylang (Cananga Odorata) essential oil and ascorbic acid loaded olive oil-in-water emulsion. Chiang Mai J Sci 46: 353-360.

SUK VRE, LATIF FM, TEO YY & MISRAN M. 2020. Development of nanostructured lipid carrier (NLC) assisted with polysorbate nonionic surfactants as a carrier for l-ascorbic acid and Gold Tri.E 30. J Food Sci Technol 57: 3259-3266. DOI 10.1007/s13197-020-04357-x.

SUK VRE & MISRAN M. 2017. Development and characterization of DOPEPEG2000 coated oleic acid liposomes encapsulating anticancer drugs. J Surfac Deterg 20: 321-329. DOI 10.1007/s11743-016-1914-8.

TADROS T. 2013. Gibbs Adsorption Isotherm. In: Tadros T (Eds) Encyclopedia of Colloid and Interface Science. Springer, Berlin, Heidelberg. DOI 10.1007/978-3-642-20665-8_97.

TAN HW & MISRAN M. 2013. Polysaccharide-anchored fatty acid liposome. Int J Pharm 441: 414-423. DOI 10.1016/j.ijpharm.2012.11.013.

TEO YY, MISRAN M, LOW KH & ZAIN SM. 2011. Effect of unsaturation on the stability of C18 polyunsaturated

fatty acids vesicles suspension in aqueous solution. Bull Korean Chem Soc 32: 59-64. DOI 10.5012/bkcs.2011.32.1.59.

TOUGHAN MM, SALLAM AAA & EL-LATIF AOA. 2017. Pyrethroid resistance and its mechanism in field populations of the sand termite, Psammotermes hypostoma Desneux. World Acad Sci Eng Technol Int J Bioeng Life Sci 11: 699-704.

UGBOMEH AP & DIBOYESUKU AT. 2019. Studies on termite infestation of buildings in Ase, a rural community in the Niger Delta of Nigeria. J Basic Appl Zool 80: 1-7. DOI 10.1186/s41936-019-0100-8.

XIA J, XIA Y & NNANNA IA. 1995. Structure-Function Relationship of Acyl Amino Acid Surfactants: Surface Activity and Antimicrobial Properties. J Agric Food Chem 43: 867-871. DOI 10.1021/jf00052a004.

YAP YH, AZMI AA, MOHD NK, YONG FSJ, KAN SY, THIRMIZIR MZA & CHIA PW. 2020. Green Synthesis of Silver Nanoparticle Using Water Extract of Onion Peel and Application in the Acetylation Reaction. Arab J Sci Eng 45: 4707-4807. DOI 10.1007/s13369-020-04595-3.

YEA DN, LEE SM, JO SH, YU HP & LIM JC. 2018. Preparation of environmentally friendly amino acid-based anionic surfactants and characterization of their interfacial properties for detergent products formulation. J Surfac Deterg 21: 541-552. DOI 10.1002/jsde.12051.

ZHANG GJ, CHAI CX, TAN TT, XU BC, ZHOU YW, LIU HQ, ZHAO L & WANG N. 2016. Green synthesis and surface properties of acyl glycine surfactants derived from vegetable oils. Ten Surfac Deterg 53: 284-290. DOI 10.3139/113.110435.

How to cite

SAZALEE SNF, RUSLAN NAAA, NORDIN N, AZMI WA, SUK VRE, MISRAN M, YONG TS, TEIK KK & CHIA PW. 2022. Synthesis of *N*-acyl glycine surfactant from palm oil as green repellent and toxicant to termite (*Microcerotermes diversus*). An Acad Bras Cienc 94: e20201601. DOI 10.1590/0001-3765202220201601.

Manuscript received on October 09, 2020;
accepted for publication on September 02, 2021

SYAMIMI N.F. SAZALEE[1]
https://orcid.org/0000-0003-1452-7201

NUR A.A.A. RUSLAN[1]
https://orcid.org/0000-0001-7196-1448

NURHAMIZAH NORDIN[1]
https://orcid.org/0000-0003-2896-5990

WAHIZATUL A. AZMI[1]
https://orcid.org/0000-0003-2713-815X

VICIT R.E. SUK[2]
https://orcid.org/0000-0002-7960-459X

MISNI MISRAN[2]
https://orcid.org/0000-0002-9849-9190

TAN S. YONG[3]
https://orcid.org/0000-0002-2742-7181

KOAY K. TEIK[3]
https://orcid.org/0000-0001-6415-7646

POH W. CHIA[1,4]
https://orcid.org/0000-0001-5462-4773

[1]Universiti Malaysia Terengganu, Faculty of Science and Marine Environment, Eco-Innovation Research Interest Group, 21030 Kuala Nerus, Terengganu, Malaysia

[2]Research & Development Centre, KL-Kepong Oleomas Sdn. Bhd., Lot 1 & 2, Solok Waja 3, Bukit Raja Industrial Estate, PO Box 83, 41710 Klang, Selangor, Malaysia

[3]Green2U Solution Sdn Bhd, 41-1, Jalan Puteri 5/7, Bandar Puteri, 47100 Puchong, Selangor, Malaysia

[4]Universiti Malaysia Terengganu, Institute of Marine Biotechnology, 21030 Kuala Nerus, Terengganu, Malaysia.

Correspondence to: **Poh Wai Chia**
E-mail: pohwai@umt.edu.my

Author contributions

Syamimi Nur Fatin Sazalee, Nur Aimi Aqilah Ahmad Ruslan and Nurhamizah Nordin: Investigation, Data curation. Syamimi Nur Fatin Sazalee and Poh Wai Chia: Data curation, Roles/Writing - original draft. Tan Soon Yong, Koay Kean Teik, Wahizatul Afzan Azmi, Vicit Rizal Eh Suk and Misni Misran: Sample preparation, Resources.