Preparation of Ceramic Support Coating Tin (II) Octoate: Effect of Ethylene Glycol Solvent and Dimethyl Formamide Solvent

Abdul Muiz Abd Jalil, Amira Nadzirah Suhaidi, Dr Norliza Ibrahim

Faculty of Chemical Engineering, Universiti Teknologi Mara

Abstract—**Tin (II) Octoate usually used as catalyst in condensation polymerization in production of Polylactic acid (PLA). Problem comes as the phase of catalyst same as product and reactant which made catalyst difficult to be recycle and if this catalyst remain in product PLA plastic, the plastic quality will be affected. Experiment has been done to immobilize the Tin (II) Octoate in form of sol gel before coated on solid support which is ceramic. Parameters are chosen to determine the coating which are effect of Ethylene Glycol (EG) and Dimethyl Formamide (DMF) on the preparing the coating solution and this structure are characterized by Adhesion test and Field Emission Scanning Electron Microscopy Energy Dispersive X- ray (FESEM-EDX). The result showed that the coating layer that using Dimethyl Formamide (DMF) as a solvent contain about 36.96% tin (Sn) element which higher that the coating used Ethylene Glycol as solvent which only 11.43% content of tin (Sn) element. This showed that Dimethylformamide solvent is more efficient to be used as solvent and produce better coating characteristic in term of catalyst distribution and strength of catalyst attachment on ceramic support.**

Keywords— Tin (II) Octoate, dip sol gel coating and immobilization of catalyst on ceramic support.

I. INTRODUCTION

Tin (II) Octoate is metal catalyst which is widely in plastic production especially in polymerization process. This catalyst is used in polymerization of Lactic acid (LA) to produce Polylactic acid (PLA) plastic. Polylactic acid is a polymer which compostable which has high potential to reduce societal solid waste disposal problem. Currently Polylactic acid is used in production of food packaging due to low toxicity level which suitable and safely used by consumer [15]. Since this process can achieve high molecular weight made this process very popular in polymer industry. This is because catalyst Tin (II) Octoate can act as an initiator which can provide high reaction velocity and high transformation [20].

Even the reaction can achieve high yield of plastic, this process has some disadvantage which comes from the catalyst. In previous invention of industrial process, this catalyst is mixed together with raw material both exist in liquid form which made homogenous phase with the reactant Lactic acid (LA) result to difficulties to remove the catalyst at the end of process and harder to recycle the catalyst. The products need to be purified through purification process in order to separate catalyst and product which made the process more complicated and expensive [14].

This residue will reside in the plastic product and affect the structure of the plastic product. Besides that, Tin (II) Octoate can be classified as heavy metal and processing this catalyst will contribute to creation of waste which can lead to disposal problem. Before this, there a study which has been introduce as an alternative way in handling problem as usage of catalyst can cause corrosion and toxicity. The study is about strategies is by immobilizing homogenous catalyst on solid support which known as heterogenization process [25]. Immobilizing homogenous catalyst actually can help isolation of surface active species to prevent agglomerization of catalyst and made the catalyst can be used in longer period. Besides that, immobilizing catalyst on support can made the process more efficient as the product does not need purification process which made process more easy and realistic [16]. So this experiment is running based on immobilization method which forming a coating to immobilize Tin (II) Octoate on solid support. This approach has been chosen as the immobilization catalyst on solid support so that the catalyst usage can be recycled [19]. The objective of this experiment is to know the effect of solvent used to make gel of Tin (II) Octoate solution and the characterization of the coating on ceramic support. The sol gel will be coated on ceramic surface as ceramic has high ratio of surface to volume in order to achieved high catalyst density. The ceramic surface contain fine pore which provide surface attachment and this increase the effectiveness as the pores increase surface area.

Then proceed to main goal which to determine the effect of solvent on preparing the sol gel solution so that the catalyst can be immobilize on ceramic support. The solvent act as homogenizing the solution, control drying behavior of coating and optimize flow properties so that the solution can be distributed equally on surface coating. Besides that, solvent can be used to improve pigment wetting and homogenize the solution mixture [23]. In this study, Ethylene Glycol (EG) and Dimethyl Formamide (DMF) are used as a solvent in preparing sol gel solution containing catalyst before coated on ceramic.

Ethylene Glycol (EG) is organic solvent that usually used main ingredient in antifreeze solution. The characteristic of Ethylene Glycol (EG) is colourless and has molecular weight around 62.07 g/mol. This solvent has high boiling point which 197 °C which made the solvent more suitable in coating application. Solvent with higher boiling point will create coating thickness and tend to produce coating having smoother and more uniform appearance [3]. Alternative solvent is Dimethyl Formamide (DMF) which solvent that exist as clear liquid with amine odor. This liquid has low vapor pressure and good solubility for very wide range of substances. This solvent also has high boiling point which about 153 °C. Besides that, this solvent can dissolve in water and has potential to enhance uniformity and coverage monolayer of coating particle on large surface [4].

The solvent efficiency will be decided on coating structure in term of catalyst distribution, surface attachment and strength of coating. This experiment will determine the potential of these two

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solvent in order to produce a better sol gel coating solution which can produce well distribute catalyst concentration on surface coating and the strong attachment of coating layer.

II. METHODOLOGY

A. Materials

This section should explain procedures and the methods that you completed within your study. Be sure to describe your participants, measurements, and, any treatments, training sessions, or protocols that were followed with the scope of your research. Your method section should end with a description of any data analysis that was carried out.

B. Preparation of waste power from ceramic waste

The raw sample ceramic waste obtained is already in granular formed and placed in plastic container. The ceramic granule size is about more than 150 μm. The ceramic granule was grinded by using grinder for 3 minutes. After the grinding process, the ceramic powder was separated by using sieving process. The sieve tray size was chose which are 150 μm and 125 μm. Then this tray was cleaned by using vacuum cleaner (Model SC-301N) and Swan air compressor. Then ceramic powder was separated by using Endecotts Octagon 2000 Dgital Sieve Shaker for 10 miutes duration in order to obtain 125 μm ceramic powders.

C. Preparation of paste for fabrication of ceramic support

60 grams of fine ceramic powder was weighed by using analytical balance. Then 20 ml of diluted weight PVA solution was measured by using measuring cylinder. After that, both components was mixed gradually in a metal bowl. The powder need to be added into the bowl until the mixture creating a dough form. Next was compression process which the dough mixture compressed through compression machine. First male and female mold was cleaned using plain water to removed dirt from mold surface. Then female mold was placed on plate surface in the machine and dough mixture was placed inside the female mold ring. Then the opening ring was closed with male mold and both slightly compressed by rotating screw clamp. After that, valve on the left side was closed by rotating the valve clockwise direction. The hydraulic pump pressed until the pressure reach 150 kg/m^2 and after that the valve on left side is released. Then rotate the screw screw clamp to take out the mold and the mold was placed in inverted position. A screw was placed on top of the mold and pressed by the screw clamp to obtain the ceramic coin shape sample. This sample was dried in oven for 24 hours at 110°C. After that, the sample sintered in a furnace for 2 hours at 1100 °C.

D. Preparation of thin layer Tin (II) Octoate on fabricated ceramic support

The method used to prepare the thin layer Tin (II) Octoate by sol gel dip coating. This sol gel layer was prepared using Tin (II) Octoate as catalyst, Ethylene Glycol and Polyvinyl Alcohol (PVA) as a binder. 10 ml Tin (II) Octoate which mixed 100 ml of Ethylene Glycol (EG) and this solution was stirred for 30 minutes. Then the solution mixed with 1 ml with nitric acid and stirred at temperature 90 °C for 30 minutes. After that, the solution mixed with 10 ml binder which is Polyvinyl Alcohol (PVA) for 6 hours then the solution is left for aging phase for 24 hours at room temperature to produce sol gel solution. The ceramic was transferred into petri dish with specific volume of Tin (II) Octoate solution to coat the surface of ceramic with sol gel solution. This process was repeated by using alternative solvent which replaced solvent Ethylene Glycol with Dimethyl Formamide (DMF) to produce sol gel solution. Then after the ceramic coated with solution, the ceramic was sintered at 200 °C - 600 °C to form a thin layer of coating.

III. RESULTS AND DISCUSSION

A. Morphology of Tin (II) Octoate coating on ceramic surface

After the Tin (II) Octoate has been coated on ceramic support, the coated layer has been analysed in order to determine the structure of coating layer. The structure can be seen perfectly by using field emission scanning electron microscopy (FESEM).
In this study, two sample have been analysed which were sample one using Ethylene Glycol (EG) as solvent and sample two using Dimethylformamide (DMF) as solvent. Figure 1 below showed the detail structure of coating layer which using Ethylene Glycol as solvent which view under FESEM machine:

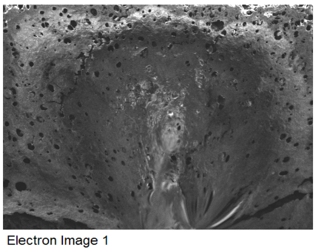

Figure 1: Structure of Tin (II) Octoate coating on ceramic surface using Ethylene Glycol

Figure 2 below showed the detail structure of coating layer which using Dimethylformamide as solvent which view under FESEM machine:

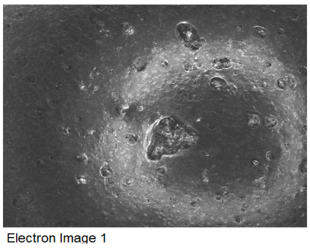

Figure 2: Structure of Tin (II) Octoate coating on ceramic surface using Dimethylformamide

Based on the result obtained, the structure of both sample have been analysed and observed. The structure of Tin (II) Octoate coating on ceramic surface using Dimethylformamide in figure 2 look more smooth and clean compared to structure from figure 1 which the coating layer produce using Ethylene Glycol as solvent.

This showed that surface coating layer is affected by the usage of the different solvent which Dimethylformamide (DMF) and Ethylene Glycol (EG). The viscosity of each solvent will play an important role in creating better coating layer. The viscosity for Dimethylformamide (DMF) is 0.79 lesser than the Ethylene Glycol which is 16.1. In order to create smooth coating surface, the viscosity of solvent liquid must be located in range between 0.5 to 5 [27]. Since the viscosity of Dimethylformamide solvent is in range compared to viscosity Ethylene Glycol made the Dimethylformamide able to form a better coating layer. Besides that, Dimethylformamide solvent has an ability to enhance uniformity and coverage monolayer of coating particle on large surface area which made solvent able to produce better coating surface [26].

B. Distribution and concentration of Tin (II) Octoate coating layer

The experiment also focused on the distribution and concentration of catalyst Tin (II) Octoate on coating layer. The presence of Tin (II) Octoate catalyst can be traced by energy dispersive X-Ray (EDX) machine. Two sample using different solvent which Dimethylformamide and Ethylene Glycol were characterized in order to trace the presence of catalyst Tin (II) Octoate.

First sample consist of coating layer which used Ethylene Glyol was characterized by using energy dispersive X-Ray (EDX) and the result is represented by the figure 3 below:

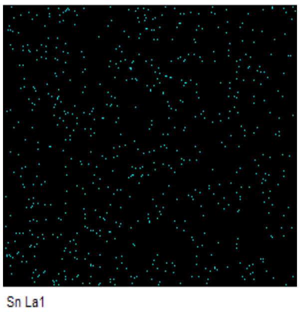

Figure 3: Distribution of component Sn contain in coating layer by using Ethylene Glycol

Figure 4 below showed distribution of component Sn contain in coating layer by using Dimethylformamide as a solvent:

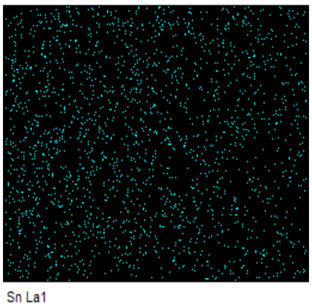

Figure 4: Distribution of component Sn contain in coating layer by using Dimethylformamide

Based on the figure 3 and 4, the result obtained showed that several element existed in coated layer such as carbon, oxygen, silica and tin component. Both figure 3 and 4 illustrate the composition carbon which contain in the coating layer and stated as a highest compared to other element. This carbon element actually is comes from material used to prepared the coating solution such as Tin (II) Octoate, Polyvinyl Alcohol (PVA), Ethylene Glycol (EG) and Dimethylformamide (DMF). All these material contribute to carbon composition in the coating layer which detected by the energy dispersive microscopy (EDX).

Same goes for oxygen component presence in coating layer but not as high compared to carbon element. Oxygen element also is contributed from coating material such Tin (II) Octoate, Dimethylformamide (DMF), Polyvinyl Alcohol (PVA), Ethylene Glycol (EG) and Nitric acid. Actually the oxygen component and tin (Sn) element related to each other as both component are importance in creating the coating layer. This is because this experiment objective is to immobilize the tin (Sn) element on ceramic surface through a coating layer. But to immobilize the tin (Sn), a bond must be formed in order to bind the tin (Sn) element so that the created coating layer that attach on the ceramic support has the tin (Sn) component. This bond referred to chemical bond which formed from peptization process and this bond related to the oxygen element.

From this figure 4, tin (Sn) element distribution is differ for each solvent used and from the observation tin (Sn) distribution on coating which using Dimethylformamide as a solvent has high intensity of tin (Sn) compared to coating using Ethylene Glycol as a solvent. This result actually related to peptization bond and at the same time related to the oxygen component presence in the coating layer. If oxygen component concentration is high on coating layer, the tin (Sn) concentration which presence in the coating also high.

Oxygen content in the coating layer which using Ethylene Glycol solvent is low compared coating layer which using Dimethylformamide and this result will affect the tin (Sn) distribution which stated in the figure 4 coating layer which using Dimethylformamide is contain more tin (Sn) about 36.96% compared to coating layer which using Ethylene Glycol solvent which stated only 11.43% tin (Sn) presence in the coating.

Based on the previous study, researcher made an experiment on formulation to prepare the coating solution in form of sol gel [32]. The study is more focused on the ratio of material used like Tin (II) Octoate catalyst, Polyvinyl Alcohol (PVA) binder and Ethylene Glycol as solvent used. Besides that, these three material will be mixed with Nitric acid (HNO_3) and each portion must in correct amount so that the sol gel can produce. According to Rafizah Zaiton (2017), the catalyst, binder and Nitric acid added made the solution undergoes hydrolysis at pH 2.8 and mixed with binder Polyvinyl Alcohol (PVA) which proceed to evaporation and condensation to form sol gel at pH 4.

The critical part is the portion of each material must in correct amount and the role of Nitric acid to convert the solution into sol gel appearance. The Nitric acid is importance in creating sol gel as this acid acts as peptization agent [29]. This peptization process actually create a bond which known as oxygen covalent bonding and this bond responsible to hold the tin (Sn) element on the coating layer. This bonding also same like other research which attaching aluminium alkoxide through oxygen covalent bonding [28]. Oxygen component will act to bind the tin (Sn) through the covalent bonding. This statement also support the result which showed if the element of oxygen presence is large amount, so more tin (Sn) component can be found in the coating like in figure 4.

The solvent used in this study also has specific important role in creating the coating layer on ceramic support. Solvent works to make the coating can distributed equally on the ceramic surface while retaining the tin (Sn) content on the coated layer. In order to form a uniform thickness of coating, solvent with high boiling point will choose like Ethylene Glycol and Dimethylformamide since solvent will affect the rate vaporization [31]. Besides that, the solvent effect also can be seen on the distribution of tin (Sn) element on the coating surface. From figure 4, the distribution of tin (Sn) for coating that using Dimethylformamide look more uniform than coating that using Ethylene Glycol solvent. This statement is support by the role of viscosity of the solvent and the range usually in between 0.5 to 5 in order to create smooth coating surface [27]. So if the coating thickness is uniform, the distribution of tin (Sn) content in the coating layer also will be automatically uniform.

In addition, the type of solvent used also importance in obtaining smooth coating and the coating surface is free from cracking after sintering. The solvent characteristic will influence the formation of the layer when applying sol gel solution on the ceramic support. Some research has been done on the effect on solvent in creating coating layer and the research briefly explained about Dimethylformamide solvent effect on coating layer formation. The result from the research found that the Dimethylformamide solvent made the sol gel solution has smaller pore size which avoiding the surface of coating layer became cracking after drying and sintering [33]. The research also explained that by increasing pore size of the gel can lead to increase the permeability and reduce drying stress but at the same time large pores formation in the coating layer can lead to the coating became weak and crack easily [30].

Lastly is about silica (Si) element which has been detected along with the coating layer for both sample. This silica (Si) component can be classified as trace element as the amount of silica is too minimum compared to other element in the coating layer. This silica component actually is contribute from the ceramic support as the support is comes from ceramic waste. This waste may contain trace element such as silica (Si) due the support has been use previously as a support for many reaction. The ceramic waste is collected previously and grinded in order to form ceramic powder before mix with the binder to produce ceramic support which used in this experiment which act as material to be coated to attach tin (Sn) element.

C. Attachment and strength of Tin (II) Octoate coating layer on ceramic support

Then proceed to the adhesion test which applied to both sample. This test is important in order to measure the strength of coating layer attachment on the ceramic support. Basically the strength of coating layer is depend on one factor which help the coating attach on support which is pore of the ceramic support and the pore on the surface of the ceramic also contribute to the attachment of the coating.

The adhesion test is ran by using standard method which stated in test method D3359 which require razor blade and pressure sensitive tape. An X cut is made on the ceramic support by using razor blade in order to create the square area which later will be test by using pressure sensitive tape which to extract the data on the attachment strength and the step is illustrated on the figure 5 below:

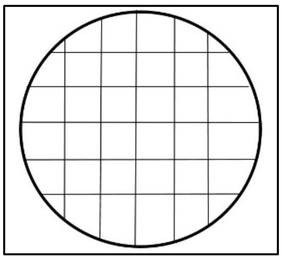

Figure 5: X cut is made on sample of the ceramic support

The result from the adhesion test for the ceramic support which has coating layer using the Ethylene Glycol as solvent and the result can be seen on the figure 6 below:

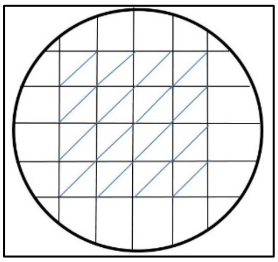

Figure 6: Adhesion test for ceramic support which has coating layer using the Ethylene Glycol as solvent

Based on the result figure 6, adhesion test showed the result which about all the square which draw by the razor knife is pulled out by the tape. The square all together has 16 square box and the result is 16/16 is wipe out by the pressure sensitive tape. If the result compared to the standard on the adhesion test in test method D3359, this result is classified in 0B. 0B classification which detachment occur is more than 65% and this flaking detachment is worse than grade 1. From the result, the strength of coating layer which used Ethylene Glycol is completely low and easily to be detach.

The result from the adhesion test for the ceramic support which has coating layer using the Dimethyformamide as solvent and the result can be seen on the figure 7 below:

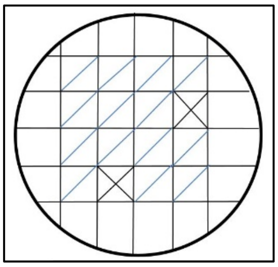

Figure 7: Adhesion test for ceramic support which has coating layer using the Dimethylformamide as solvent

Based on the figure 7, adhesion test showed the result which almost all the square which draw by the razor knife is pulled out by the tape but not all detach from the surface. The square all together has 16 square box and the result is 14/16 is wipe out by the pressure sensitive tape. If the result compared to the standard on the adhesion test in test method D3359, this result is classified in 0B. 0B classification which detachment occur is more than 65% and this flaking detachment is worse than grade 1. From the result, the strength of coating layer which used Dimethylformamide is completely low and easily to be detach but if compared to Ethylene Glycol, Dimethylformamide has better attachment since not all the square is detach from surface ceramic support.

IV. CONCLUSION

In conclusion, this experiment has conducted and considered as a success in characterize the coating layer which used as a method to immobilize the Tin (II) Octoate catalyst on ceramic support by using field emission scanning electron microscopy – energy dispersive X – Ray (FESEM-EDX). The objective of this study was achieved which produce ceramic support coating by using different type of solvent which are Ethylene Glycol and Dimethylformamide. These coating sample has been characterized on the effect of solvent on the coating layer, distribution of the tin (Sn) component and attachment strength by using FESEM-EDX and adhesion test. This research showed that Dimethylformamide is suitable solvent used in immobilizing Tin (II) Octoate compared to Ethylene Glycol. The coating layer contain about 36.96% of tin (Sn) for coating that using Dimethylformamide solvent is higher compared to Ethylene Glycol which stated only 11.43% tin (Sn) presence in the coating. In term of attachment, coating layer which using Dimethylformamide as solvent is provide better attachment strength compared coating layer which using Ethylene Glycol as solvent.

ACKNOWLEDGMENT

Thank you to my supervisor and Universiti Teknologi Mara. I would like express my gratitude to my supervisor, Dr Norliza Binti Ibrahim for her guidance and assistance in helping me to complete this research project successfully. Her encouragement and enthusiasm in me throughout actually increasing my motivation in doing this research. She always knew where to look for the answers to obstacles while leading me to the right sources, theories and perspectives. Other than that, I also wish to express my appreciation to and Miss Amira Nadzirah Binti Suhaidi who always assist my research project to be in the right path and with her guidance which helped me a lot in completing this research project. Here I would like to thank all the researcher and contributors to this particular field because without their previous researches, I would not able to complete this research due to lack of information and I hope that I can be part of them contributing to this subject matter. Lastly I would like to express my gratitude to my family member and my friends which give morale support in completing this task. And most of all to Allah SWT, who gave me strength and knowledge to do this project, the perseverance and determination to continue doing this project in spite of struggles that come along the way.

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