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Broad Exposure to Science and Technology 2019 (BEST2019) IOP Conf. Series: Materials Science and Engineering 673 (2019) 012016 IOP Publishing doi:10.1088/1757-899X/673/1/012016 1 Optimization of biodegradable plastic production using response surface methodology M Riza1\*, S Syaubari1, A Andriansyah2, R Dewi3 and L Ernita1 1 Chemical Engineering Department, Universitas Syiah Kuala, JI. Tgk. Syech Abdul Rauf No.7

Banda Aceh 23111 Aceh, Indonesia <mark>2 Laboratory of Industrial Computation and Optimization, Industrial Engineering, Universitas Syiah Kuala, Jl. Tgk. Syech Abdul Rauf No.7 Banda Aceh 23111 Aceh, Indonesia 3 Chemical Engineering Department, Universitas Malikussaleh Lhokseumawe, Aceh, Indonesia \*email: medyan\_riza@unsyiah.ac.id Abstract. Optimization is a searching technique for variable values considered optimum, e ? ective and e ? cient, in order to reach the desirable results.</mark>

An experiment was carried out to see the relationship between response variable and the independent variable. Several comparison tests can be obtained to gain the level that will create optimal response. The aim of this research is to apply the Response Surface methodology in order to obtain the optimum condition for the process variable in producing biodegradable plastic with lemongrass as antioxidant.

Fixed variables in this research are tapioca starch, 69-79oC gelatinization temperature and the total mixture weight that consists of starch, poly(NIPAM) chitosan, lemongrass oil, acetic acid and water. The response variables are tensile strength and break of elongation of each biodegradable plastic that is produced. The application of response surface methodology on each biodegradable plastic can be used to obtain the

## independent variable that makes the optimal response variable.

The predicted result of the optimum value for tensile strength and elongation of break lies on the composition treatment of Poly(NIPAM)-chitosan 0.35 grams, glycerol 3.5 grams and 36,55% lemongrass oil with tensile strength result of 3.98 MPa and elongation of break of 36.55%. 1. Introduction The biggest waste around the world today is from plastic-based materials [1].

Plastic is commonly used in food packaging, packaging from household items as well as in other sectors. Plastic is a kind of material that is not easy to be degraded by microbes. The number of plastic waste is increasing every year and can cause many problems, such as environmental pollution, the spreading of diseases and also to?o rolem[2Many ? orts have been done to tack Ithe lastwe ?s,they e: recycling plastic and burning plastic.

However on there is only a few percentage of plastic that can be recycled; therefore another alternative solution is needed to handle this situation. Biodegradable plastic can be commonly interpreted as plastic that can be recycled and can be degraded naturally. Conventional plastic is plastic that is made up of petroleum, coal and natural gas.

Meanwhile biodegradable plastic is plastic that is made up of natural polymers such as cellulose (starch), collagen, casein, protein or lipid [3]. Biodegradable plastic is a material that its chemical structure can change under certain circumstances; in which microorganisms such as, bacteria, fungi and algae, can a ? ect its properties [5].

Starch is the main material in producing biodegradable plastic, because starch Broad Exposure to Science and Technology 2019 (BEST2019) IOP Conf. Series: Materials Science and Engineering 673 (2019) 012016 IOP Publishing doi:10.1088/1757-899X/673/1/012016 2 has biodegradable properties, easier processing and has an economical value.

Starch produces materials such as, cassavas, bananas, corns, potatoes and others are easily available in Indonesia [4]. Many researchers have utilized starch as main material in producing biodegradable plastic. One of them is biodegradable plastic that is made up of starch, chitosan and glycerol mixture [2].

Glycerol fnto?xthlao lastip roperties that are starch- bedwhh elac,?iba oth. Glycerol has a low molecular weight so that it can be considered to be used as plasticizer [4]. Chitosan is an additional material in producing biodegradable plastic that can function to ?xthe nsareno plastic produced [5].

Chitosan contains antioxidants and antimicrobials that can be used as substances for packaging [6]. Chitosan is a substance that can inhibit oxygen on the packaging [7]. Chitosan also functions as preservative fr iodabpc.Csapps wat -resistant, non-toxic and anti-bacterial that can function as preservatives and anti-microbial.

N-isopropylacrylamide (NIPAM) is an organic compound that can be synthesized to become polymer or can be crosslinking polymer that is biodegradable in nature. NIPAM is one of the acrylamide monomers that are white, odourless and shaped as solid crystals that are soluble in water and very reactive through amide reaction or by its duplicate bonds. This compound can make a long polymer chain that is called poly(NIPAM).

Poly(NIPAM) is a thermo sensitive material that can be shaped into hydge aplican fply(NAM) a t een oin u?eld ch s,actinas electrophoresis membrane, bio mical emrane,?olatcasarasos dliqs n liquid waste processing, microsphere synthesis for drug transmission, thickener or to create other copund o he ageo oIPhasor e?nt arariss,soth there are orars hat ope n rdto?xthe e?encs.Oo ding roing h other monomers. Poly(NIPAM) is a smart polymer that is susceptible to change in temperatures.

Bse o tha cicoi?cn a me r of crosslinking towards poly(NIPAM) and chitosan in the process of producing biodegradable plastic is hoped to increase the physical quality, thermal and plastic degradation in the plastic produced. The addition of poly(NIPAM) in the production of biodegradable plastic can create a stronger plastic structural matrix.

In order to understand how far an optimum process is a ? ected by a number of variables, a large amount of experiment data is needed, where it requires a long time to be obtained automatically and it also requires a large cost. There are several mathematical and statistical techniques that are used to as approaches in order to gain understanding of the optimum condition of a process, without the need of mda.Oo technis espnssurfe etdlo(R] articial ura network to create a mathematical model for biodegradable plastic.

Cinnamon oil is added in to NIPAM- chitosan and glyserol. [9] optimizes the results of linear and multilinear regression results using simulated annealing algorithm to obtain a best composition of biodegradable plastic produced. Almost similarly with previous works, this study investigates the combination of biodegradable plastic compositions by adding lemongrass oil.

RSM incorporates statistical and mathematical techniques that can be used to generate and analyze responses (composition if poly(NIPAM)-chitosan, glycerol and lemongrass

oil), that are a ? ected by some free variables. The RSM can also be used as an e ? ort in looking for the right function to predict responses and determine the values of independent variables that can optimize responses, with achieving the aim of optimizing responses [10]. Many researcher have used this method to obtain best results.

[11] uses the this method to optimize preparation of biocomposites based on poly and durian peel cellulose. The biodegradability of acrylic acid grafting onto polypropylene has been optimed by RSM [12]. 2. Methodology 2.1. Equipment and materials The equipment that will be used in this research to produce biodegradable plastic are, magnetic stirrer, beaker glass, Erlenmeyer, chemical glass, aluminium foil, digital weighing scale, hot plate, desiccator, casting glass, oven, stirrer stick, tweezers and petri dish.

The materials needed for this research to produce biodegradable plastic are tapioca starch, glycerol, chitosan, lemongrass oil, 1% acetic acid, Broad Exposure to Science and Technology 2019 (BEST2019) IOP Conf. Series: Materials Science and Engineering 673 (2019) 012016 IOP Publishing doi:10.1088/1757-899X/673/1/012016 3 ethanol, NIPAM monomers, distilled water, azobisisobutironiltril initiator (AIBN), and mercaptopropionic acid initiator (MPA). 2.2.

Research design This research uses Central Composite Design (CCD) and uses Design Expert version 10 three factors, they are: what is included in the RSM to see the optimum condition towards the e ? ects of treatment of the poly(NIPAM)-chitosan, glycerol and lemongrass oil treatment towards the results of tensile strength and elongation of break of the biodegradable plastic produced. The trial design of CCD can be seen in Table 1. 2.3.

Research procedure NIPAM polymerization is carried out by dissolving 10 grams of NIPAM into 20 ml of Ethanol in an Erlenmeyer. Then 1 ml AIBN is added and 0.6 ml MPA. The mixture is stirred until it is completely dissolved. The mixture is then incubated with a temperature of 60oC for 20 hours. The product produced is precipitate that can be taken by adding diethyl ether solution.

The sediment that is obtained is then dried inside the oven for 12 hours at 80oC temperature. Table 1. Limitations and change level variable/independent variables. Variable Limitations and Level -1.682 (- a) -1 0 +1 1.682 a) Pnipam-chitosan (X1) 0.11 0.35 0.45 0.55 0.93 Glycerol (X2) 0.48 1.50 2.50 3.50 5.89 Lemongrass oil (X3) 0.08 0.25 0.35 0.45 0.76 Tapioca starch (10 grams) is dissolved with 50 ml of distilled water in a chemical glass and stirred for 25 minutes.

Then when the mixture of starch and distilled water is homogenous, the starch mixture is placed on top of a hot plate. Hot plate is used to speed up the reaction by increasing the temperature. Magnetic stirrer was used to stir the mixture in a homogenous form. Magnetic stirrer was used to av oid lumps within the starch during heating and to ensure even heat distribution.

The starch is heated until it reaches 70 oC in temperature for 20 minutes. Next, glycerol plasticizer, poly(NIPAM)-chitosan and lemongrass oil is added into the starch. During the addition of glycerol, the poly(NIPAN)-chitosan, lemongrass oil and starch mixture must be kept stirred for 15 minutes to avoid lumps and speed up the mixture homogenization between the starch and glycerol, as well as the chitosan and lemongrass oil.

After the mixture thickens, the chemical glass is removed from the hot plate. The solution should be kept stirred until it reaches normal temperature around 25 – 30oC for 30 minutes to keep the thickness of the mixture stable. After the temperature of the starch solution is normal, then casting is conducted on top of a glass plate with 1mm thickness that has been given tape on the sides.

The purpose of the tape is to keep the solution intact within the glass plate. The pouring of the starch solution has to be done slowly and carefully. The thin later that is formed on top of the glass plate is entered into an oven with a temperature of 80oC and kept at it for 4 hours to harden and dry. The thin layer from the glass plate is then removed and kept inside a desiccator for a day in a place with no direct sunlight. 3.

Result and Discussion This research is conducted with 19 treatments where X1 variable for the composition of Poly(NIPAM)-chitosan mixture, X2 for the composition of glycerol and X3 for the composition of lemongrass oil. The response variables are the tensile strength and elongation of break of the plastic produces. Broad Exposure to Science and Technology 2019 (BEST2019) IOP Conf. Series: Materials Science and Engineering 673 (2019) 012016 IOP Publishing doi:10.1088/1757-899X/673/1/012016 4 Table 2. Experiment data results and model prediction results towards tensile strength and elongation of break.

Run Level Value Variable X Tensile Strength (MPa) Elongation of Break (%) Pnipam-chitosan (gr) X1 Glycerol (gr) X2 Lemongrass oil (gr) X3 Data Prediction % Error Data Prediction % Error 1 0.35 1.5 0.45 2.97 3.29 -10.77 25.92 25.86 0.23 2 0.55 3.5 0.25 3.57 3.88 -8.74 31.4 25.81 17.81 3 0.45 2.5 0.08 3.86 3.94 -2.05 31.32 28.42 9.25 4 0.45 2.5 0.35 3.77 3.62 3.95 14.48 26.63 -83.94 5 0.45 0.48 0.35 3.44 3.21 6.75 6.32 9.52 -50.6 6 0.45 2.5 0.35 3.1 3.62 -16.81 25.64 26.63 -3.88 7 0.45 2.5 0.35 2.78 3.62 -30.25 25.32 26.63 -5.19 8 0.35 1.5 0.25 3.02 3.03 -0.2 18.92 10.22 45.98 9 0.35 3.5 0.25 4.08 4.21 -3.26 31.08 36.55 -17.6 10 0.45 5.89 0.35 4.24 4.82 -13.7 16.32 17.15 -5.09 11 0.45 2.5 0.35 3.73 3.62 2.92 19.96 26.63 -33.44 12 0.45 2.5 0.35 4.17 3.62 13.16 31.04 26.63 14.19 13 0.35 3.5 0.45 3.77 4.02 -6.62 53.52 41.11 23.19 14 0.93 2.5 0.35 2.79 2.89 -3.58 11.48 11.29 1.61 15 0.45 2.5 0.76 3.66 4.11 -12.33 39 41.6 -6.67 16 0.55 1.5 0.45 3.17 3.67 -15.75 34.76 23.64 32 17 0.11 2.5 0.35 2.75 3.18 -15.78 35.44 37.76 -6.56 18 0.55 1.5 0.25 3.08 3.57 -15.92 17.24 24 -39.2 19 0.55 3.5 0.45 2.79 3.52 -26.31 11.32 14.37 -26.93 From Table 2, it can be concluded that the composition of poly(NIPAM)-chitosan, glycerol and lemongrass oil have an e ect towards the values of tensile strength and elongation of break of the biodegradable plastic that is produces.

The highest tensile strength value is obtained from this experiment based on the results prediction model is from run 10, which is 4.24 MPa on the 0.45 gram poly(NIPAM)-chitosan, 5.89 grams glycerol and 0.35 gram lemongrass oil composition. Meanwhile, the lowest tensile strength value is obtained from the experiment on run 17, which is 2.75 MPa with the composition of 0.11 gram poly(NIPAM)-chitosan, 2.5 grams glycerol and 0.35 gram lemongrass oil.

The highest value for elongation of break obtained from the experiment on run 13, which is 53.52 with the composition of 0.35 gram poly(NIPAM)-chitosan, 3.5 grams glycerol and 0.45 gram lemongrass oil. Meanwhile the highest elongation of break value is obtained from the results prediction model in on experiment run 15 which is 41.60% with the composition of 0.45 gram poly(NIPAM)- chitosan, 2.5 grams glycerol and 0.76 gram lemongrass oil.

Meanwhile the lowest elongation of break value is obtained from the experiment run 5, which is 6.32% with the composition of 0.45 gram poly(NIPAM)-chitosan, 0.48 gram glycerol and 0.35 gram lemongrass oil. For the lowest elongation of break value that was obtained based on the results prediction model is the same with the one from run 5, which is 9.52%.

However for the centre point condition; 5 experiments were conducted with the composition of poly(NIPAM)-chitosan 0.45 gram, 2.5 grams glycerol and 0.35 gram lemongrass oil. The tensile strength value that was obtained on the centre point run 4 was 3.77 MPa, run 6 was 3.1 MPa, run 7 was 2.78 MPa, run 11 was 3.73 MPa and run 12 was 4.17 MPa. Meanwhile the elongation of break value obtained on the centre point on run 4 was 14.48%m run 6 was 25.64%, run 7 was 25.32%, run 11 was 19.96% and run 12 was 31.04% Tab of Probability F is bigger than 0.0500 which is 0.3056 for tensile strength model and 0.1098 for elongation of break.

The Prob > F value serves to look into the suitability between every coe cient and the strength of interaction between each independent variables. If the Prob > F value is less than F-value, ect towards the response. Broad Exposure to Science and Technology 2019 (BEST2019) IOP Conf. Series: Materials Science and Engineering 673 (2019) 012016 IOP Publishing doi:10.1088/1757-899X/673/1/012016 5 Table 3. Tensile strength analysis of variance.

Source Sum of square df Mean square F value P-value Prob>F Characteristic Model 2.68 9 0.3 1.42 0.3056 Insignificant A 0.005 1 0.005 0.024 0.881 B 0.46 1 0.46 2.17 0.1745 C 0.2 1 0.2 0.95 0.3548 A2 0.38 1 0.38 1.82 0.2103 B2 0.014 1 0.014 0.065 0.8049 C2 0.16 1 0.16 0.76 0.4063 AB 0.5 1 0.5 2.39 0.1562 AC 0.064 1 0.064 0.3 0.5955 BC 0.25 1 0.25 1.17 0.3067 Residual 1,890 9 0.21 Lack of Fit 0.64 5 0.13 0.41 0.8232 Insignificant Pure error 1.25 4 0.31 Cor Total 4.58 18 Table 4.

Elongation of break analysis of variance. Source Sum of square df Mean square F value P-value Prob>F Characteristic Model 1679.81 9 186.65 2.35 0.1098 Insignificant A 358.74 1 358.74 4.51 0.0626 B 302.3 1 302.3 3.8 0.083 C 26.59 1 26.59 0.33 0.5772 A2 300.62 1 300.62 3.78 0.0837 B2 128 1 128 1.61 0.2363 C2 61.38 1 61.38 0.77 0.4024 AB 0.039 1 0.039 0.0004 0.9829 AC 363.18 1 363.18 4.57 0.0613 BC 81.53 1 81.53 1.03 0.3376 Residual 715.43 9 79.49 Lack of Fit 557.02 5 111.4

2.81 0.169 Insignificant Pure error 158.41 4 39.6 Cor Total 2395.24 18 Table 5 and table 6 are the results of variance analysis for each tensile strength and elongation of break statistical models. It can be seen that each of the tensile strength and elongation of break break R2 values are 0.5865 and 0.7013.

Thimteidti at riabs ,X2andX3gaa ig ? ect towards the tensile strength and elongation of break models. The suggested model for tensile strength and elongation of break is quadratic model. The actual mathematical model obtained is to predict the value of tensile strength (TS) and elongation of break (EOB) is as follows: = -1, 20552 + 10, 11597 ?? 1 + 1, 506 ?? 2 + 2, 032 ?? 3 - 2, 187 ?? 1 ?? 2 - 4, 125 ?? 1 ?? 3 - 1, 412 ?? 2 ?? 3 - 3, 425 ?? 1 2 + 0, 0275 ?? 2 2 + 3, 4010 ?? 3 2 ( 1 ) ?????? = - 134, 3753 + 259, 982 ?? 1 + 51, 96163 ?? 2 + 215, 34639 ?? 3 - 61, 3 ?? 1 ?? 2 - 400 ?? 1 ?? 3 - 27, 7 ?? 2 ?? 3 + 0, 9497 ?? 1 2 - 2, 083 ?? 2 2 + 63, 4299 ?? 3 2 ( 2 ) Broad Exposure to Science and Technology 2019 (BEST2019) IOP Conf. Series: Materials Science and Engineering 673 (2019) 012016 IOP Publishing doi:10.1088/1757-899X/673/1/012016 6 Table 5. Tensile strength statistical model summary. Model source Std Dev. R-squared Adjust R-square Predicted R-squares press Linear 0.48 0.2374 0.0849 -0.434 6.57 2FI 0.49 0.3588 0.0383 -0.4637 6.70 Quadratic 0.46 0.5865 0.1729 -23.107 15.16 suggested Cubic 0.56 0.7264 -0.2313 + aliased Table 6.

Elongation of break statistical model summary. Model source Std Dev. R-squared Adjust R-square Predicted R-squares press Linear 10.84 0.2642 0.117 -0.3688 3278.62 2FI 10.30 0.4687 0.2031 -0.6956 4059.96 Quadratic 8.92 0.7013 0.4026 -1.3278 5575.73 suggested Cubic 6.29 0.9339 0.7024 + aliased Tee enh dedas mim othacawittandbpc il baks.T testg ftensile reh couc ut dilo mtoardthimon n the ib oa ndotpasts sticity.

Figure 1(a) shows the value of tensile strength increases as the composition of poly(NIPAM)-chitosan and glycerol increase. The increase of tensile strength is a ? ected by the higher number of poly(NIPAM)-chitosan used. This is because the higher the number of poly(NIPAM)-chitosan, the mire hydrogen bonds that can be found in the plastic, therefore the chemical bonds become stronger and harder to break because it will require a larger energy to break the bond [1].

Figure 1(b) shows the value of tens ilstrgth vinnosig?ccnath composition of poly(NIPAM)-chitosan and lemongrass oil composition increases. This is due to the increase of lemongrass oil can decrease the mechanical nature so that it can be easily broken. This happens because there is a dispersed oil phase within the plastic matrix that causes the plastic structure torunin isconu ros.Engan fbis eedas e erctao gtcngei the plastic when it is pulled it breaks.

The higher the value of elongation of break then the plastic is more elastic, so that the material pulled is more stretched. Plastic with lower elongation of break will be brittle in ur[8 ?re [d can e n t ngan fbwiinasas e composition of glycerol increases and the elongation of break decreases as the poly(NIPAM)chitosan composition incse is de pstier'abty ucblens ncre he exilito plastic.

Meanwhile with the increase of poly(NIPAM)-chitosan, it decreases the percentage of elongation of break. This is due to the smaller distance between the intermolecular bonds. The addition of lemongrass oil also tends to decrease the percentage of elongation break. This is due to the fact that oil is hydrophobic in nature so that it does not dissolve in the starch solution that will cause less elasticity on the plastic and can be broken easily. Broad Exposure to Science and Technology 2019 (BEST2019) IOP Conf.

Series: Materials Science and Engineering 673 (2019) 012016 IOP Publishing doi:10.1088/1757-899X/673/1/012016 7 c f Figure 1. 3D Plot of the Poly(NIPAM)-chitosan, glycerol, and lemongrass oil composition effects towards the values of tensile strength (a, b, c) and elongation of break (d, e, f). 4. Conclusion This study investigates the optimum composition of biodegradable plastic production. Lemongrass oil is used as antioxidant in the experiments. The experiments are designed using RSM to get optimal Broad Exposure to Science and Technology 2019 (BEST2019) IOP Conf. Series: Materials Science and Engineering 673 (2019) 012016 IOP Publishing doi:10.1088/1757-899X/673/1/012016 8 composition formed biodegradable plastic. The independent variables in this experiments are poly(NIPAM) -chitosan, glycerol, and lemongrass oil.

The tensile strength and elongation of break values are a ? ected by the composition of Poly(NIPAM)-chitosan, glycerol, and lemongrass oil. Based on analysis, the prediction results of optimum value of tensile strength and elongation of break depended on the composition treatment of 0.35 grams of Poly(NIPAM)chitosan, 3.5% glycerol and 36.55% lemongrass oil.

Based on the Central Composite Design (CCD) on Design Expert 10 software, it was found out that the suitable model for tensile strength and elongation of break values is a quadratic model where each of the R2 value are 0.5865 and 0.7013. Acknowledgments This research is supported by Universitas Syiah Kuala, Ministry of Research, Technology and Education of Indonesia, in accordance with Letter of Appointment Agreement of Research Professor of Fiscal Year 2019 Number: 23/UN11.2/PP/PNBP/SP3/2019 Date February 8, 2019.

Thanks and high appreciation to Rector and Head of LPPM Universitas Syiah Kuala. References [1] CniwaP Ia a ?rM 15 Jurnal Teknik Kimia 20 [2] Fachry A R and Sartika A 2012 Jurnal Teknik Kimia 18 [3] Leejarkpai T, Suwanmanee U, Rudeekit Y and Mungcharoen T 2011 Waste management 31 1153 – 1161 [4] Lazuardi G P and Cahyaningrum S E 2013 UNESA Journal of Chemistry 2 [5] Joseph C S, Prashanth K H, Rastogi N, Indiramma A, Reddy S Y and Raghavarao K 2011 Food and Bioprocess Technology 4 1179 – 1185 [6] Jeon Y J, Kamil J Y and Shahidi F 2002 Journal of agricultural and food chemistry 50 5167 – 5178 [7] Butler B, Vergano P, Testin R, Bunn J and Wiles J 1996 Journal of food science 61 953 – 956 [8] Riza M, Mukhlishien, Sentia P et al.

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