

**RECEIVED**  
15 October 2019**REVISED**  
21 November 2019**ACCEPTED FOR  
PUBLICATION**  
2 December 2019**PUBLISHED**  
23 December 2019

## Comparison on Physical Properties Between Polyether-Based and Castor-Based Polyurethane Grouting Materials

Nur Izzah Atirah Mat Hussain<sup>2</sup>, Noor Najmi Bonnia<sup>1\*</sup>, Radin Siti Fazlina Nazrah Hirzin<sup>2</sup>, Ernie Suzana Ali<sup>3</sup>

<sup>1</sup>Materials Science and Technology Department, Faculty of Applied Sciences, Universiti Teknologi MARA, 40450 Shah Alam, Malaysia.

<sup>2</sup>Polymer Science and Technology Department, Faculty of Applied Sciences, Universiti Teknologi MARA, 40450 Shah Alam, Malaysia.

<sup>3</sup>Applied Physic Department, Faculty of Science and Technology, Universiti Sains Islam, 71800 Nilai, Negeri Sembilan, Malaysia

**E-mail:** noornajmi@uitm.edu.my\*

**ABSTRACT:** Polyurethane grouting is an excellent maintenance technology for many infrastructures. Most of commercialized polyurethane grouts (PUG) are polyether-based which are expensive and hazardous. Therefore, in this study, two types of PU which were polyether PUG (petrochemical-based) and castor-based PUG (natural-based) were fabricated to compare their foam reaction time and apparent density. Ratio of isocyanate to polyol (NCO:OH) were varied in four compositions. The rise time of both type of PUG decreased with increasing NCO:OH ratio whereas opposite trend was obtained for tack free time of both PUG. The apparent density of both types of PUG increased with increasing NCO:OH ratio. Overall, CPUG composites had a longer foam reaction time and higher apparent density compared to PPUG composites where the results obtained were in the range of industrial grout properties (cream time: 1-5s, rise time: 90-365s, tack free time: 120-510s, apparent density: 100-300kg/m<sup>3</sup>).

**Keywords:** Polyether-based PUG; Castor-based PUG; NCO:OH ratio, Foam reaction time, Physical properties

### 1. Introduction

Grouting is as a technique that is widely used to repair and strengthen broken and loosened matrices [1,2]. Polymer grouting technology is an excellent maintenance technology for infrastructures as it is economical and very efficient technique to seal crack. Over time, problems on concrete infrastructures arise such as settlement issues, crack and formation of sinkholes which resulting a requirement of continuous maintenance that cause an increase in budget. Therefore, a low cost maintenance using excellent grouting material is introduced to solve problems of settlement issues [3].

One of the grouting materials that has gained attention is polyurethane. Polyurethane exhibit extraordinary properties such as light weight, good mechanical properties and good thermal stability compared to other materials [4]. Polyurethane can also adhere strongly to many substrates which make it useful in many applications [5]. Besides, it also has the ability to expand in a short time which makes it suitable to be used for sealing cracks on concrete structures. Polyurethane grout is a rigid, closed cell foam which is produced from exothermic chemical reaction between polyol and isocyanate. Variations in the type of polyol used and the equivalent ratio of isocyanate to polyol can influence the properties of polyurethane produced which may fulfilled the requirement of specific applications.

Currently, almost 90% of polyurethane production in the industry used polyether polyol as their main component [6]. However, due to the high demand of polyether polyol, their prices have rapidly risen. The main source of polyether polyol which was crude oil and coal had issues which were increasing in depletion rate and required high technology processing system in extraction of polyol [7,8]. Therefore, natural based polyol (castor polyol) is introduced in the production of polyurethane to replace polyether polyol and overcome problems that arised.

Castor oil is one type of vegetable oil that can be used directly as a polyol without any chemical modification process in the presence of OH functional group as its chemical structure [9]. NCO:OH ratio which is defined as the equivalent ratio between isocyanate an polyol is varied in order to produce rigid PUG with excellent physical properties [10]. The aim of this study is to produce polyether and castor-based PUG that meets the industrial standard of grouting materials and to compare their properties.

## 2. Experimental

### 2.1 Materials

Two types of polyol that is used in this research were castor oil (OH value: 161.62 mg/KOH/g, equivalent weight: 347 gmol<sup>-1</sup>) distributed by Progressive Scientific Sdn. Bhd. and polyether-based polyol which is poly propyleneoxy sucrose (OH value: 414 mg/KOH/g, equivalent weight: 135.51 gmol<sup>-1</sup>) supplied by Growchem Sdn. Bhd. Isocyanate (4,4-methylenediphenyl diisocyanate) with NCO content: 31%, molecular weight: 360gmol<sup>-1</sup>, was supplied by Growchem Sdn. Bhd., blowing agent (1,1-dichloro-1-fluoroethane) was provided by Airgas USA, surfactant (polyalkyleneoxidemethylsiloxane copolymer) was manufactured by Momentive Amer Ind., pentamethyldipropylenatriamine (PMDETA) which acts as blowing catalyst and dimethylcetylhexamine (DMCHA) which acts as gelling catalyst where both catalyst supplied by Huntsman (Singapore) Pte Ltd.

### 2.2 Polyurethane Grout Materials Preparation

**Table 1** shows formulations of PUG produced in this work. Four different NCO:OH ratios of polyether-based polyurethane grout (PPUG) and castor-based polyurethane grout (CPUG) composites were produced. PPUG composites were produced by mixing polyether polyol, blowing agent, surfactant, gelling catalyst and blowing catalyst together in a plastic cup using mechanical stirrer for about 2 minutes at 3000 rpm. Then, isocyanate was added into the mixture and mixed at 3000 rpm for about 20 seconds [11]. Later, the mixtures were poured into an acrylic mould and conditioned at room temperature for about 24 hours. Then, the samples were demoulded and conditioned at room temperature for about 36 hours before being tested. The same step was repeated in producing CPUG composites.

**Table 1.** Polyurethane grouting material formulations

Samples		NCO:OH ratio	Blowing agent	Blowing catalyst (pbw)	Gelling catalyst (pbw)	Surfactant (pbw)
Polyether-based PUG	PPUG1	2.0:1	1	0.4	0.4	2
	PPUG2	2.2:1	1	0.4	0.4	2
	PPUG3	2.4:1	1	0.4	0.4	2
	PPUG4	2.6:1	1	0.4	0.4	2
Castor-based PUG	CPUG1	2.0:1	1	0.4	0.4	2
	CPUG2	2.2:1	1	0.4	0.4	2
	CPUG3	2.4:1	1	0.4	0.4	2
	CPUG4	2.6:1	1	0.4	0.4	2

## 3. Results and discussion

### 3.1 Foam Reaction Time

**Table 2** shows the foam reaction time for both PPUG and CPUG composites. Cream time corresponds to the time taken for the foam reaction to take place. The rise time is the time taken for the freely rising foam to stop growing while tack free time is the time taken for the outer skin of the foam loses its stickiness [9,12]. There are slight differences in the cream time of both PPUG and CPUG composites despite the NCO:OH ratio increasing. The cream time for PPUG1 and PPUG2 constant at 2s and increase to 3s for PPUG3 and PPUG4. For CPUG composites, the cream time increase from 3s to 4s and remain constant until CPUG4. There was a slight change in cream time where the differences were only 1s. This indicates that NCO:OH ratio did not much influence the cream time of both PPUG and CPUG composites. Overall, the cream time for PPUG composites were in the range between 2-3s while for CPUG composites, the range was between 3-4s. Despite that, the cream time obtained for both PPUG and CPUG composites were in agreement with commercial polyurethane grout's cream time (<20s) [13].

**Table 2.** Foam reaction time

	Samples	Cream time, s	Rise time, s	Tack free time, s
<b>Polyether-based PUG</b>	PPUG1	2	180	190
	PPUG2	2	170	260
	PPUG3	3	155	295
	PPUG4	3	145	300
<b>Castor-based PUG</b>	CPUG1	3	340	430
	CPUG2	4	320	435
	CPUG3	4	300	470
	CPUG4	4	280	475

The rise time for both PPUG and CPUG composites decreased with increasing of NCO:OH ratio. This was attributed to the increase in activation energy during the foaming reaction which led to the increment of reaction rate and thus resulted in shorter rise time [9]. Overall, the rise time for PPUG composites were in the range of 145-180s while for CPUG composites, the values were in the range of 280-340s. Tack free time of both PPUG and CPUG composites increased with increasing NCO:OH ratio. The increase of NCO:OH ratio cause the increment of NCO monomer. Lower free energy of unreacted monomer (NCO) caused them to be exposed to the free surfaces that resulted the stickiness on the outer skin of the foam and led to longer tack free time [12]. Overall, the tack free time for PPUG composites were in the range of 190-300s while for CPUG composites, the values were in the range of 430-475s. In comparison, CPUG composites had a longer rise time and tack free time compared to PPUG composites. This was because castor polyol had lower OH value compared to polyether polyol [9]. Low OH value of castor polyol caused a lower urethane crosslinking thus resulted in lower exothermic reaction and reduced the foaming reaction in CPUG composites [14]. Despite that, the rise time and tack free time of both PPUG and CPUG composites obtained in agreement with commercial polyurethane grout properties where the rise time were in the range of 110-460s while tack free time values were in the range of 140-470s [15,16].

### 3.2 Apparent Density

**Fig. 1** shows the apparent density of PPUG and CPUG composites. Apparent density is the overall density including the skin of the foam [17]. This property affects other physical and mechanical properties of the PUG produced. Both PPUG and CPUG's apparent density increased with increasing NCO:OH ratio. The increased in isocyanate caused an increased in allophanate crosslinking. Allophanate crosslinking is a crosslinking that form as the excess isocyanate was unable to react with OH functional group from polyol formed a crosslink with urethane linkage. These resulted an increment in crosslinking density, hence, increase the apparent density of PPUG and CPUG composites [12]. The apparent density of PPUG composites were in range between 139.8-173.96 kg/m<sup>3</sup> while for CPUG composites, the values were in the range between 202.15-222.1 kg/m<sup>3</sup>. Overall, the apparent density of CPUG composites was higher than PPUG samples. This is due to the low OH functional group in castor polyol that caused a decrease in blowing efficiency which resulted in less bubbles produced in CPUG matrix. These give CPUG composites a higher apparent density [14]. Despite that, the results obtained for both PPUG and CPUG composites concur and in parallel with commercial polyurethane grout's density (90-360kg/m<sup>3</sup>) [18].

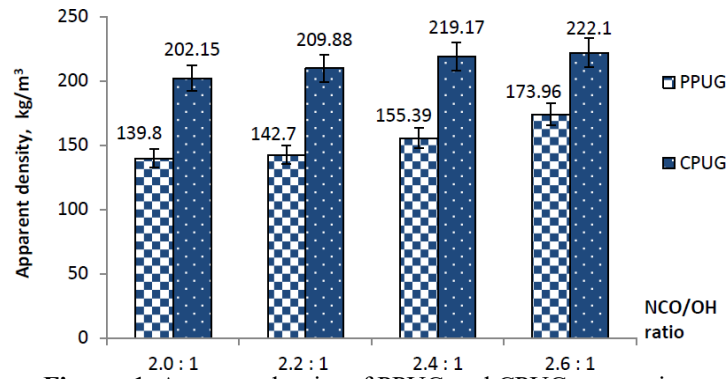


Figure. 1. Apparent density of PPUG and CPUG composites

#### 4. Conclusions

To summarize, the rise time decreased while tack free time increased with increasing NCO:OH ratio for both PPUG and CPUG composites. The apparent density for both PPUG and CPUG composites also increased with the increment in NCO:OH ratio. Foam reaction time and apparent density of both PPUG and CPUG composites obtained were in the range of industrial grout properties. Overall, CPUG composites had higher foam reaction time and apparent densities compared to PPUG composites.

#### Acknowledgement

The authors acknowledge International Polyurethane Technology Foundation for International Grant Scheme 100-IRMI/INT/ 16/6/2 (004/2018) and Universiti Teknologi MARA for the research facilities.

#### References

1. H. Gullu, Const. Build. Mater 93 (2015) 891-910.
2. H. Gullu, Construction Building Materials 127 (2016) 220-236.
3. A.M.M. Fakhar, A. Asmaniza, IOP Conference Series: Materials Science and Engineering 136 (2016) 1-10.
4. D.K. Backus, M.J. Crucini, Int. J. Economics 50 (2000) 185-213.
5. X. Zhang, H. Li, Q. Cao, L. Jin, F. Wang, Sagepub Journal 36 (2018) 436-444.
6. K. H. Badri, Polyurethane: InTech, Rijeka, Croatia, 2012, pp. 447-470.
7. K.F. Adekunle, Journal of Polymer Chemistry 5 (2015) 34-40.
8. I. Banik, M.M. Sain, Journal of Reinforced Plastic Composite 27 (2008) 357-373.
9. A.K. Yusuf, P.A. P. Mamza, A.S. Ahmed, U. Agunwa, International Journal of Scientific and Research Publications 6 (2016) 548-556.
10. G. Runumi, K.N. Utpal, M.A. Sarwar, S.M. Dayal, Journal of World Applied Science 21 (2013) 276-83.
11. E.S. Ali and S.A. Zubir, MATEC Web of Conference 39 (2016) 1-5.
12. S.H. Kim, B.K. Kim, Macromolecular Research 16 (2018) 467-72.
13. Azo-Grout Product Data Sheets. Retrieved from <https://azogROUT.com/azo-grout-product-data-sheets-sobre-productos/> on 15 August 2019.
14. H. Lim, S.H. Kim, B.K. Kim, Polymers For Advanced Technologies 19 (2008) 1729.
15. Alchemy-Spetec. Retrieved from <https://alchemy-spetec.com/up-content/uploads/03-64-01-Injection-Grouting-Hydrophilic-and-Hydrophobic.pdf> on 17 August 2019.
16. SikaFix HH Hydrophilic. Retrieved from <https://usa.sika.com/en/construction/department-of-transportation/grouts/polyurethane-grouts/sikafix-hh-hydrophilic.html>
17. Y. Wei, F. Wand, X. Gao, Y. Zhang, Journal Materials and Civil Engineering 29 (2017).
18. J. Bodi, Z. Bodi, J. Scucka, P. Martinec, Intech-PU Grouting Technologies (2012) 307-333.

© 2019 by the authors. Published by EMS ([www.electroactmater.com](http://www.electroactmater.com)). This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution license (<http://creativecommons.org/licenses/by/4.0/>).

“Paper presented at PhyMas2019, 31 October 2019, Faculty of Applied Sciences, Shah Alam, Selangor, Malaysia.”