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**From:** Communications in Science and Technology  
**Sent:** 13 September 2021 7:36  
**To:** Yulian Firmana Arifin  
**Subject:** [CST] Submission Acknowledgement

Yulian Firmana Arifin:

Thank you for submitting the manuscript, "Volume Change in Compacted Claystone-Bentonite Mixtures as Affected by the Swamp Acidic Water" to Communications in Science and Technology. With the online journal management system that we are using, you will be able to track its progress through the editorial process by logging in to the journal web site:

Manuscript URL: <https://cst.kipmi.or.id/journal/authorDashboard/submission/540>  
Username: yarifin

If you have any questions, please contact me. Thank you for considering this journal as a venue for your work.

Communications in Science and Technology  
The following message is being delivered on behalf of Communications in Science and Technology.

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## Authors

Agus Kurniawan (akurniawan)

Yulian Firmana Arifin (yarifin)

## Messages

Note

From

Dear Authors,

akurniawan

Thank you for considering Communications in Science and Technology.

2021-11-25

Please see the comments of the reviewers on the article entitled: "Volume Change in Compacted Claystone-Bentonite Mixtures as Affected by the Swamp Acidic Water". I suggest you consider these comments, suggestions and questions and revise your article accordingly. The revised version of your submission is due by December 3, 2021.

08:56 AM

For your guidance, reviewers' comments are appended below. If you decide to revise the work, please submit a list of changes or a rebuttal against each point which is being raised and highlight the changes in manuscript when you submit the revised manuscript.

To submit a revision, please go to <https://cst.kipmi.or.id/> (login as an Author) within 10 days; after this time the manuscript will be considered as withdrawn.

Yours sincerely,

Agus Kurniawan

Editor

Communications in Science and Technology

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Editor:

Comments are available in the file attached.

Reviewer #1:

Comments are available in the file attached.

Reviewer #2:

- Fig. 4 : it is better to show the volume change graph than

there is no change

- Is there any effect of montmorillonite in this study?

Conclusions:

- Is there any optimum mixture of clay, bentonite and acidic water that can be used as clay liner, landfill barrier or others?

- It is better to provide final/ important statement of the result in the conclusions related to the effect of swamp acidic water on volume change of mixture material not only the results summary

Reviewer #3:

The manuscript entitled "Volume Change in Compacted Claystone?Bentonite Mixtures as Affected by the Swamp Acidic Water" can be accepted in CST after some points below are clarified:

1. The content and topic is interesting; however, some places are difficult to understand thus it is recommended to use Professional English editing.

2. The subject identified in the work is still localized, it is recommended to bring the international scope. Hence, the similar place and location in other countries can obtain the benefit from this research.

3. The authors said that "This value is greater than that required for clay liners in many countries (i.e.,  $1.0 \times 10^{-9}$  m/s)." Which is the reference concluding this statement?

4. There is a shift in peak oriented in the range of wave number of about 3400-1 in Figure 3. Please explain the phenomenon of the result.

5. In Fig. 4b, there is a delay of significant increase in deformation for 80C20B-10(A) and then the sample leads to the maximum deformation. Please clarify the result.

6. The sentences of "As seen in Fig. 5 (a), the swelling percentage that occurs in the acidic water is higher than those in pure water for the samples with initial water content of 10% and 15% and bentonite content of 5% and 10%." is confused to be understood. Please revise it.

7. It is recommended to shorten the conclusion. The significant results should be mainly described rather than all results. The itemized conclusion should be avoided.

[akurniawan, 540-1557-1-5-20211125 \(1\).doc](#)

[akurniawan, 540-Article Text-1553-1-18-20211125 \(2\).doc](#)

► Dear Editor and Reviewers,

yarifin

2021-11-28

I hereby submit the revised article based on the reviewers' comments and suggestions. We appreciate the editor and

11:34 PM

From

Note

## Response Authors to Reviewer's Comments/Remarks

### Volume Change in Compacted Claystone–Bentonite Mixtures as Affected by the Swamp Acidic Water

**Authors:** Yulian Firmana Arifin, Muhammad Arsyad, Jeane Monica, Setianto Samingan Agus

Dear Editor and Reviewers,

I hereby submit the revised article based on the reviewers' comments and suggestions. We appreciate the editor and reviewers' feedback and suggestions for improving the quality of this article. Additionally, I indicate that professional English editing will be performed immediately upon acceptance of this manuscript for publication. Some of the improvements we made in response to the editor and reviewer's comments are listed below. Due to the absence of lines for column 2, the terms "Column 1" and "Column 2" are used in the updated section.

No	Editor and Reviewer's comments	Authors response
	<b>Editor</b>	
	Title_ Does the title accommodate variations in the pH levels of swamp water	The title is made more general to accommodate the use of acidic water directly from the field. The general pH of acidic water from references has been added in the Introduction in <b>Line 71-81 column 2.</b>
	Abstract_ make a more technical sentence regarding the possible effect on clay due to swamp water. Abstract_ no need to repeat the percentage symbol, just use it at the end of the sentence Abstract_ Use spaces where needed	The abstract has been revised in response to reviewer comments.
	Introduction_ Is there a role for radioactivity in this research, otherwise it doesn't need to be mentioned	The sentence has been revised in <b>Line 2.</b>
	8000m <sup>3</sup> _Use space	All units have been corrected according to suggestions from the editor.
	Figures 7 and 9 must be rearranged again so that they match the topics discussed and do not overlap with the conclusion section	The figures have been rearranged.
	Conclusion_ Rephrase with "the sample tends to compressed"	Conclusion has been revised as shown in <b>Line 571 Column 2</b>
	Add quantitative parameters to strengthen this conclusion	Conclusion has been revised as shown in <b>Line 571 Column 2</b>
	<b>Reviewer 1</b>	
	The abstract section should be less than 150 words. The description of your methodology could be shortened.	Abstract has been revised with 150 words.

	Abstract_ This was not mentioned in the conclusion section, yet it appears in the abstract. If this is a significant finding, it might be better to elaborate this in the conclusion.	The abstract has been revised to reflect feedback from reviewers.
	Line 80 Col. 2 <u>Materials and Methods</u> Please write a short paragraph describing your overall research method.	Short paragraph has been inserted in the article in <b>Line 92-97 Column 2</b> .
	Line 192 Col. 2 <u>Results and Discussions</u>	Typo and fixed <b>Line 318 Column 2</b>
	Lines 308-311 <u>Future research on longer periode test</u> If the condition does not fluctuate much, why is it necessary to conduct a longer period of observation?	Apart from that, clay, particularly bentonite, has a very time-dependent behavior, and the clayliner used must be able to sustain contamination throughout the waste decomposition process, which can take up to 50 years.  The sentence has been revised in <b>Line 324-329 Column 1</b>
	Fig. 3 <u>Functional group</u>	Typo and fixed <b>Line 318 Column 1</b>
	Line 505 <u>decreases</u>	Typo and fixed <b>Line 526 Column 1</b>
	Line 518 <u>indicates</u>	Typo and fixed <b>Line 563 Column 1</b>
	<b>Reviewer 2</b>	
	- Fig. 4 : it is better to show the volume change graph than deformation if possible, or explain the correlation between deformation and volume change	We do thank the reviewer for conscientious review. The sentence below has been added in <b>Line 330-331 column 2</b> .  Because the test was conducted on an oedometer, the only deformation that happens is in the vertical direction, with no change in the horizontal. As a result, the amount of deformation is proportional to the volume change of the sampel.
	- Is there any change of pH value before and after mixing the materials in this study? It is better to show if any, or mention it if thereis no change	The authors express their gratitude to the reviewer for the advice on pH samples before and after the test. The authors had planned this before the research was conducted, but the test was not carried out for several reasons, including (1) there was a test that had to be carried out after the test was completed (i.e., the moisture content of the sample), which required the sample to be oven-dried. Naturally, this altered the sample's condition, making pH testing unfeasible. (2) The sample was divided following the

		test for the FTIR, XRD, and XRF tests, as well as the majority of the tests indicated in point (1). So, although the pH after the test is fascinating to discuss, it was not done. (3) Paralel tests were difficult due to long duration of the test (i.e 12 days) for one sample, limited equipments, and tool usage queue.
	- Is there any effect of montmorillonite in this study?	The sentences below have been added in <b>Line 401 Column 2 – 426 Column 1</b> .  Although both include montmorillonite, bentonite contains a greater proportion of the mineral than natural soils (ref). Thus, by adding bentonite to the mixture, the amount of montmorillonite in it is increased. Clay and montmorillonite contain are thought to have a distinct influence on swelling behavior, ranging from minor to major. However, it was revealed that the latter element had a greater influence than the former (ref).
	Conclusions: - Is there any optimum mixture of clay, bentonite and acidic water that can be used as clay liner, landfill barrier or others? - It is better to provide final/ important statement of the result in the conclusions related to the effect of swamp acidic water on volume change of mixture material not only the results summary	The initial moisture content of the sample affects the swelling of compacted claystone bentonite mixtures in acidic water. The sample tends to compress when the moisture level is higher than the wet of optimum. Compression increases as the amount of bentonite in the mixture increases. There is a noticeable behavioral difference between samples having more than 10% bentonite. Compression occurs faster in this condition than in pure water. A mixture with 20% bentonite content compacted at dry to optimum moisture content is the best for mitigating the negative effects of acidic water. The conclusion has been revised in <b>Line 569 Column 2</b>
	<b>Reviewer 3</b>	
	The manuscript entitled "Volume Change in Compacted Claystone?Bentonite Mixtures as Affected by the Swamp Acidic Water" can be accepted in CST after some points below are clarified:	We do thank the reviewer for conscientious review. The following are revisions in response to the comments and suggestions of the Reviewer 3.

1.	The content and topic is interesting; however, some places are difficult to understand thus it is recommended to use Professional English editing.	Professional English editing will be performed immediately upon acceptance of this manuscript for publication.
2.	The subject identified in the work is still localized, it is recommended to bring the international scope. Hence, the similar place and location in other countries can obtain the benefit from this research.	This can occur in any location with a large area of peat wetland. Tcvetkov (2017) provides data on countries with peat swamp areas, including Russia (150 million ha), Indonesia (26 million ha), the United States of America (40 million ha), Canada (170 million ha), Finland (10 million ha), China (3.5 million ha), Sweden (7 million ha), and Ireland (1.2 million ha), as well as the remaining 12.3 million ha in Malaysia, Germany, Poland, the United Kingdom, and Belarus. Wind-Mulder et al (1996) reported water chemistry data from four peat swamp areas in Canada showing the average pH was 3.7–3.9 with a predominant of $\text{SO}_4^{2-}$ .  The sentences and new references have been added in <b>Line 71-81 column 2</b> .
3.	The authors said that "This value is greater than that required for clay liners in many countries (i.e., $1.0 \times 10^{-9}$ m/s)." Which is the reference concluding this statement?	A reference has been inserted at the end of the sentence in <b>Line 115 column 2</b> (reference No. 25).
4.	There is a shift in peak oriented in the range of wave number of about 3400-1 in Figure 3. Please explain the phenomenon of the result.	The impact of acid on montmorillonite is almost similar whenever the-OH extracting happens at a wave length between 3441 $\text{cm}^{-1}$ (Akpomi et al., 2016) and 3427 $\text{cm}^{-1}$ (Ozcan et al., 2005).  The sentences and references have been added in <b>Lines 332 Column 2</b> .
5.	In Fig. 4b, there is a delay of significant increase in deformation for 80C20B-10(A) and then the sample leads to the maximum deformation. Please clarify the result.	The effect of acid water on bentonite has begun to be seen at low water content. In Fig. 4(b), there is a significant delay in increasing deformation for 80C20B-10(A), and then the sample leads to the maximum deformation. The high concentration of ions contained in acidic water results in a balancing process with the soil water inside. As shown in FTIR in Figure 3, after the acidic water began to be absorbed by the bentonite surface,

		<p>modifications occurred which resulted in an increase in the amount of water absorbed on the surface. This resulted in high swelling that occurred, as indicated by the vertical deformation of the sample.</p> <p>The sentences have been added in <b>Line 392-402</b></p>
6.	<p>The sentences of "As seen in Fig. 5 (a), the swelling percentage that occurs in the acidic water is higher than those in pure water for the samples with initial water content of 10% and 15% and bentonite content of 5% and 10%." is confused to be understood. Please revise it.</p>	<p>Fig. 5(a) shows that the 10 percent bentonite level to be the limit of the distinct swelling behavior of the samples. At bentonite less than 10%, the swelling in the acidic water is higher than that in pure water for the samples with an initial water content of 10% and 15%.</p> <p>The sentences have been revised in <b>Line 427-431 Column 2</b></p>
7.	<p>It is recommended to shorten the conclusion. The significant results should be mainly described rather than all results. The itemized conclusion should be avoided.</p>	<p>The initial moisture content of the sample affects the swelling of compacted claystone bentonite mixtures in acidic water. The sample tends to compress when the moisture level is higher than the wet of optimum. Compression increases as the amount of bentonite in the mixture increases. There is a noticeable behavioral difference between samples having more than 10% bentonite. Compression occurs faster in this condition than in pure water. A mixture with 20% bentonite content compacted at dry to optimum moisture content is the best for mitigating the negative effects of acidic water.</p> <p>The conclusions can be found in <b>Line 571 Column 2</b></p>



# Volume Change in Compacted Claystone–Bentonite Mixtures as Affected by the Swamp Acidic Water

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## Article history:

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## Abstract

Water containing sulfuric acid with a pH of up to 3 is prevalent in swampy areas. This article focuses on the effects of the solution on volume change of compacted claystone–bentonite mixtures. Claystone was from Banjarbakula landfill. The claystone was mixed with bentonite on a 5, 10, 15, and 20% dry mass basis. Samples have dry density of 16 kN/m<sup>3</sup> and moisture content of 10, 15, and 20%. The oedometer examined the samples' swelling and compression in both pure and acidic water. Characterization tests (i.e., XRF, XRD, and FTIR) were also performed. The results show that swelling and compression are affected by initial moisture and bentonite content. Samples with a moisture content of 20% showed compression in acidic water. Acidic water changes the water absorbed on the clay surface without altering the mineral. A mixture containing 20% bentonite compacted to optimum moisture content is best for reducing acidic water effects.

**Keywords:** claystone; bentonite; swelling; compression; clay liner

## 1. Introduction

Numerous materials have been proposed as waste barriers. One of them is a mixture of claystone and bentonite. Along with the clay minerals it contains, claystone is used to recycle waste material from excavation [1]. Previously, claystone from excavation had been considered as an undesirable construction material, particularly when it came into contact with water [2], [3]. During the development of the Banjarbakula landfill in Banjarbaru, South Kalimantan, Indonesia, an approximately 8000 m<sup>3</sup> of claystone was dumped as the material was regarded as an undesirable material. Nevertheless, there are some economic benefits utilizing this material. Therefore, both the economic and environmental concerns are addressed from the use of this material [1].

Hydraulic conductivity, shear strength, compressibility and swelling characteristics are among the properties that are commonly evaluated in relation to the use of bentonite-based materials as a landfill barrier. These properties are strongly influenced by the bentonite content in the mixtures. Khalid et al. [4] found that the influence of bentonite on the geotechnical properties is more evident at a bentonite percentage of more than 10% for clay–bentonite mixtures. Meanwhile, adding more than 20% bentonite to silty sand has no effect on the hydraulic conductivity of the clay liner [5].

Clay liners, as a barrier, are extremely prone to interaction with substances other than water. In the nuclear waste

repository, the sealing material will interact with the saline solution of the surrounding host rock. This will have an effect on the canister's corrosion, the swelling and self-sealing capability of the bentonite back fill, and a sophisticated geochemical calculation [6]. Wang et al. [1] found that due to the high sample density and low salinity of the water utilized, water chemistry had no effect on the swelling behavior of compacted claystone–bentonite mixes. Swelling pressure of compacted claystone–bentonite mixture is affected by the final dry density of bentonite in the mixture, while the claystone used is considered to behave as sand [1].

Claystone, on the other hand, is highly impacted by the minerals it contains. Its combination with bentonite will have an effect on the mixture's overall behavior. The swelling capacity of bentonite is also impacted by the chemistry of saturating fluids; the higher the salinity, the lower the sample's swelling capacity, which has a negligible effect on samples with a high density (i.e., 17–19 kN/m<sup>3</sup>) [1].

Besides density, water salinity has an effect on hydromechanical materials containing a large amount of smectite (i.e., 50% bentonite) [6]. Apart from swelling characteristics, Siddiqua et al. [6] examined the influence of salt on compression and swelling indices (i.e.,  $c_c$  and  $c_s$ ) obtained by consolidation tests.  $c_c$  was found to decrease in the presence of saline solution, indicating its influence on the sample's compressibility behavior. On natural stiff clay, similar results were reported by Ngunyen et al. [7]. Clays with a high smectite content experience more alterations than others.

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57 Sealing materials may also interact with acidic liquids 117  
 58 addition to salt. Acidic water, which is generated by acid rain 118  
 59 and has a pH of 3–4.5, reduces the shear strength 119  
 60 sedimentary and igneous residual soils and increases the 120  
 61 permeability [8]. Acid rain infiltration into the soil causes 121  
 62 leaching of  $\text{Fe}^{3+}$  and  $\text{Al}^{3+}$ , which plays an important role 122  
 63 cementation. The effect of acid rain on the development 123  
 64 soil erosion was also investigated by Matsumoto et al. [9]. The 124  
 65 release of  $\text{Al}^{3+}$  owing to fluids with pH of 2–6 was also 125  
 66 observed in the study, which resulted in the development 126  
 67 soil erosion. Meanwhile, Ahmed et al. [10] found that 127  
 68 swelling ratio reduced when the pH in soil pores decreases 128  
 69 due to acid water. Gratchev and Towhata [11] investigated 129  
 70 potential of changes in the compressibility of marine clay 130  
 71 to soil contamination from past mismanagement of waste. It 131  
 72 reported that acid water increases or decreases 132  
 73 compressibility index depending on the minerals and 133  
 74 structure. Le et al. [12] investigated a coastal acid sulfate 134  
 75 in Australia that contains sulfidic mineral (i.e.,  $\text{FeS}_2$ ). The 135  
 76 results of the compressibility test show that the physical 136  
 77 structure of the soil is influenced by  $\text{H}^+$  and  $\text{Ca}^{2+}$  cations. In 137  
 78 short time, the effect appears to be insignificant. Besides time 138  
 79 however, the combination of pore water chemical 139  
 80 composition, compressive pressure, and moisture content 140  
 81 affects the permeability of the acid sulfate soil [12]. 141

82 Acidic water has also been reported to cause damage 142  
 83 industrial areas due to contamination with sulfuric acid, which 143  
 84 is widely used in the paper industry, petroleum refining 144  
 85 copper leaching, inorganic pigments, and the organic chemical 145  
 86 industry [13]. In the soil, it was found that 1N  $\text{H}_2\text{SO}_4$  results 146  
 87 in the formation of gypsum and cornelite, whereas 4N  $\text{H}_2\text{SO}_4$  147  
 88 formed aluminite and chloritoid. Mineral changes in the black 148  
 89 cotton soil used in the study resulted in an increase in percent 149  
 90 swelling. In addition, acid solution with a high 150  
 91 concentration also produces a greater swelling potential [13].  
 92 Numerous researchers have also reported soil heaving induced 151  
 93 by acidic solutions [14]–[16]. Sridharan et al. [14] studied 152  
 94 incidence of floor, pavement and foundation distress in 153  
 95 fertilizer factory. The damage was determined to be the result 154  
 96 of heave induced by phosphoric acid reacting with soil in 155  
 97 acidic environment. Assa'ad [15] reported the incline of 156  
 98 storage tank at the chemical fertilizer factory in Aqaba, Jordan  
 99 was caused by phosphoric acid leaking and interacting with 157  
 100 the subgrade soil. Like a gel, phosphate compounds are 158  
 101 formed and filled the pores, which results in trapped gas 159  
 102 resulting from the chemical process. The pressure generated 160  
 103 causes the tank to lift when it is empty. Rama Vara Prasad 161  
 104 al. [17] investigated the swelling potential of three soils 162  
 105 namely black cotton soil, sodium bentonite, and kaolin 163  
 106 using two acidic solutions (i.e.,  $\text{H}_2\text{SO}_4$  and  $\text{H}_3\text{PO}_4$ ). The 164  
 107 results show that the swelling potential of montmorillonite 165  
 108 soils is influenced by the type of cation exchangeable. The 166  
 109 cation exchange reaction and the dissolution of some minerals 167  
 110 resulted in mineral changes in the montmorillonite soil which 168  
 111 affected its swelling behavior. In kaolinite soils, adsorption 169  
 112  $\text{H}^+$  at the broken ends results in a face-to-edge association 170  
 113 the particle, which results in an increase in the swelling 171  
 114 potential of the soil, coupled with changes in soil mineralogy 172  
 115 Chen et al. [18] investigated the compressibility of kaolin 173  
 116 soil using pore fluid with a larger dielectric constant than

water, such as acetic acid. The results indicate that the  
 compression and swelling index samples in the solution are  
 smaller than those in water. Meanwhile, Wahid et al. [19]  
 concluded that kaolinite is not affected by salinity but pH,  
 which attacks the tip of the particle. The compression that  
 occurs under constant load is caused by the interaction of  
 kaolinite with acid solution as a result of sliding between  
 particles and is irreversible.

In South Kalimantan, the area is predominantly swampy  
 and low land. In areas where the soil is predominantly peat,  
 the presence of sulfuric acid in river water causes the pH to  
 vary from 3.4–4.2. The pH does not increase even during  
 rainy season due to high precipitation resulting in rising water  
 levels in the river, thereby preventing the entry of seawater  
 into the river [20]. This can occur in any location with a large  
 area of peat wetland. Tcvetkov [21] provides data on countries  
 with peat swamp areas, including Russia (150 million ha),  
 Indonesia (26 million ha), the United States of America (40  
 million ha), Canada (170 million ha), Finland (10 million ha),  
 China (3.5 million ha), Sweden (7 million ha), and Ireland  
 (1.2 million ha), as well as the remaining 12.3 million ha in  
 Malaysia, Germany, Poland, the United Kingdom, and  
 Belarus. Wind-Mulder et al [22] reported water chemistry data  
 from four peat swamp areas in Canada shows the average pH  
 of 3.7–3.9 with a predominant of  $\text{SO}_4^{2-}$ . Therefore, the acidic  
 water has a high potential of reacting with the clay liner that  
 surrounds it. This paper aims to examine the effect of swamp  
 acidic water on the volume change (i.e., swelling and  
 compression) of the claystone–bentonite mixture. An  
 oedometer was used to evaluate samples of claystone and  
 bentonite mixtures with various compositions in acid water as  
 immersion.

## 2. Materials and Methods

Both natural and fabricated clays (i.e., claystone and  
 bentonite) were used in this study. In addition, the acidic  
 water utilized was obtained directly from a swampy area in  
 order to explore its composition and effects on the clay liner.  
 Overall sample preparation, compaction, and volume change  
 tests were carried out in the laboratory at room temperature.

### 2.1. Claystone

The claystone used was taken from the Banjarbakula  
 landfill project site. The soil was not used in the project and  
 was disposed of. The claystone has a moisture content of  
 2.76%, Gs 2.6, a liquid limit (LL) of 40%, a plastic limit (PL)  
 of 20%, and a shrinkage limit (SL) of 15%. The material  
 consists of 4.5% sand, 43.9% silt, and 51.6% clay. According  
 to the Unified Soil Classification System (USCS) [23], the  
 claystone is classified as an anorganic clay with low to  
 medium plasticity (CL). The main exchangeable cation  
 claystone used was  $\text{Ca}^{2+}$  4.3 meq/g and the remainder was  $\text{Na}^+$   
 0.3 meq/g,  $\text{Mg}^{2+}$  0.1 meq/g, and  $\text{K}^+$  0.3 meq/g. At a dry unit  
 weight of 16 kN/m<sup>3</sup>, the compacted claystone has a hydraulic  
 conductivity of  $7.9 \times 10^{-9}$  m/s [24]. This value is greater than  
 that required for clay liners in many countries (i.e.,  $1.0 \times 10^{-9}$   
 m/s) [25].

172 2.2. Bentonite

173 The bentonite used is commercial bentonite with the main  
 174 exchangeable cation  $\text{Ca}^{2+}$  18.7 meq/g. The others (i.e.,  $\text{Na}^{+}$ ,  
 175  $\text{Mg}^{2+}$ , and  $\text{K}^{+}$ ) are 0.34 meq/g, 0.2 meq/g, and 0.58 meq/g,  
 176 respectively. The bentonite has a moisture content of 14.17%,  
 177 a specific gravity of 2.71, LL 351.71%, PL 44.68%, and  
 178 41.89%, and a plasticity index (PI) of 307.03%. The material  
 179 consists of 1.4% fine sand, 8.3% silt, and 90.3% clay.

180 2.3. Acidic water

181 Acidic water was taken from a river in Tanipah village,  
 182 the Barito Kuala district in South Kalimantan. The water has  
 183 pH of 3.4–3.6. This pH tends to remain constant throughout  
 184 the year, both in the dry and rainy seasons. The chemical  
 185 composition of the acidic water is shown in Table 1.

186 The chemical compounds dominant in the solution are  
 187  $\text{Ca}^{2+}$ ,  $\text{SO}_4^{2-}$ , and  $\text{Cl}^{-}$ . The high concentration of sulfate ions is  
 188 a result of pyrite oxidation occurring in the soil [20].  
 189 Commonly, the  $\text{SO}_4^{2-}/\text{Cl}^{-}$  ratio is used to determine the  
 190 influence of sulfuric acid on pyrite oxidation on the  
 191 composition of river water in swamp areas.

192 Table 1. Chemical compositions of the acidic water used.

Chemical compound	$\text{K}^{+}$	$\text{Ca}^{2+}$	$\text{Na}^{+}$	$\text{Fe}^{3+}$	$\text{Mn}^{2+}$	$\text{SO}_4^{2-}$	$\text{Cl}^{-}$
mg/l	4.21	158.86	6.910	4.876	1.427	261.02	153.4

193 2.4. Sample preparation

194 Bentonite was mixed with claystone at a percentage of 5%,  
 195 10%, 15%, and 20% based on its dry weight. Water was  
 196 added to the mixtures at a certain amounts to provide the  
 197 samples an initial moisture contents ( $w$ ) of 10%, 15%, and  
 198 20%. The sample target water contents were based on results  
 199 of the Proctor standard compaction test on the claystone with  
 200 an optimum moisture content (OP) of 15% obtained. As a  
 201 result, the water contents of 10% and 20% fall on the dry of  
 202 optimum (DOP) and the wet of optimum (WOP) water  
 203 content, respectively. After that, the mixtures were statically  
 204 compressed with a hydraulic jack to produce samples a dry  
 205 unit weight ( $\gamma_d$ ) of 16 kN/m<sup>3</sup>. The samples have a diameter of  
 206 63.4mm and a height of 20mm. The initial conditions and  
 207 sample identifications (Sample IDs) are shown in Table 2.  
 208 The names are given following the sample conditions, such as  
 209 composition and initial water content.

210 2.5. Swelling and Compression tests

211 Two tests were carried out in the oedometer, namely the  
 212 swelling potential and the compression tests. These tests were  
 213 performed following the standard ASTM procedures (i.e.,  
 214 ASTM D4829–11 [26] and ASTM D2435–04 [27]). The  
 215 water used in the test was pure water with a pH of  $\pm 7$  and  
 216 swamp acidic water with a pH of 3.4. The tests using the two  
 217 waters were carried out separately. For the test with pure  
 218 water, the sample in the oedometer was immersed in the water  
 219 under a pressure of 6.9 kPa to obtain the sample's swelling

220 strain. After equilibrium was reached, which was observed  
 221 from constant dial gauge readings, the sample was loaded and  
 222 subsequently unloaded following the consolidation test  
 223 procedure [27]. Similar procedures were also carried out for  
 224 the samples tested using swamp acidic water.

225 2.6. Sample characterisation

226 The investigation into the effects of acidic water on the  
 227 mixtures of claystone and bentonite commences with the  
 228 Atterberg limit tests, which were carried out to determine the  
 229 liquid limit, plastic limit, and plasticity index of the samples.  
 230 Similar approach was also adopted by a number of other  
 231 researchers [8][17][28].

Table 2. Sample initial conditions

Sample ID	Claystone (%)	Bentonite (%)	$\gamma_d$ (kN/m <sup>3</sup> )	$w$ (%)
100C–10	100	0	16	10
100C–15	100	0	16	15
100C–20	100	0	16	20
95C5B–10	95	5	16	10
95C5B–15	95	5	16	15
95C5B–20	95	5	16	20
90C10B–10	90	10	16	10
90C10B–15	90	10	16	15
90C10B–20	90	10	16	20
85C15B–10	85	15	16	10
85C15B–15	85	15	16	15
85C15B–20	85	15	16	20
80C20B–10	80	20	16	10
80C20B–15	80	20	16	15
80C20B–20	80	20	16	20

232 The acidic water has a physical influence on clay and can  
 233 also cause chemical and mineral changes with the clay. The  
 234 alterations in the mineral contents were investigated using X-  
 235 ray diffraction (XRD) analysis for the samples before and  
 236 after test with the acidic water. In addition, Fourier-transform  
 237 infrared spectroscopy (FTIR) test was used to analyze the  
 238 functional groups of materials tested with pure water and  
 239 acidic water. Finally, the samples' chemical compositions  
 240 were measured using X-ray fluorescence (XRF).

241 3. Results and Discussions

242 3.1. Effect of swamp acidic water on sample characterisation.

243 Figs. 1(a) and 1(b) depict the influence of acidic water on the  
 244 claystone-bentonite mixture's liquid limit (LL) and plastic  
 245 limit (PL), respectively. Fig. 1(a) shows that LL increases  
 246 with increasing bentonite concentration in both the pure water  
 247 and acidic water tests. This is plausible since the LL of the  
 248 bentonite is greater than that of the claystone. It is also evident  
 249 that the influence of bentonite content on the LL is observed  
 250 at the bentonite concentrations greater than 10%. This finding

253 is consistent with that reported by Khalid et al. [4], who found  
 254 that bentonite has an impact on the clay-bentonite  
 255 combination at a concentration of higher than 10%. The LL  
 256 the samples tested with the acidic water is consistently greater  
 257 than those tested with pure water containing more than 10%  
 258 bentonite, as shown in Fig. 1(a). LL increases by up to 16%

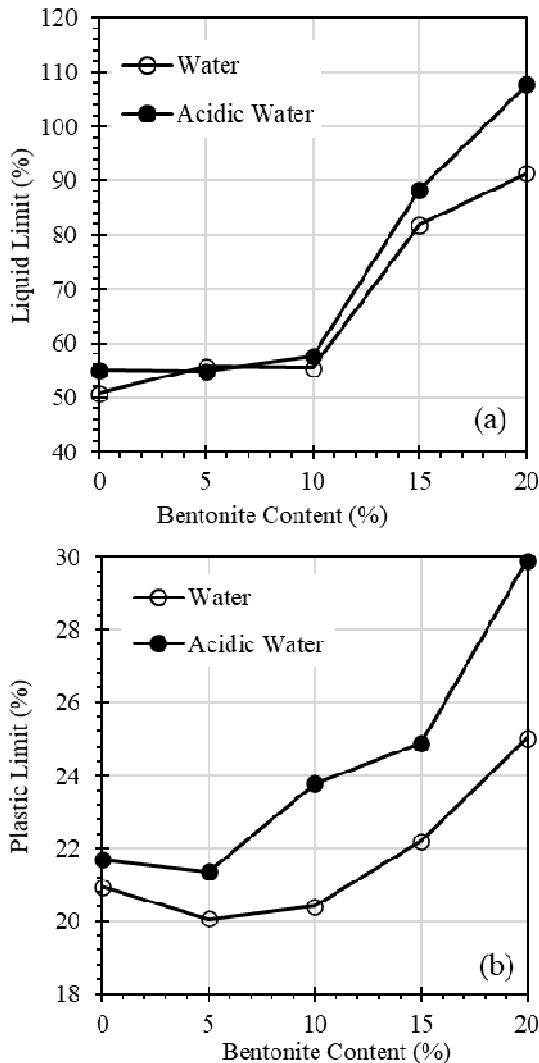


Fig. 1. Effect on acidic water on liquid limit and plastic limit of claystone-bentonite mixtures

259 the 20% bentonite content. At the same bentonite content, LL  
 260 is also higher by up to 16% for the samples tested in the acidic  
 261 water. An insignificant increase in the PL is also observed  
 262 when testing the samples with the acidic water. The greatest  
 263 difference in the PL of around 5% is shown at the 20%  
 264 bentonite content. Since only minor change in the PL is  
 265 observed for the tests using the acidic water, the change in the  
 266 PI is almost similar to the change in the LL. An increase in the  
 267 LL and PI with reducing pH of the soil water was also  
 268 reported by Bakhshipour et al. [8].

269 Table 3 shows the oxides contents of claystone, bentonite,  
 270 and claystone-bentonite mixtures before and after interacting  
 271 with the acidic water obtained from the XRF test. The samples  
 272 tested were taken from those after the consolidation test with  
 273 different bentonite and initial water contents. According to  
 274 samples ID, the samples consist of claystone, bentonite, and

the mixes with varying bentonite percentages (i.e., 5%  
 (95C5B) and 20% (80C20B)), and also different initial  
 moisture contents (i.e., 10% and 20%). As shown in the table,  
 claystone and bentonite predominately contain SiO<sub>2</sub> with a  
 percentage of 55.6% and 54.6%, respectively, followed by  
 Fe<sub>2</sub>O<sub>3</sub> as the next oxide with a content of 19.3% and 23.4%,  
 respectively. Both materials also contain almost the same  
 Al<sub>2</sub>O<sub>3</sub>, which is 15% and 14%, respectively. The rest is K<sub>2</sub>O,  
 CaO and TiO<sub>2</sub>.

284 Table 3. Oxides of claystone, bentonite, dan claystone-bentonite mixtures

Sample ID	Condition	Compound (%)					
		Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	K <sub>2</sub> O	CaO	TiO <sub>2</sub>	Fe <sub>2</sub> O <sub>3</sub>
Claystone (C)	Before test	15	55.6	4.33	3.22	1.83	19.3
Bentonite (B)	Before test	14	54.6	0.56	4.10	1.82	23.4
95C5B-10	After test	14	55.1	3.90	2.98	1.91	21.2
95C5B-20	After test	14	53.8	4.01	3.01	1.93	21.4
80C20B-10	After test	13	54.8	3.43	3.17	1.93	22.6
80C20B-20	After test	14	53.1	3.41	3.33	1.90	22.6

285

286 Bakhshipour et al. [8] reported leaching of Al<sup>2+</sup>, Fe<sup>3+</sup>, Si<sup>2+</sup>,  
 287 K<sup>+</sup> and Ca<sup>2+</sup> due to acid rain infiltration, which resulted in  
 288 reduced sample strength. Artificial acid rain (AAR) was  
 289 prepared by adding a certain volume of 0.005 M nitric acid  
 290 (HNO<sub>3</sub>) and sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) to deionized distilled water  
 291 with pH values of 2, 3, 4, 5, and 5.6. In this study, samples  
 292 soaked in the acidic water with chemical contents as shown in  
 293 Table 1 did not affect the samples' oxide contents. The  
 294 contents of Al<sub>2</sub>O<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>, and SiO<sub>2</sub> as shown in Table 3 did  
 295 not alter for the samples with 5% and 20% bentonite contents.  
 296 Neither cation exchange nor leaching occurred during the  
 297 swelling and consolidation processes.

298 Fig. 2 shows the XRD results of the claystone and  
 299 bentonite samples (i.e., the bottom curve) and those after

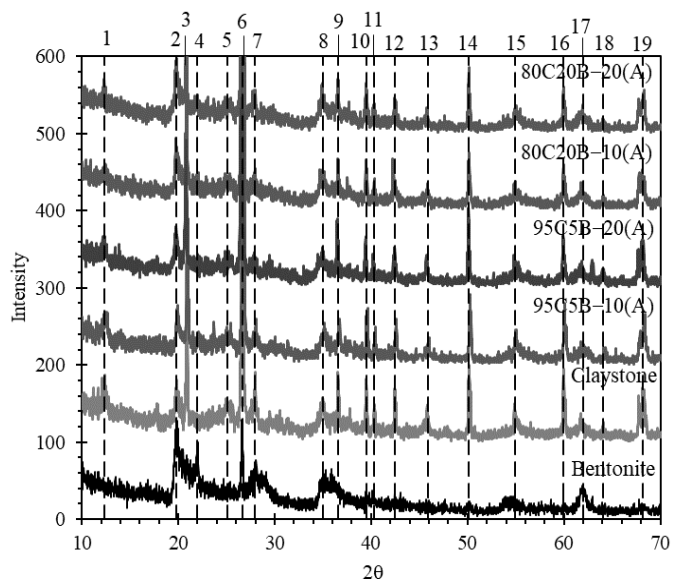


Fig. 2. Mineralogy of samples before and after tested by the acidic water

interacting with the acidic water (denoted by (A)) for different bentonite content (i.e., 5% and 20%) and initial water content (i.e., 10% and 20%). As shown, the claystone sample contains more minerals than the bentonite sample, based on the number of peaks created by the XRD test. This is due to the fact that the claystone sample was collected directly from nature without any purification or other processes. Claystone is composed of various minerals, including kaolinite (1), illite (2, 11), quartz (3, 6, 10, 12, 14, 19), vermiculite (5, 16), feldspar (7), montmorillonite (8, 17), chlorite (9), mica (13), and kaolinite (15, 18). Numerous minerals are found in bentonite, including illite (2), feldspar (4), quartz (6), and montmorillonite (8, 17).

Table 4 summarizes the mineral composition of the samples before and after testing, based on the XRD test results shown in Fig. 2. As seen in Table 4, no minerals were dissolved or formed as a result of interacting with the acidic water solutions. This result differs from the findings of previous investigations, including Sivapullaiah et al. [13] and Rama Vara Prasad et al. [17]. This discrepancy might be explained by the low concentration and brief duration of the interaction (approximately 14 days) with the acidic water. According to Le et al. [12], the combination of acidic water concentration and interaction time has an effect on the clay solution's interaction with the soil. Apart from that, clay, particularly bentonite, has a very time-dependent behavior and the clayliner used must be able to sustain contamination throughout the waste decomposition process, which can take up to 50 years, future research will be conducted over a long period of time.

Table 4. Sample's mineralogy before and after test

Mineral	Before test		After consolidation (A)			
	Claystone	Bentonite	95C5B-10	95C5B-20	80C20B-10	80C20B-20
Illite	√	√	√	√	√	√
Quartz	√	√	√	√	√	√
Vermiculite	√	×	√	√	√	√
Feldspar	√	√	√	√	√	√
Mont.	√	√	√	√	√	√
Chlorite	√	×	√	√	√	√
Mica	√	×	√	√	√	√
Kaolinite	√	×	√	√	√	√

√ = available, × = not available

Fig. 3 shows the results of the FTIR test to determine the functional groups of the samples used, including their condition after interacting with the swamp acidic water. Samples with 10% and 20% bentonite contents were tested. In the figure, letters A and W signifies that the samples tested with the acidic and pure water, respectively. As seen, the peaks are found in the high wavelength region, i.e., at 1630, 3402, 3416, and 3620  $\text{cm}^{-1}$ . These peaks each indicate the presence of clay minerals (i.e., 3618–3628  $\text{cm}^{-1}$ ) [29]. The development of OH was found at 3402–3445  $\text{cm}^{-1}$  which is the interlayer and intralayer of the H bond [30]. Saputra et al. [31] also found a montmorillonite hydroxyl (OH) peak at 3434  $\text{cm}^{-1}$ . Ravindra-Reddy et al. [32] reported that this peak indicates the presence of water on the mineral surface. While

at low wavelengths of 1009, 695, 528, and 470  $\text{cm}^{-1}$ , these peaks are the peaks of the  $\text{SiO}_4$  tetrahedron [32]. Where at 466–470 and 528–535  $\text{cm}^{-1}$  is an indication of the presence of clay and silica minerals.

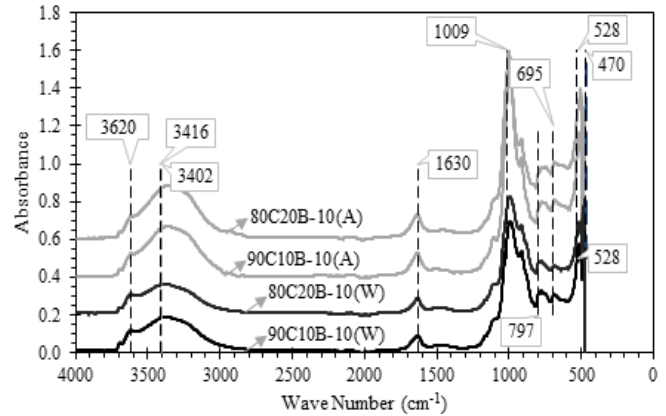


Fig. 3. Functional group of samples tested with pure water (W) and the acidic water (A)

The FTIR results confirm that the material used has clay minerals where  $\text{SiO}_4$  is present at 3618–3628  $\text{cm}^{-1}$  [29]. Moreover, the indications of the presence of the mineral montmorillonite can be seen from the development process at a wavelength of 3402–3445  $\text{cm}^{-1}$  when interacting with pure water. The expansion over this range (i.e. 3402–3445  $\text{cm}^{-1}$ ) for the samples interacting with the acidic water (A) was higher than those with pure water (W). This shows that the clay surface absorbs more water when interacting with acidic water, as seen from the –OH extraction at a wavelength of 3402–3445  $\text{cm}^{-1}$ . The amount of bentonite in the mixture does not appear to affect the extraction intensity of the –OH samples. The impact of acid on montmorillonite is almost similar whenever the –OH extracting happens at a wave length between 3441  $\text{cm}^{-1}$  [33] and 3427  $\text{cm}^{-1}$  [34].

### 3.2. Effect of swamp acidic water on swelling

Because the test was conducted on an oedometer, the only deformation that happens is in the vertical direction, with no change in the horizontal. As a result, the amount of deformation is proportional to the volume change of the sample. Figs. 4(a)–(f) shows the typical swelling development of claystone–bentonite mixtures with time when interacting with pure water (ie, Figs. 4(a), 4(c), and 4(e)) and the acidic water (i.e., Figs. 4(b), 4(d), and 4(f)) under a 6.9kPa load plotted on a semi–logarithmic scale. For the sample with 10% initial moisture content shown in Fig. 4(a), the deformation samples increased slowly in the early stages of the test (i.e., up to 20 minutes). Primary swelling occurs rapidly thereafter up to a certain point (i.e., up to 300–4000 minutes depending on the bentonite content in the mixture) slopes and reaches the maximum deformation. The maximum deformation recorded is referred to as the maximum swelling of the sample. The maximum swelling of claystone is reached in less than 100 minutes. Fig. 4(b) shows that the initial swelling occurs gradually in the beginning, up to 20 minutes, followed by primary swelling up to 300 minutes for the samples with 5% bentonite content, and 3000 minutes for those with 20%

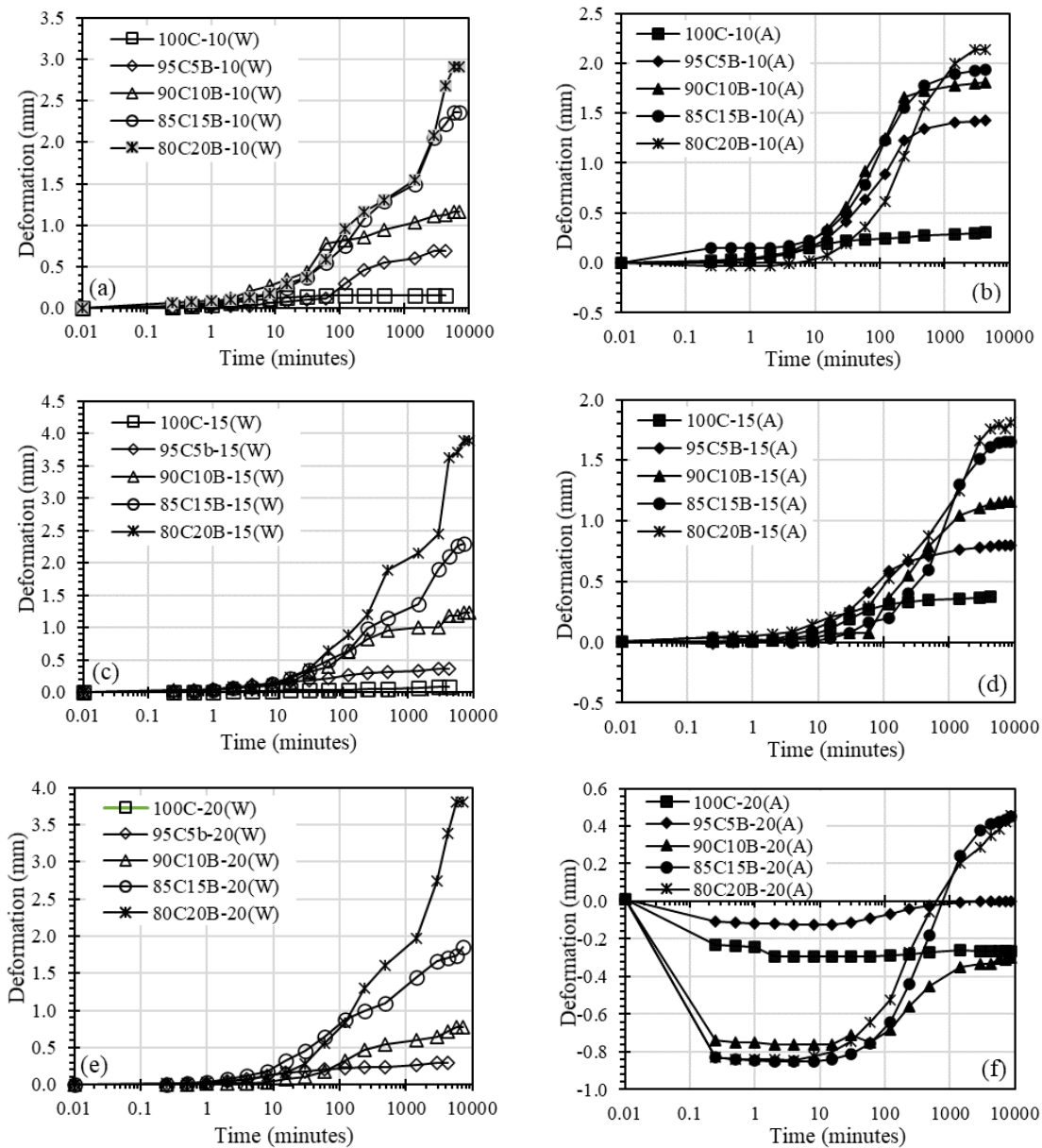


Fig. 4. Swelling development by time of claystone–bentonite mixture samples with initial moisture content of (a)–(b) 10%, (c)–(d) 15%, and (e)–(f) 20%.

388 bentonite. Insignificant compression occurs in sampl<sup>406</sup>  
 389 80C20B-10(A) which contains 20% bentonite. The sampl<sup>407</sup>  
 390 behavior is also observed in the sample with the init<sup>408</sup>  
 391 moisture content of 15%, as shown in Figs. 4(c) and 4(d). <sup>409</sup>  
 392 The effect of acid water on bentonite has begun to be see<sup>410</sup>  
 393 at low water content. In Fig. 4(b), there is a significant del<sup>411</sup>  
 394 in increasing deformation for 80C20B-10(A), and then <sup>412</sup>  
 395 sample leads to the maximum deformation. The high<sup>413</sup>  
 396 concentration of ions contained in acidic water results in<sup>414</sup>  
 397 balancing process with the soil water inside. As shown <sup>415</sup>  
 398 FTIR in Figure 3, after the acidic water began to be absor<sup>416</sup>  
 399 by the bentonite surface, modifications occurred whic<sup>417</sup>  
 400 resulted in an increase in the amount of water absorbed on <sup>418</sup>  
 401 surface. This resulted in high swelling that occurred, <sup>419</sup>  
 402 indicated by the vertical deformation of the sample. <sup>420</sup>

403 Different behavior is noticed in the samples with an init<sup>421</sup>  
 404 moisture content of 20%, where all samples that interact<sup>422</sup>  
 405 with the acidic water tend to experience shrinkage (<sup>423</sup>

compression). Only two samples (i.e., 85C15B-20(A) and <sup>406</sup>  
 80C20B-20(A)) with 15% and 20% bentonite content, <sup>407</sup>  
 respectively, swelled back past their initial conditions. From <sup>408</sup>  
 this behavior, it can be seen that swamp acidic water has an <sup>409</sup>  
 effect on the mixtures with high bentonite content or high <sup>410</sup>  
 initial water content. Claystones containing clay minerals such <sup>411</sup>  
 as kaolinite and illite are not much affected by the acidic <sup>412</sup>  
 water. This can be seen from the results of the Atterberg limit <sup>413</sup>  
 tests (Fig. 1). The unremarkable effect was caused by the <sup>414</sup>  
 adsorption of H<sup>+</sup> at the broken end, resulting in a face-to-edge <sup>415</sup>  
 association of the particle [17] [18] [19].

Swelling occurs due to the absorption of water by the clay <sup>416</sup>  
 surface. Swelling increases with increasing percentage of <sup>417</sup>  
 bentonite in the mixture. Although both include <sup>418</sup>  
 montmorillonite, bentonite contains a greater proportion of the <sup>419</sup>  
 mineral than natural soils [35]. Thus, by adding bentonite <sup>420</sup>  
 to the mixture, the amount of montmorillonite in it is increased. <sup>421</sup>  
 Clay and montmorillonite contain are thought to have a <sup>422</sup>

424 distinct influence on swelling behavior, ranging from minor 456  
 425 major. However, it was revealed that the latter element had 457  
 426 greater influence than the former [35]. The montmorillonite 458  
 427 containing bentonite, according to Pusch et al [36], requires 459  
 428 2–3 layers of water molecules to meet the hydration force 460  
 429 Other researchers even reported 4 layers of water molecules 461  
 430 are required [37]. The thickness and complete hydrated layer 462  
 431 of water molecules in bentonite vary depending on 463  
 432 exchangeable cation. Assuming the specific surface area 464  
 433 bentonite is 500 m<sup>2</sup>/g and the water unit weight of 1 g/cm<sup>3</sup> 465  
 434 Arifin [38] reported that the water content to satisfy 466  
 435 hydration force is 22.7%, 14.1%, 23.9%, and 15.4% for 467  
 436 Mg, Ca, Na, and K types of bentonite, respectively. This water 468  
 437 content can even be greater because the surface water density 469  
 438 is reported to be possibly more than 1 gr/cm<sup>3</sup> [39]. After 470  
 439 this water absorption, the role of surface hydration decreases 471  
 440 In an attempt to equalize ion concentrations, water molecules 472  
 441 tend to diffuse toward the surface. 473

442 Numerous studies have previously observed that mineral 474  
 443 changes occur when soils, particularly those containing 475  
 444 montmorillonite, interact with acidic solutions [13], [12], [14], 476  
 445 [19]. This was not the case in this study, as shown in Fig. 477  
 446 and Table 4. There was even no cation exchange as shown in 478  
 447 Table 3. The volume change that occurred in the sample was 479  
 448 due to the difference in the concentration of cations in the 480  
 449 porewater and the acidic water. This process is known as 481  
 450 osmotically-induced consolidation or osmotic consolidation 482  
 451 [40], [41]. The high concentration of cations present in the 483  
 452 acidic water (Table 1) results in the outward flow from within 484  
 453 to balance these conditions. When the water content of the 485  
 454 sample is high, the concentration of cations in the pore water 486  
 455 decreases and tends to release water, which results in 487

decrease in the soil volume (Fig. 4(f)).

The results of the maximum percentage of swelling and compression that occurred in the sample (Fig. 4) are summarized in Fig. 5. Fig. 5(a) shows that the 10 percent bentonite level to be the limit of the distinct swelling behavior of the samples. At bentonite less than 10%, the swelling in the acidic water is higher than that in pure water for the samples with an initial water content of 10% and 15%. In this condition, the behavior of claystone containing kaolinite is more dominant. In kaolinite soils, acidic water will affect the tip of the particle, resulting in a face to edge association which results in a higher swelling potential [17] [18] [19]. At the higher bentonite contents (i.e., 15 and 20%) where the hydration force is higher, the swelling is greater when the sample interacts with pure water than with the acidic water.

At 20% moisture content, the sample compression is higher compared to the swelling. Even at a bentonite content of less than 15%, the sample tends to compress. This compression is problematic when occurs horizontally as it results in cracks [42], [43]. During lateral compression, the shear strength of the soil decreases and its permeability increases. This behavior needs to be considered in determining an acceptable zone as a clay liner in a landfill application.

These findings are consistent with the results obtained from the FTIR test, where the samples 90C10B-10(A) and 80C20B-10(A) have higher peaks, especially at a wavelength of 3402–3445 cm<sup>-1</sup> (Fig. 3). At this wavelength, the samples absorb more -OH, so that the swelling is high (90C10B-10(A)) as shown in Fig. 5(a). Meanwhile, the swelling seems to be smaller in the 80C20B10(A) sample than in the 80C20B10(W) sample (Fig. 5(a)) owing to compression, as seen in Fig. 5(b).

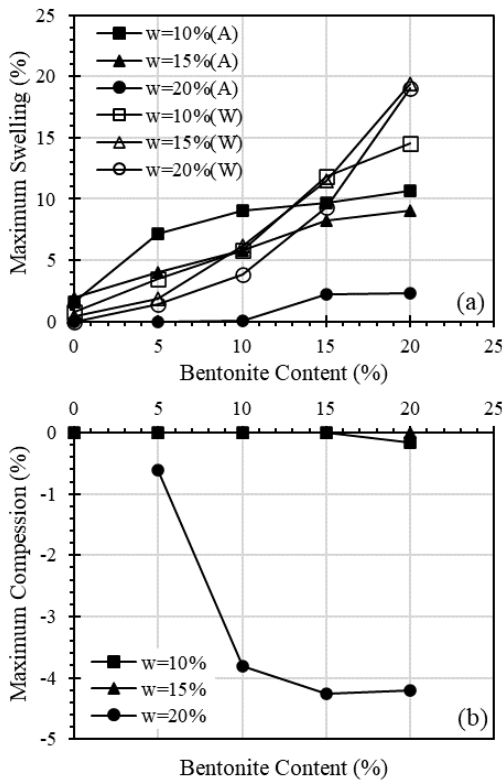


Fig. 5. (a) Maximum swelling and (b) maximum compression as a function bentonite content.

### 488 3.3. Effect of swamp acidic water on compression of sample

489 Figs. 6(a)–6(d) show the results of the consolidation test in 900  
 490 normalized void ratio versus logarithmic pressure for samples 901  
 491 with bentonite content of 5, 10, 15, and 20%, respectively. 902  
 492 The normalized void ratio was used so that the effect of 903  
 493 bentonite content and acidic water on the initial void ratio 904  
 494 after swelling could be excluded in the assessment. Each 905  
 495 sample's initial void ratio was added with a number to start at 906  
 496 1.0. For the same sample, the number was appended to all of 907  
 497 the void ratio data. In general, it is seen that the volume 908  
 498 change indicated by the largest change in void ratio occurs in 909  
 499 the sample with an initial moisture content of 10%. This is 910  
 500 due to the orientation of clay particles, which tend to fluctuate 911  
 501 at low water contents and the dominant formation of 912  
 502 macropores [38]. Macropores, or interaggregate pores, are 913  
 503 pores that exist between soil aggregates. When the sample is 914  
 504 compressed, the part that is greatly reduced is macropores 915  
 505 [44].

506 The results in Fig. 6 also show that the sample compacted 916  
 507 at 10% water content interacted with the acidic water to 917  
 508 produce the largest volume change. However, when compared 918  
 509 to the one tested in the pure water, this change is still smaller. 919  
 510 In the acidic water with higher concentrations, the 920  
 511 intergranular attraction force increases so that the particles 921  
 512 tend to flocculate [28]. Resistance to external forces becomes 922  
 513 greater and results in lower compressibility. This result is

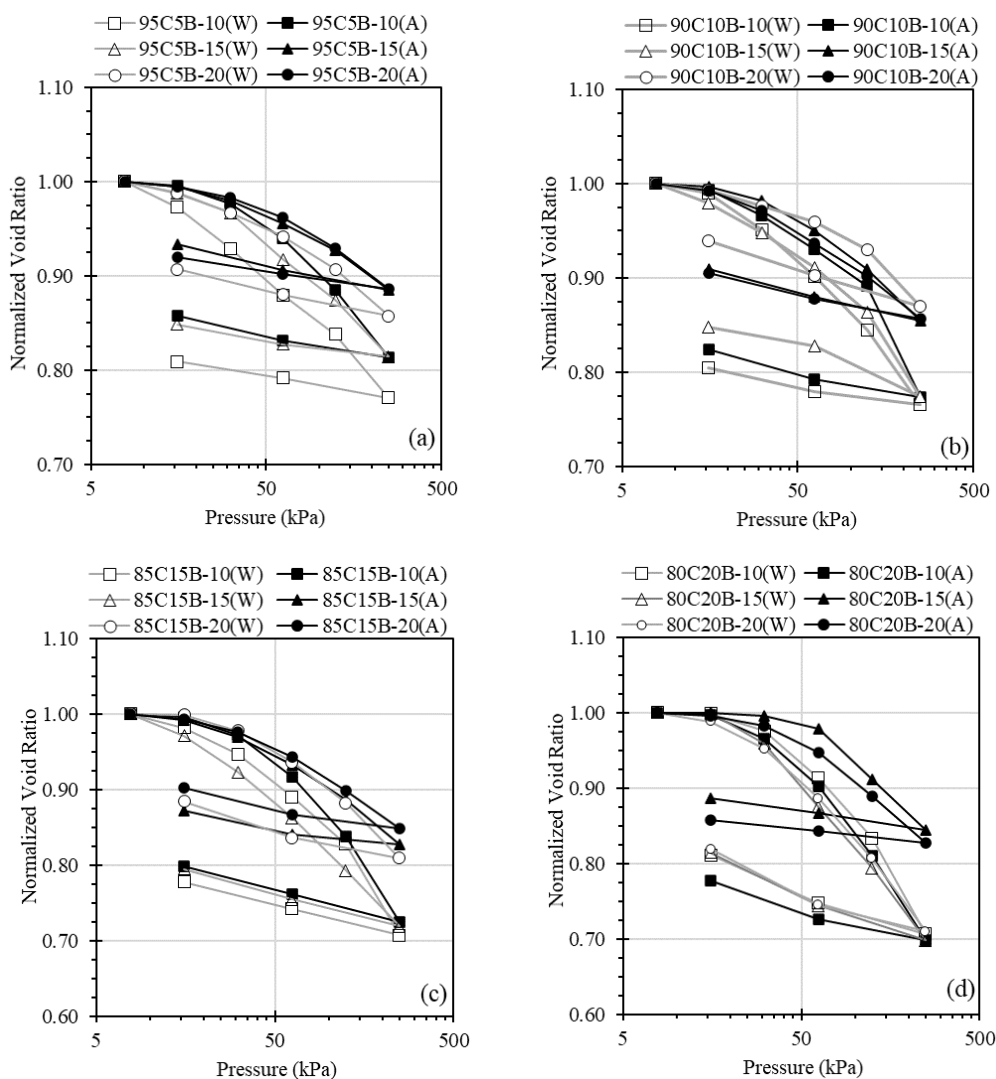


Fig. 6. Figure Normalized void ratio–logarithmic pressure relationship of compacted claystone–bentonite mixtures for Samples with (a) 5% bentonite, (b) 10% bentonite, (c) 15% bentonite, and (d) 20% bentonite. (Note: W=pure water, A=the acidic water).

514 supported by the FTIR test as shown in Fig. 3. The sample 518  
 515 tested with the acidic water shows more -OH extraction 519  
 516 wavelength of  $3402\text{--}3445\text{ cm}^{-1}$  due to the low compressibility 520  
 517 that leaves more water on the surface of the clay minerals. 521

518 Figs. 7(a) and 7(b) show the coefficient of compression ( $c_c$ ) 522  
 519 and swelling index ( $c_s$ ) of the claystone–bentonite mixture 523  
 520 interacting with pure water (W) and the acidic water (A). 524  
 521 general, it can be seen that  $c_c$  increases with increasing 525  
 522 bentonite content. In addition, the samples with lower initial 526  
 523 moisture contents exhibit higher  $c_c$  regardless of the solution 527  
 524 in which the consolidation test was performed. The effect of 528  
 525 acidic water on  $c_c$  was seen in the samples with an initial 529  
 526 water content of 10%, whereas for those with 15% and 20% 530  
 527 water content, the  $c_c$  from the tests in the acidic water was 531  
 528 smaller than that in pure water. 532

529 Gratchev and Towhata [11] reported that the effect of 533  
 530 acidic water on soil compressibility is influenced by mineral 534  
 531 soil structure, and diffuse double layer. When interacting with 535  
 532 acidic water, in certain soils, mineral leaching occurs 536  
 533 resulting in high compressibility. Changes in soil mineralogy 537  
 534 were not found in this study, as shown in Fig. 2 and Table 538  
 535 At bentonite contents up to 10%, where the dominant 539  
 536 behavior of bentonite is not maximum, soil structure tends 540  
 537 to be more flocculated when interacting with the acidic water 541

518 due to adsorption of  $\text{H}^+$  at the tip of the soil particles [17] [18]  
 519 [19]. Such a structure results in a large amount of  
 520 compressibility. However, when bentonite effect begins to be  
 521 prevalent i.e. at the percentage of more than 10%, the  $c_c$  value  
 522 decreases due to the collapse of the diffuse double layer [45]  
 523 [11].

As with the  $c_c$  value, for the compression in pure water,  $c_s$   
 also increases with increasing bentonite content in the  
 mixture. At the same bentonite content,  $c_s$  for the sample with  
 higher initial water contents tends to produce higher  $c_s$  value  
 due to the high repulsion between sample particles. Different  
 results have been seen in the tests with the acidic water, where  
 at high bentonite contents (i.e., 20%),  $c_s$  value decreases for  
 samples prepared at high initial water contents (i.e., 15% and  
 20%) tended to decrease due to collapse of the diffuse double  
 layer structure.

Besides the magnitude of volume change parameters  
 presented by  $c_c$  and  $c_s$ , the time effect needs to be given  
 consideration. This can be presented by the coefficient of  
 consolidation ( $c_v$ ). Fig. 8 shows the variation in  $c_v$  values as a  
 function of bentonite content. In general,  $c_v$  decreases with  
 increasing bentonite content. This condition is increasingly  
 seen at high bentonite contents, which is caused by reduced  
 sample permeability as the pores between claystone are filled



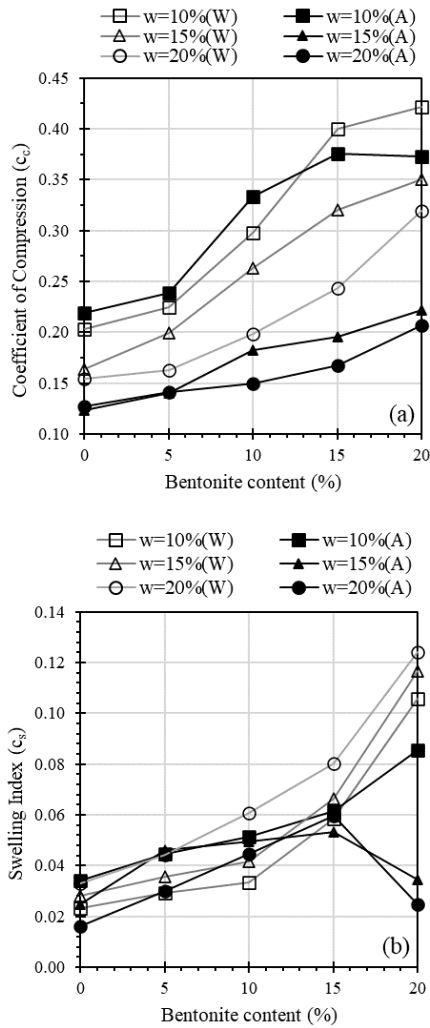


Fig. 7. Effects of acidic water on the compression parameters as a function of bentonite content (a) Coefficient of compression, and (b) swelling index.

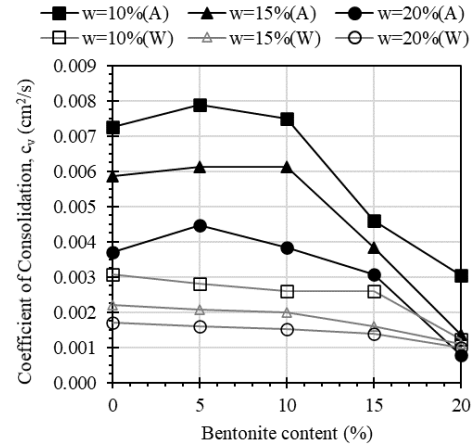


Fig. 8. Coefficient of consolidation as a function bentonite content.

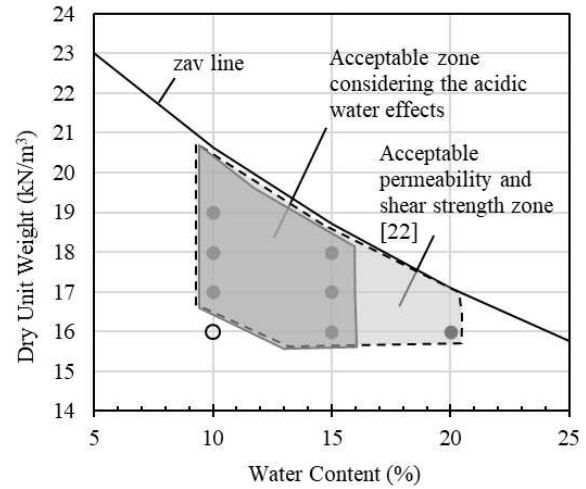


Fig. 9. Acceptable zone of claystone–bentonite mixture considering acidic water effects.

562 with bentonite [46]. At the same bentonite content, the same  
 563 with a higher moisture content has a smaller  $c_v$ . This indicates  
 564 the dominance of micropores in the behaviour of compacted  
 565 mixtures [38][44] at low permeability.  
 566 For the tests in the acidic water, a higher  $c_v$  is seen at the  
 567 same bentonite content when compared with the tests in pure  
 568 water. This is observed especially at low bentonite contents,  
 569 where flocculation of particles occurs due to clay interaction  
 570 with the acidic water. This results in greater permeability and  
 571 thus accelerates the consolidation process. As mentioned  
 572 above, the bentonite content is higher than 10% where the  
 573 diffuse double layer is dominant. The distance between particles  
 574 tends to be small due to the drop of the diffuse double layer  
 575 sample that interacts with the acidic water. This causes the  
 576 consolidation process to be more prolonged.  
 577 Arifin et al [24] recommended an acceptable zone for  
 578 claystone–bentonite mixtures considering their permeability  
 579 and shear strength. The widest zone is for the mixture with  
 580 20% bentonite content, which covers almost the entire area  
 581 both the wet of optimum (WOP) zone (i.e.,  $w=20\%$ ) and the  
 582 dry of optimum (DOP) zone (i.e.,  $w=10\%$ ), as shown in  
 583 in the dash line filled with light gray. However, other aspects  
 584 such as the potential for desiccation, resistance to chemical  
 585 attack, interfacial friction with the geomembrane, and the

ability to deform without cracking, should also be considered  
 [47]. Considering the effect of the acidic water on the volume  
 change (compression), especially in the samples with a  
 moisture content of 20% (i.e., the WOP zone), the acceptable  
 zone for the clay liner shown in dark gray is narrower than  
 previously (Fig. 9).

#### 4. Conclusions

Volume changes of compacted claystone–bentonite mixtures in the form of swelling and compression affected by swamp acidic water have been described and discussed. The initial moisture content of the sample affects the swelling of compacted claystone bentonite mixtures in acidic water. The sample tends to compress when the moisture level is higher than the wet of optimum. Compression increases as the amount of bentonite in the mixture increases. There is a noticeable behavioral difference between samples having more than 10% bentonite. Compression occurs faster in this condition than in pure water. A mixture with 20% bentonite content compacted at dry to optimum moisture content is the best for mitigating the negative effects of acidic water.

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# Volume Change in Compacted Claystone–Bentonite Mixture as Affected by the Swamp Acidic Water

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## Abstract

Water containing sulfuric acid with a pH up to 3 is prevalent in swampy areas. This article focuses on the effects of the solution on volume change of compacted claystone–bentonite mixture. Claystone was obtained from Banjarbakula landfill and it was mixed with bentonite on a 5, 10, 15, and 20% dry mass basis. Samples possessed the dry density of 16 kN/m<sup>3</sup> and moisture content of 10, 15, and 20%. The odometer examined the samples' swelling and compression in both pure and acidic water. Characterization tests i.e., XRF, XRD, and FTIR were also performed. The results showed that swelling and compression were affected by initial moisture and bentonite content. Samples with a moisture content of 20% showed compression in acidic water. Acidic water changed the water absorbed on the clay surface without altering the mineral. A mixture containing 20% bentonite compacted to optimum moisture content was found at best in reducing the acidic water effects.

Keywords: claystone; bentonite; swelling; compression; clay liner

## 1. Introduction

Numerous materials have been proposed as waste barrier one of which is a mixture of claystone and bentonite. Along with the clay minerals it contains, claystone is used to recycle waste material from excavation [1]. Previously, claystone from excavation was considered as an undesirable construction material, particularly when it came into contact with water [2], [3]. During the development of the Banjarbakula landfill in Banjarbaru, South Kalimantan, Indonesia, an approximately 8000 m<sup>3</sup> of claystone was dumped for being seen undesirable. In fact, the economic and environmental concerns should be addressed from the use of this material [1], considering some economic benefits from the utilization of this material.

Hydraulic conductivity, shear strength, compressibility and swelling characteristics are some of the properties commonly evaluated in relation to the use of bentonite-based materials as a landfill barrier. These properties are strongly influenced by the bentonite content in the mixture. Khalid et al. [4] found that the influence of bentonite on the geotechnical properties was more evident at a bentonite percentage of more than 10% for clay–bentonite mixture. Meanwhile, adding more than 20% bentonite to silty sand had no effect on the hydraulic conductivity of the clay liner [5].

Clay liners, as a barrier, are extremely prone to interaction with substances other than water. In the nuclear waste repository, the sealing material will interact with the saline

solution of the surrounding host rock. This will bring an effect on the canister's corrosion, the swelling and self-sealing capability of the bentonite back fill, and a sophisticated geochemical calculation [6]. Wang et al. [1] found that, due to the high sample density and low salinity of the water utilized, water chemistry had no effect on the swelling behavior of compacted claystone–bentonite mix. The swelling pressure of compacted claystone–bentonite mixture is affected by the final dry density of bentonite in the mixture, while the claystone used is considered to behave as sand [1].

Claystone, on the other hand, is highly impacted by the minerals it contains. Its combination with bentonite will bring effect on the mixture's behavior overall. The swelling capacity of bentonite is also determined by the chemistry of saturating fluids; the higher the salinity, the lower the sample's swelling capacity in which has a negligible effect on samples with a high density (i.e. 17–19 kN/m<sup>3</sup>) [1].

Besides density, water salinity has an effect on hydromechanical materials containing a large amount of smectite (i.e. 50% bentonite) [6]. Apart from swelling characteristics, Siddiqua et al. [6] examined the influence of salt on compression and swelling indices (i.e.,  $c_c$  and  $c_l$ ) obtained by consolidation tests.  $c_c$  was found to decrease in the presence of saline solution, indicating its influence on the sample's compressibility behavior. On natural stiff clay, the similar results were reported by Ngunyen et al. [7]. Clays with a high smectite content experienced more alterations than others.

Sealing materials may also interact with acidic liquids in

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82 addition to salt. Acidic water, which is generated by acid rain  
 83 and has a pH of 3–4.5, reduces the shear strength  
 84 sedimentary and igneous residual soils as well as increases  
 85 their permeability [8]. Acid rain infiltration into the soil  
 86 causes leaching of Fe<sup>3+</sup> and Al<sup>3+</sup>, which plays an important  
 87 role in cementation. The effect of acid rain on the  
 88 development of soil erosion was also investigated by  
 89 Matsumoto et al.[9]. The release of Al<sup>3+</sup> owing to fluids with  
 90 pH of 2–6 was also observed in the study in which it resulted  
 91 in the development of soil erosion. Meanwhile, Ahmed et al.  
 92 [10] found that the swelling ratio reduced when the pH in  
 93 pores decreased due to acid water. Gratchev and Towhata [11]  
 94 investigated the potential of changes in the compressibility  
 95 marine clay due to soil contamination from the past waste  
 96 mismanagement. It is reported that acid water increases  
 97 decreases the compressibility index dependent upon the  
 98 minerals and soil structure. Le et al. [12] investigated a coastal  
 99 acid sulfate soil in Australia containing sulfidic mineral (FeS<sub>2</sub>).  
 100 The results of the compressibility test showed that the  
 101 physical structure of the soil was determined by H<sup>+</sup> and Ca<sup>2+</sup>  
 102 cations. In a short time, the effect appeared to be insignificant.  
 103 Besides time, however, the combination of pore water  
 104 chemical composition, compressive pressure, and moisture  
 105 content affected the permeability of the acid sulfate soil [12].  
 106 Acidic water has also been reported to cause damage to  
 107 industrial areas for being contaminated with sulfuric acid  
 108 which has been widely used in paper industry, petroleum  
 109 refining, copper leaching, inorganic pigments, and organic  
 110 chemical industry [13]. In the soil, it was found that 1N  
 111 H<sub>2</sub>SO<sub>4</sub> resulted in the formation of gypsum and corneilite  
 112 whereas 4N H<sub>2</sub>SO<sub>4</sub> formed alunite and chloritoid. Mineral  
 113 changes in the black cotton soil used in the study resulted in  
 114 an increase in percent swelling. In addition, acid solution with  
 115 a higher concentration also produced a greater swelling  
 116 potential [13]. Numerous researchers have also reported soil  
 117 heaving induced by acidic solutions [14]–[16]. Sridharan et al.  
 118 [14] studied the incidence of floor, pavement and foundation  
 119 distress in a fertilizer factory. The damage was determined to  
 120 be the result of heave induced by phosphoric acid reaction  
 121 with soil in an acidic environment. Assa'ad [15] reported the  
 122 incline of the storage tank at the chemical fertilizer factory in  
 123 Aqaba, Jordan was caused by phosphoric acid leaking and  
 124 interacting with the subgrade soil. Like a gel, phosphoric  
 125 compounds are formed and fill the pores, which generate the  
 126 trapped gases from the chemical process. The generated  
 127 pressure causes the tank to lift when it is empty. Rama Varan  
 128 Prasad et al. [17] investigated the swelling potential of the  
 129 soils, namely black cotton soil, sodium bentonite, and  
 130 kaolinite, using two acidic solutions (i.e., H<sub>2</sub>SO<sub>4</sub> and H<sub>3</sub>PO<sub>4</sub>).  
 131 The results then showed that the swelling potential of  
 132 montmorillonite soils was determined by the type of cation  
 133 exchangeable. The cation exchange reaction and the  
 134 dissolution of some minerals resulted in mineral changes in  
 135 the montmorillonite soil, which affected its swelling behavior.  
 136 In kaolinite soils, the adsorption of H<sup>+</sup> at the broken ends  
 137 resulted in a face-to-edge association of the particle, which  
 138 caused an increase in the swelling potential of the soil  
 139 coupled with changes in soil mineralogy. Chen et al. [18]  
 140 investigated the compressibility of kaolinite soil using a  
 141 fluid with a dielectric constant larger than water, such as

acetic acid. The results indicated that the compression and  
 swelling index samples in the solution were smaller than that  
 of in water. Meanwhile, Wahid et al. [19] concluded that  
 kaolinite is not affected by salinity but pH, which attacks the  
 tip of the particle. The compression that occurs under constant  
 load is caused by the interaction of kaolinite with acid  
 solution as a result of sliding between particles and is  
 irreversible.

In South Kalimantan, the area is predominantly swampy  
 and low land. In areas where the soil is predominantly peat,  
 the presence of sulfuric acid in river water causes the pH to  
 vary from 3.4–4.2. The pH does not increase even during  
 rainy season due to high precipitation resulting in increasing  
 water levels in the river, thereby preventing the entry of  
 seawater into the river [20]. This can occur in any locations  
 with a large area of peat wetland. Tevetkov [21] provided data  
 in countries with peat swamp areas, including Russia (150  
 million ha), Indonesia (26 million ha), the United States of  
 America (40 million ha), Canada (170 million ha), Finland (10  
 million ha), China (3.5 million ha), Sweden (7 million ha),  
 and Ireland (1.2 million ha), as well as the remaining 12.3  
 million ha in Malaysia, Germany, Poland, the United  
 Kingdom, and Belarus. Wind-Mulder et al. [22] reported that  
 water chemistry data from four peat swamp areas in Canada  
 showed the average pH of 3.7–3.9 with a predominant of  
 SO<sub>4</sub><sup>2-</sup>. Therefore, the acidic water has a high potential of  
 reacting with the clay liner surrounding it. This paper aims to  
 examine the effect of swamp acidic water on the volume  
 change (i.e., swelling and compression) of the  
 claystone–bentonite mixture. An odometer was used to  
 evaluate samples of claystone and bentonite mixtures with  
 various compositions in acid water as immersion.

2. Materials and Methods

This study used both natural and fabricated clays (i.e.,  
 claystone and bentonite). Meanwhile, the acidic water utilized  
 was directly obtained from a swampy area to explore its  
 composition and effects on the clay liner. Overall sample  
 preparation, compaction, and volume change tests were  
 carried out in the laboratory at room temperature.

2.1. Claystone

The claystone used was taken from the Banjarbakula  
 landfill project site. The soil however not used in the project  
 and was disposed of. The claystone had a moisture content of  
 2.76%, Gs 2.6, a liquid limit (LL) of 40%, a plastic limit (PL)  
 of 20%, and a shrinkage limit (SL) of 15%. The material  
 meanwhile consisted of 4.5% sand, 43.9% silt, and 51.6%  
 clay. According to the Unified Soil Classification System  
 (USCS) [23], the claystone is classified as an inorganic  
 clay with low to medium plasticity (CL). The main exchangeable  
 cation claystone used was Ca<sup>2+</sup> 4.3 meq/g and the remainder  
 was Na<sup>+</sup> 0.3 meq/g, Mg<sup>2+</sup> 0.1 meq/g, and K<sup>+</sup> 0.3 meq/g. At a  
 dry unit weight of 16 kN/m<sup>3</sup>, the compacted claystone had a  
 hydraulic conductivity of 7.9×10<sup>-9</sup> m/s [24]. This value is  
 greater than the one required for clay liners in many countries  
 (i.e., 1.0×10<sup>-9</sup> m/s) [25].

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241 2.2. Bentonite

242 The bentonite used was commercial one with the main  
 243 exchangeable cation of Ca<sup>2+</sup> 18.7 meq/g and the others (i.e.  
 244 Na<sup>+</sup>, Mg<sup>2+</sup>, and K<sup>+</sup>) were 0.34 meq/g, 0.2 meq/g, and 0.293  
 245 meq/g, respectively. The bentonite had a moisture content  
 246 14.17%, a specific gravity of 2.71, LL 351.71%, PL 44.68265  
 247 SL 41.89%, and a plasticity index (PI) of 307.03%. The  
 248 material meanwhile consisted of 1.4% fine sand, 8.3% silt,  
 249 and 90.3% clay.

250 2.3. Acidic water

251 Acidic water was taken from a river in Tanipah village,  
 252 Barito Kuala district in South Kalimantan. The water had a pH  
 253 of 3.4–3.6. This pH tends to remain constant throughout the  
 254 year in both the dry and rainy seasons. Table 1 presents  
 255 chemical composition of the acidic water.  
 256 The chemical compounds dominant in the solution  
 257 included Ca<sup>2+</sup>, SO<sub>4</sub><sup>2-</sup>, and Cl<sup>-</sup>. The high concentration of  
 258 sulfate ions was as a result of pyrite oxidation occurred in the  
 259 soil [20]. Commonly, the SO<sub>4</sub><sup>2-</sup>/Cl<sup>-</sup> ratio is used to determine  
 260 the influence of sulfuric acid on pyrite oxidation on the  
 261 composition of river water in swamp areas.

262 Table 1. Chemical compositions of the acidic water used.

Chemical compound	K <sup>+</sup>	Ca <sup>2+</sup>	Na <sup>+</sup>	Fe <sup>3+</sup>	Mn <sup>2+</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>
mg/l	4.21	158.86	6.910	4.876	1.427	261.02	153.4

263 2.4. Sample preparation

264 Bentonite was mixed with claystone at a percentage of 5%,  
 265 10%, 15%, and 20% based on its dry weight. Water was then  
 266 added to the mixtures at a certain amounts to provide the  
 267 samples as the initial moisture contents (w) of 10%, 15%, and  
 268 20%. The sample target water contents were based on the  
 269 results of the Proctor standard compaction test on the  
 270 claystone with an optimum moisture content (OP) of 15%  
 271 obtained. As a result, the water contents of 10% and 20%  
 272 on the dry of optimum (DOP) and the wet of optimum (WOP)  
 273 water content, respectively. Subsequently, the mixtures were  
 274 statically compressed with a hydraulic jack to produce  
 275 samples of dry unit weight (γ<sub>d</sub>) of 16 kN/m<sup>3</sup>. The samples had  
 276 a diameter of 63.4mm and a height of 20mm. Table 2  
 277 illustrates the initial conditions and sample identifications  
 278 (Sample IDs). The names have been given following the  
 279 sample conditions, such as composition and initial water  
 280 content.

281 2.5. Swelling and Compression tests

282 Two tests were carried out in the odometer, namely the  
 283 swelling potential and the compression tests. These tests were  
 284 performed based upon the standard ASTM procedures (i.e.  
 285 ASTM D4829–11 [26] and ASTM D2435–04 [27]). The  
 286 water used in the test was pure water with a pH of ±7 and  
 287 swamp acidic water with a pH of 3.4. The tests using the  
 288 waters were carried out separately. For the test with pure

289 water, the sample in the odometer was immersed in the water  
 290 under a pressure of 6.9 kPa to obtain the sample's swelling  
 291 strain. After equilibrium was reached in which it was  
 292 observed from constant dial gauge readings, the sample was  
 293 loaded and subsequently unloaded in accordance to the  
 294 consolidation test procedure [27]. Similar procedures were  
 295 also carried out for the samples tested using swamp acidic  
 296 water.

297 2.6. Sample characterization

298 The investigation on the effects of acidic water on the  
 299 mixtures of claystone and bentonite commences with the  
 300 Atterberg limit tests was carried out to determine the liquid  
 301 limit, plastic limit, and plasticity index of the samples. Similar  
 302 approach has been also adopted by a number of other  
 303 researchers [8][17][28].

Table 2. Sample initial conditions

Sample ID	Claystone (%)	Bentonite (%)	γ <sub>d</sub> (kN/m <sup>3</sup> )	w (%)
100C–10	100	0	16	10
100C–15	100	0	16	15
100C–20	100	0	16	20
95C5B–10	95	5	16	10
95C5B–15	95	5	16	15
95C5B–20	95	5	16	20
90C10B–10	90	10	16	10
90C10B–15	90	10	16	15
90C10B–20	90	10	16	20
85C15B–10	85	15	16	10
85C15B–15	85	15	16	15
85C15B–20	85	15	16	20
80C20B–10	80	20	16	10
80C20B–15	80	20	16	15
80C20B–20	80	20	16	20

306 The acidic water has a physical influence on clay and can  
 307 cause chemical and mineral changes with the clay. The  
 308 alterations in the mineral contents were investigated using X-  
 309 ray diffraction (XRD) analysis for the samples before and  
 310 after the test with the acidic water. In addition, Fourier-  
 311 transform infrared spectroscopy (FTIR) test was used to  
 312 analyze the functional groups of materials tested with pure  
 313 water and acidic water. Finally, the samples' chemical  
 314 compositions were measured using X-ray fluorescence (XRF).

315 3. Results and Discussions

316 3.1. Effect of swamp acidic water on sample characterization

317 Figs. 1(a) and 1(b) depict the influence of acidic water on  
 318 the claystone-bentonite mixture's liquid limit (LL) and plastic  
 319 limit (PL), respectively. Fig. 1(a) shows that LL increased  
 320 with the increasing bentonite concentration in both the pure  
 321 water and acidic water tests. This was plausible since the LL

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359 of the bentonite was found greater than that of the claystone.  
 360 It is also evident that the influence of bentonite content on  
 361 LL was observed at the bentonite concentrations greater than  
 362 10%. This finding is consistent with what was reported  
 363 Khalid et al. [4], who found that bentonite had an impact  
 364 the clay-bentonite combination at a concentration of higher

372 with the acidic water. The greatest difference in the PL of  
 373 around 5% was shown at the 20% bentonite content. Since  
 374 only minor change in the PL was observed for the tests using  
 375 the acidic water, the change in the PI was almost similar to the  
 376 change in the LL. An increase in the LL and PI with reducing  
 377 pH of the soil water was also reported by Bakhshipour et al.  
 378 [8].

379 Table 3 shows the oxides contents of claystone, bentonite,  
 380 and claystone-bentonite mixtures before and after interacting  
 381 with the acidic water obtained from the XRF test. The samples  
 382 tested were taken from those after the consolidation test with  
 383 different bentonite and initial water contents. According to  
 384 samples ID, the samples consisted of claystone, bentonite, and  
 385 the mixes with varying bentonite percentages (i.e., 5%  
 386 (95C5B) and 20% (80C20B)), and different initial moisture  
 387 contents (i.e., 10% and 20%). As shown in the table, claystone  
 388 and bentonite predominately contained SiO<sub>2</sub> with a percentage  
 389 of 55.6% and 54.6%, respectively, followed by Fe<sub>2</sub>O<sub>3</sub> as the  
 390 next oxide with a content of 19.3% and 23.4%, respectively.  
 391 Both materials also contained almost equal Al<sub>2</sub>O<sub>3</sub>, which is  
 392 15% and 14%, respectively. The rests were K<sub>2</sub>O, CaO and  
 393 TiO<sub>2</sub>.

394 Table 3. Oxides of claystone, bentonite, dan claystone-bentonite mixtures

Sample ID	Condition	Compound (%)					
		Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	K <sub>2</sub> O	CaO	TiO <sub>2</sub>	Fe <sub>2</sub> O <sub>3</sub>
Claystone (C)	Before the test	15	55.6	4.33	3.22	1.83	19.3
Bentonite (B)	Before the test	14	54.6	0.56	4.10	1.82	23.4
95C5B-10	After the test	14	55.1	3.90	2.98	1.91	21.2
95C5B-20	After the test	14	53.8	4.01	3.01	1.93	21.4
80C20B-10	After the test	13	54.8	3.43	3.17	1.93	22.6
80C20B-20	After the test	14	53.1	3.41	3.33	1.90	22.6

395 Bakhshipour et al. [8] reported the leaching of Al<sup>2+</sup>, Fe<sup>3+</sup>,  
 396 Si<sup>2+</sup>, K<sup>+</sup> and Ca<sup>2+</sup> due to acid rain infiltration, which resulted  
 397 in the reduced sample strength. Artificial acid rain (AAR)  
 398 was prepared by adding a certain volume of 0.005 M nitric acid  
 399 (HNO<sub>3</sub>) and sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) to deionized distilled water  
 400 with the pH values of 2, 3, 4, 5, and 5.6. In this study, samples  
 401 soaked in the acidic water with chemical contents as shown in  
 402 Table 1 did not affect the samples' oxide contents. The  
 403 contents of Al<sub>2</sub>O<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>, and SiO<sub>2</sub> as shown in Table 3 did  
 404 not alter for the samples with 5% and 20% bentonite contents.  
 405 Neither cation exchange nor leaching occurred during the  
 406 swelling and consolidation processes.

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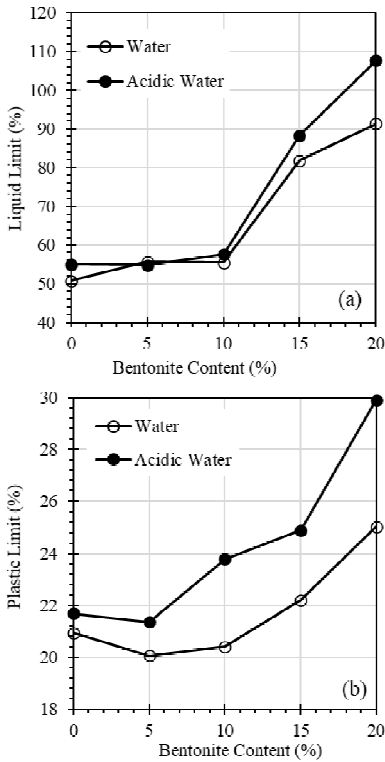


Fig. 1. Effect on acidic water on liquid limit and plastic limit of claystone-bentonite mixtures

365 than 10%. The LL of the samples tested with the acidic water  
 366 was consistently greater than those tested with pure water  
 367 containing more than 10% bentonite, as shown in Fig. 1(a).  
 368 LL increased up to 16% at the 20% bentonite content. At the  
 369 same bentonite content, LL was also found higher up to 16%  
 370 for the samples tested in the acidic water. An insignificant  
 371 increase in the PL was also observed when testing the samples

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Fig. 2 shows the XRD results of the claystone and bentonite samples (i.e., the bottom curve) and those interacting with the acidic water (denoted by (A)) for different bentonite contents (i.e., 5% and 20%) and initial water contents (i.e., 10% and 20%). As shown, the claystone sample contained more minerals than the bentonite sample, based on the number of peaks created by the XRD test. This was due to the fact that the claystone samples were collected directly from the nature without any purification or other processes. Claystone is composed of various minerals, including kaolinite (1), illite (2, 11), quartz (3, 6, 10, 12, 14, 19), vermiculite (5, 16), feldspar (7), montmorillonite (8, 17), chlorite (9), mica (13), and kaolinite (15, 18). In bentonite numerous minerals are found, including illite (2), feldspar (4), quartz (6), and montmorillonite (8, 17).

Table 4 summarizes the mineral composition of the samples before and after the test, based on the XRD test results as shown in Fig. 2. As seen in Table 4, no minerals were dissolved or formed as a result of interacting with the acidic water solutions. This result differed from the findings of previous investigations, including Sivapulliah et al. [13] and Rama Vara Prasad et al. [17]. This discrepancy might be explained by the low concentration and brief duration of the interaction (approximately 14 days) with the acidic water. According to Le et al. [12], the combination of acidic water concentration and interaction time has an effect on the solution's interaction with the soil. Apart from that, clay, particularly bentonite, has a very time-dependent behavior and the clayliner used must be able to sustain contamination throughout the waste decomposition process, which can take up to 50 years, and future research will be conducted over a longer period of time.

Table 4. Sample's mineralogy before and after the test

Mineral	Before the test		After consolidation (A)			
	Claystone	Bentonite	95C5B-10	95C5B-20	80C20B-10	80C20B-20
Illite	√	√	√	√	√	√
Quartz	√	√	√	√	√	√
Vermiculite	√	×	√	√	√	√
Feldspar	√	√	√	√	√	√
Mont.	√	√	√	√	√	√
Chlorite	√	×	√	√	√	√
Mica	√	×	√	√	√	√
Kaolinite	√	×	√	√	√	√

√ = available, × = unavailable

Fig. 3 shows the results of the FTIR test to determine the functional groups of the samples used, including their condition after interacting with the swamp acidic water. Samples with 10% and 20% bentonite contents were tested. In the figure, letters A and W signify that the samples were tested with the acidic and pure water, respectively. As seen, the peaks were found in the high wavelength region, i.e., at 1630, 3402, 3416, and 3620  $\text{cm}^{-1}$ . Each of these peaks indicated the presence of clay minerals (i.e., 3618–3628  $\text{cm}^{-1}$ ) [29]. The development of OH was found at 3402–3445  $\text{cm}^{-1}$  which is the interlayer and intralayer of the H bond [30].

Saputra et al. [31] also found a montmorillonite hydroxyl (OH) peak at 3434  $\text{cm}^{-1}$ . Ravindra-Reddy et al. [32] reported that this peak indicated the presence of water on the mineral surface. While at low wavelengths of 1009, 695, 528, and 470

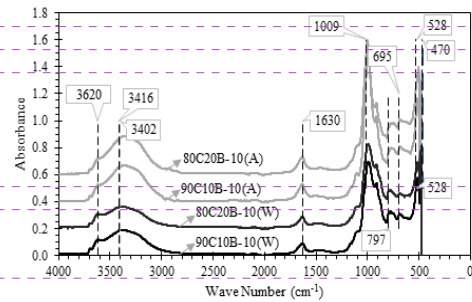


Fig. 3. Functional group of samples tested with pure water (W) and the acidic water (A)

$\text{cm}^{-1}$ , these peaks were the peaks of the  $\text{SiO}_4$  tetrahedron [32], where at 466–470 and 528–535  $\text{cm}^{-1}$  it is an indication of the presence of clay and silica minerals.

The FTIR results confirmed that the material used had clay minerals where  $\text{SiO}_4$  was present at 3618–3628  $\text{cm}^{-1}$  [29]. Moreover, the indications of the presence of the mineral montmorillonite could be seen from the development process at a wavelength of 3402–3445  $\text{cm}^{-1}$  when interacting with pure water. The expansion over this range (i.e. 3402–3445  $\text{cm}^{-1}$ ) for the samples interacting with the acidic water (A) was higher than those with pure water (W). This showed that the clay surface absorbed more water when interacting with acidic water, as seen from the –OH extraction at a wavelength of 3402–3445  $\text{cm}^{-1}$ . The amount of bentonite in the mixture did not appear to affect the extraction intensity of the –OH samples. The impact of acid on montmorillonite was almost similar whenever the –OH extracting occurred at a wave length between 3441  $\text{cm}^{-1}$  [33] and 3427  $\text{cm}^{-1}$  [34].

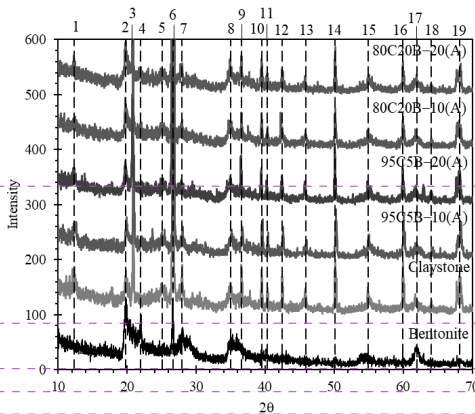


Fig. 2. Mineralogy of samples before and after tested by the acidic water

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## 517 3.2. Effect of swamp acidic water on swelling

518 Because the test was conducted on an odometer, the 518  
 519 deformation occurred was in the vertical direction, with 519  
 520 any changes in the horizontal. As a result, the amount 520  
 521 deformation was proportional to the volume change of 521  
 522 sample. Figs. 4(a)–(f) show the typical swelling developm 522  
 523 of claystone–bentonite mixtures with time when interacti 523  
 524 with pure water (i.e., Figs. 4(a), 4(c), and 4(e)) and the acid 524  
 525 water (i.e., Figs. 4(b), 4(d), and 4(f)) under a 6.9kPa 525  
 526 plotted on a semi–logarithmic scale. For the sample with 10 526  
 527 initial moisture content as shown in Fig. 4(a), the deformati 527  
 528 samples increased slowly in the early stages of the test (i 528  
 529 up to 20 minutes). Primary swelling occurred rapidl 529  
 530 thereafter up to a certain point (i.e., up to 300–4000 minu 530  
 531 dependent upon the bentonite content in the mixture) slo 531  
 532 and reached maximum deformation. The maxim 532  
 533 deformation recorded was referred to as the maxim 533  
 534 swelling of the sample. The maximum swelling of claysto 534  
 535 was reached in less than 100 minutes. Fig. 4(b) shows that the

536 initial swelling occurred gradually in the beginning, up to 20  
 537 minutes, followed by primary swelling up to 300 minutes for  
 538 the samples with 5% bentonite content, and 3000 minutes for  
 539 those with 20% bentonite. The insignificant compression  
 540 occurred in sample 80C20B-10(A), which contained 20%  
 541 bentonite. The same behavior was also observed in the sample  
 542 with the initial moisture content of 15%, as shown in Figs.  
 543 4(c) and 4(d).

The effect of acid water on bentonite has begun to be seen  
 at low water content. As shown in Fig. 4(b), there was a  
 significant delay in increasing deformation for 80C20B-  
 10(A), and then the sample led to the maximum deformation.  
 The high concentration of ions contained in acidic water  
resulted in a balancing process with the soil water inside. As  
 shown in FTIR in Figure 3, after the acidic water began to be  
 absorbed by the bentonite surface, modifications occurred and  
 resulted in an increase in the amount of water absorbed on the  
 surface. This resulted in high swelling occurred, as indicated  
 by the vertical deformation of the sample.

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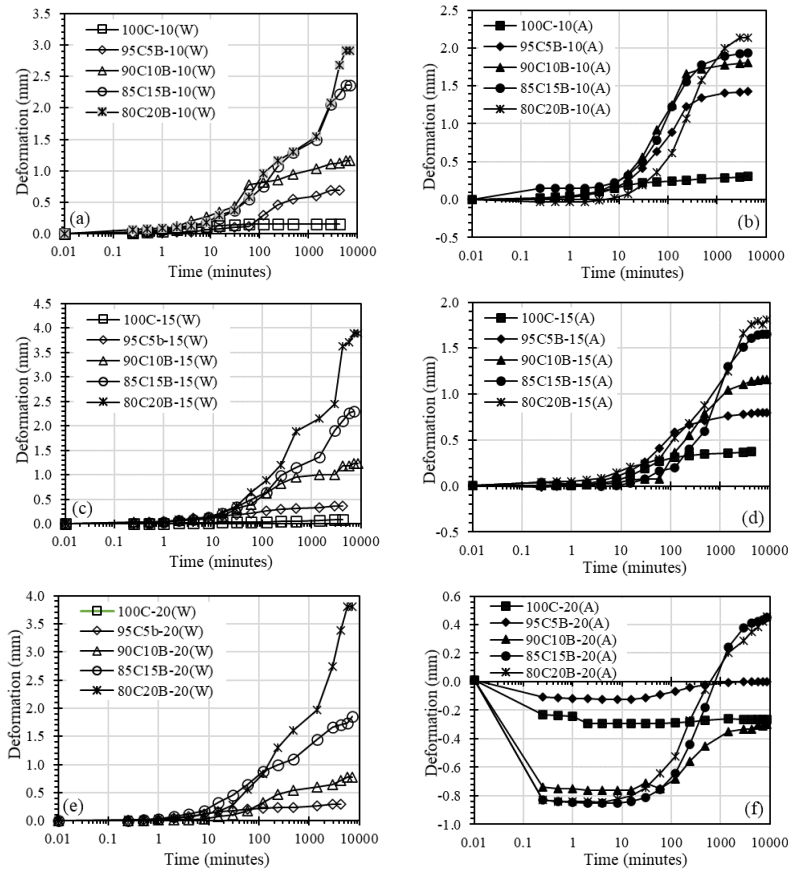


Fig. 4. Swelling development by time of claystone-bentonite mixture samples with initial moisture content of (a)–(b) 10%, (c)–(d) 15%, and (e)–(f) 20%.

578 Different behavior was noticed in the samples with 596  
 579 initial moisture content of 20%, where all samples 597  
 580 interacted with the acidic water tended to experience 598  
 581 shrinkage (or compression). Only two samples (i.e., 85C15B- 599  
 582 20(A) and 80C20B-20(A)) with 15% and 20% bentonite 600  
 583 content, respectively, swelled back past their initial 601  
 584 conditions. From this behavior, it can be seen that swar 602  
 585 acidic water has an effect on the mixtures with high bentonite 603  
 586 content or high initial water content. Claystone contain 604  
 587 clay minerals such as kaolinite and illite are not much affected 605  
 588 by the acidic water. This can be seen from the results of 606  
 589 Atterberg limit tests (Fig. 1). The unremarkable effect was 607  
 590 caused by the adsorption of H<sup>+</sup> at the broken end, resulting 608  
 591 a face-to-edge association of the particle [17] [18] [19]. 609  
 592 Swelling occurs due to the absorption of water by the clay 610  
 593 surface. It increases with the increasing percentage 611  
 594 bentonite in the mixture. Although both include 612  
 595 montmorillonite, bentonite contains a greater proportion of 613

mineral than natural soils [35]. Thus, by adding bentonite to  
 the mixture, the amount of montmorillonite in it increases.  
 Clay and montmorillonite contain are thought to have a  
 distinct influence on swelling behavior, ranging from minor to  
 major. However, it was revealed that the latter element had a  
 greater influence than the former [35]. The montmorillonite  
 containing bentonite, according to Pusch et al [36], requires  
 2–3 layers of water molecules to meet the hydration force.  
 Other researchers even reported 4 layers of water molecules  
 required [37]. The thickness and complete hydrated layers of  
 water molecules in bentonite vary depending on its  
 exchangeable cation. Assuming the specific surface area of  
 bentonite is 500 m<sup>2</sup>/g and the water unit weight of 1 g/cm<sup>3</sup>,  
 Arifin [38] reported that the water content to satisfy the  
 hydration force is 22.7%, 14.1%, 23.9%, and 15.4% for the  
 Mg, Ca, Na, and K types of bentonite, respectively. This water  
 content can even be greater because the surface water density  
 is reported to be possibly more than 1 gr/cm<sup>3</sup> [39]. After this

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water absorption, the role of surface hydration decreases. equalize ion concentrations, water molecules tend to diffuse toward the surface.

Numerous studies have previously observed that changes occur when soils, particularly those containing montmorillonite, interact with acidic solutions [13], [12], [16] [19]. This is not the case in this study, as shown in Fig. 2 and Table 4. There was even no cation exchange as shown in Table 3. The volume change occurred in the sample was due to the difference in the concentration of cations in the pure water and the acidic water. This process is known as osmotically-induced consolidation or osmotic consolidation [40], [41]. The high concentration of cations in the acidic water (Table 1) results in the outward flow from within the sample to balance these conditions. When the water content of the sample is high, the concentration of cations in the pore water decreases and tends to release water, which results in a decrease in the soil volume (Fig. 4(f)).

The results of the maximum percentage of swelling and compression occurred in the sample (Fig. 4) are summarized in Fig. 5. Fig. 5(a) shows the 10 percent bentonite level to be the limit of the distinct swelling behavior of the samples. At a bentonite content less than 10%, the swelling in the acidic water is higher than that of in pure water for the samples with an initial water content of 10% and 15%. In this condition, the behavior of claystone containing kaolinite is more dominant than kaolinite soils, acidic water will affect the tip of the particles resulting in a face to edge association, which results in a higher swelling potential [17] [18] [19]. At the high bentonite contents (i.e., 15 and 20%) where the hydration force is higher, the swelling is greater than with the acidic water when the sample interacts with pure water.

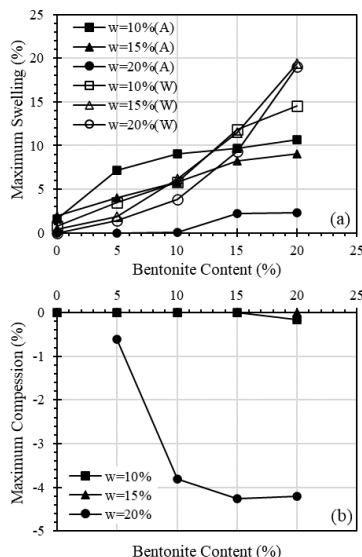


Fig. 5. (a) Maximum swelling and (b) maximum compression as a function bentonite content.

At 20% moisture content, the sample compression is higher compared to the swelling. Even at a bentonite content of less than 15%, the sample tends to compress. This compression is problematic when occurs horizontally as it results in cracks [42], [43]. During lateral compression, the shear strength of the soil decreases and its permeability increases. This behavior needs to be considered in determining an acceptable zone as a clay liner in a landfill application.

These findings are consistent with the results obtained from the FTIR test, where the samples 90C10B-10(A) and 80C20B-10(A) had higher peaks, especially at a wavelength of 3402–3445  $\text{cm}^{-1}$  (Fig. 3). At this wavelength, the samples absorbed more -OH, so that the swelling was high (90C10B-10(A)) as shown in Fig. 5(a). Meanwhile, the swelling seemed to be smaller in the 80C20B10(A) sample compared to the one in the 80C20B10(W) sample (Fig. 5(a)) owing to compression, as seen in Fig. 5(b).

### 3.3. Effect of swamp acidic water on compression of sample

Figs. 6(a)–6(d) show the results of the consolidation test in normalized void ratio versus logarithmic pressure for samples with bentonite content of 5, 10, 15, and 20%, respectively. The normalized void ratio was used so that the effect of bentonite content and acidic water on the initial void ratio after swelling could be excluded in the assessment. Each sample's initial void ratio was added with a number to start at 1.0. For the same sample, the number was appended to all of the void ratio data. In general, it can be seen that the volume change indicated by the largest change in void ratio occurred in the sample with an initial moisture content of 10%. This was due to the orientation of clay particles, which tended to fluctuate at low water contents and the dominant formation of macropores [38]. Macropores, or interaggregate pores, are pores that exist between soil aggregates. When the sample is compressed, the part that is greatly reduced is macropores [44].

The results in Fig. 6 also showed that the sample compacted at 10% water content interacted with the acidic water to produce the largest volume change. However, when compared to the one tested in the pure water, this change was still smaller. In the acidic water with higher concentrations, the intergranular attraction force increased so that the particles tended to flocculate [28]. Resistance to external forces became greater and resulted in lower compressibility. This result was supported by the FTIR test as shown in Fig. 3. The sample tested with the acidic water showed more -OH extraction at a wavelength of 3402–3445  $\text{cm}^{-1}$  due to the low compression leaving more water on the surface of the clay minerals.

Figs. 7(a) and 7(b) show the coefficient of compression ( $c_c$ ) and swelling index ( $c_s$ ) of the claystone–bentonite mixtures interacting with pure water (W) and the acidic water (A). In general, it can be seen that  $c_c$  increased with the increasing bentonite content. In addition, the samples with lower initial moisture contents exhibited higher  $c_c$  regardless of the solution in which the consolidation test was performed. The effect of acidic water on  $c_c$  was seen in the samples with an initial water content of 10%, whereas for those with 15% and 20% water content, the  $c_c$  from the tests in the acidic water was smaller than that of in pure water.

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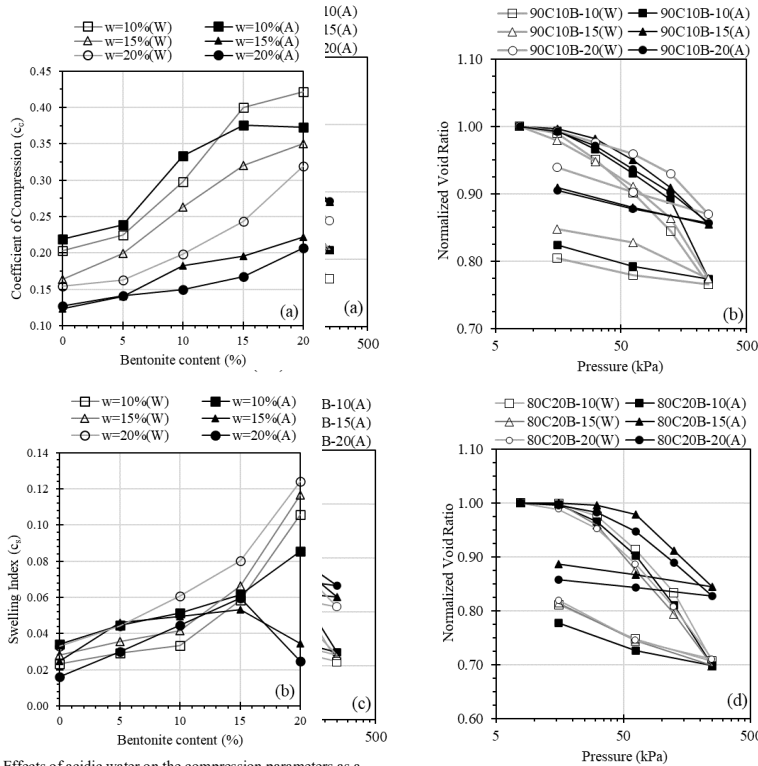


Fig. 7. Effects of acidic water on the compression parameters as a function of bentonite content (a) Coefficient of compression, and (b) swelling index.

Relationship of compacted claystone–bentonite mixtures for Samples and (d) 20% bentonite. (Note: W=pure water, A=the acidic water).

736 Gratchev and Towhata [11] reported that the effect 760  
 737 acidic water on soil compressibility is influenced by mineral 761  
 738 soil structure, and diffuse double layer. When interacting with 762  
 739 acidic water, in certain soils, mineral leaching occurs 763  
 740 resulting in high compressibility. Changes in soil mineralogy 764  
 741 were not found in this study, as shown in Fig. 2 and Table 765  
 742 At bentonite contents up to 10%, where the dominant 766  
 743 behavior of bentonite was not maximum, soil structure tended 767  
 744 to be more flocculated when interacting with the acidic water 768  
 745 due to the adsorption of  $H^+$  at the tip of the soil particles [17] 769  
 746 [18] [19]. Such a structure resulted in a large amount of 770  
 747 compressibility. However, when bentonite effect began to 771  
 748 prevalent i.e. at the percentage of more than 10%, the  $c_c$  value 772  
 749 decreased due to the collapse of the diffuse double layer [45] 773  
 750 [11]. 774  
 751 As with the  $c_c$  value, for the compression in pure water, 775  
 752 also increased with the increasing bentonite content in the 776  
 753 mixture. At the same bentonite content,  $c_c$  for the sample with 777  
 754 higher initial water contents tended to produce higher  $c_c$  values 778  
 755 due to the high repulsion between sample particles. Different 779  
 756 results have been seen in the tests with the acidic water, where 780  
 757 at high bentonite contents (i.e., 20%),  $c_c$  value decreased 781  
 758 samples prepared at high initial water contents (i.e., 15% and 782  
 759 20%) tended to decrease due to the collapse of the diffuse 783

double layer structure.

Besides the magnitude of volume change parameters presented by  $c_c$  and  $c_s$ , the time effect needs to be given consideration. This can be presented by the coefficient of consolidation ( $c_v$ ). Fig. 8 shows the variation in  $c_v$  values as a function of bentonite content. In general,  $c_v$  decreases with the increasing bentonite content. This condition is increasingly seen at high bentonite contents, which is caused by the reduced sample permeability as the pores between claystone are filled with bentonite [46]. At the same bentonite content, the sample with a higher moisture content has a smaller  $c_v$ . This indicates the dominance of micropores in the behavior of the compacted mixtures [38][44] at low permeability.

For the tests in the acidic water, a higher  $c_v$  was seen at the same bentonite content when compared with the tests in pure water. This was observed especially at low bentonite contents, where flocculation of particles occurred due to clay interacting with the acidic water. This resulted in greater permeability and thus accelerated the consolidation process. As mentioned above, the bentonite content was higher than 10% where the diffuse double layer was dominant. The distance between the particles tended to be small due to the drop of the diffuse double layer sample that interacted with the acidic water. This caused the consolidation process more

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804 prolonged.  
 805 Arifin et al [24] recommended an acceptable zone of  
 806 claystone–bentonite mixtures considering their permeability  
 807 and shear strength. The widest zone is for the mixture with  
 808 20% bentonite content, which covers almost the entire area,  
 809 both the wet of optimum (WOP) zone (i.e.,  $w=20\%$ ) and the  
 810 dry of optimum (DOP) zone (i.e.,  $w=10\%$ ), as shown in Fig. 9  
 811 in the dash line filled with light gray. However, other aspects,  
 812 such as the potential for desiccation, resistance to chemical  
 813 attack, interfacial friction with the geomembrane, and the  
 814 ability to deform without cracking, should also be considered  
 815 [47]. Considering the effect of the acidic water on the volume  
 816 change (compression), especially in the samples with a  
 817 moisture content of 20% (i.e., the WOP zone), the acceptable  
 818 zone for the clay liner shown in dark gray is narrower than  
 819 previously (Fig. 9).

820 **4. Conclusions**

821 The volume changes of compacted claystone-bentonite  
 822 mixtures in the form of swelling and compression affected by  
 823 swamp acidic water have been described and discussed. The  
 824 initial moisture content of the sample affected the swelling of  
 825 compacted claystone bentonite mixtures in acidic water. The  
 826 sample tended to compress when the moisture level was  
 827 higher than the wet of optimum. Compression increased as the  
 828 amount of bentonite in the mixture increased. There was a  
 829 noticeable behavioral difference between samples having  
 830 more than 10% bentonite. Compression occurred faster in this  
 831 condition than in pure water. A mixture with 20% bentonite  
 832 content compacted at dry to optimum moisture content was  
 833 found at best for mitigating the negative effects of acidic  
 834 water.

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 839 009.76/UN8.2/PL/2021.

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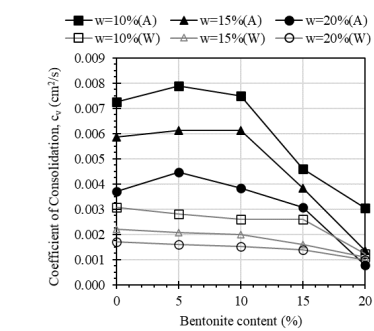


Fig. 8. Coefficient of consolidation as a function bentonite content.

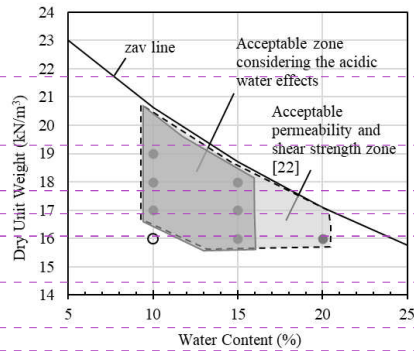


Fig. 9. Acceptable zone of claystone–bentonite mixture considering acidic water effects.

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