

# Appendix 4 – Recipe development

Biochar based capping of polluted sediment Biokolbaserad reaktiv barriär för täckning av förorenade sediment

Luleå tekniska universitet

2022-08-16



Research engineer: Wathiq al Jabbam



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# 1. Backgound

Luleå University of Technology (LTU), together with the Norwegian Geotechnical Institute (NGI) and Skellefteå municipality, is carrying out a pilot project funded by the Swedish Geological Survey (SGU) within the framework of the Governments mission *Contaminated Sediments (förorenade sediment)*. The pilot project regards the use of a biochar-based reactive barrier to cover contaminated sea sediments.

As part of the project, a pilot field experiment was carried out to cover a small area of sediment with a mixture of biochar (as reactive agent), bentonite (as the structural material) and salt (to facilitate the sedimentation of the material). The experiment was executed in the bay of Bureå outside an old industrial area, where Bure Träsliperi operated a wood grinding industry from 1928 until 1992. Wood pulp impregnated with the impregnation agent *Pulpasan,* containing methylmercury, has been transported into the Bay, and the sediment has also been contaminated with different PAH due to the trafficking of the bay during the time in which Bure Träslip was active. The sediment also contains high concentrations of trace elements.

This section describes the materials and experimental work used during this project to investigate the effectiveness of adding bentonite and biochar to cover the sea sediments. In this section, basic characteristics of sediments and a mixture of biochar and bentonite are evaluated as capping layer (cover layer) in addition to the testing procedures and methodologies of preparing samples which were utilized to obtain the main research objects.

The main goal of the project was to develop a slurry containing a mixture of bentonite clay and biochar to cover the seabed. The amount of biochar needed for the cover layer was predetermined while the amount of bentonite was tested to achieve a 5cm thick capping.

Sedimentation tests were conducted to simulate the distribution behaviors of the mixture in the field which the mixture sinks below the water and settled above the seabed as cover layer. Therefore, the mixture of bentonite and biochar should have good homogeneity and workability. Workable soil from a geotechnical point of view refers to the soil which can be easily handled and compacted homogeneously. Improving the workability of the mixture makes the mixture easy to be mixed, loaded, transported, pumped and self-compacted homogeneously, which subsequently leads to implementing the cover layer.

The key issue was to determine a proper water content to ensure homogenized and workable bentonite and biochar mixture, which leads to uniform distribution of mixture above the seabed and less water turbidity after sinking the mixture in water. The questions addressed in the study were:

- 1. How the mixture behaves with changing the water content?
- 2. Can the mixture be separated during sedimentation process?
- 3. What acceptable range of water content can provide workable mixture?
- 4. How much of the mixture will remain in suspension and how much will sink?
- 5. What is the effect of mixing duration on the consistency of mixture?
- 6. What is the effect of mixer types on obtaining homogenized mixture?
- 7. How the mixture will be distributed above the seabed?



# 2. Material

#### 2.1 Sediment

The sediment used in the project were excavated from seabed in three different site locations in Bureå, Skellefteå, Northeast part of Sweden. The site locations were named as test, reference and buck up area with dimensions 30\*30 m for each site. For each area, nine samples were excavated using van veen grabber. The sediments were classified by tests of particle size distribution, consistency limits, loss of ignition, and particle density. The physical and mechanical properties are presented in Table 1. The particle size distribution of the tested sediments when the oversize particles (mainly wood pieces) are removed or not from the samples are shown in Figure 1and Figure 2respectively.

The sediments were classified according to the Swedish standard. Organic content assessed by ignition test according to according to Swedish standards SS 027105 (1990) showed that organic content ranged between 11.9 to 35.1%. The sediments were classified as having medium to high organic content.

Parameters	Test	area	Reference area		Back up area		
Depth blow water, m							
Particle-size distribution (%)	Removing oversize's	Without removing oversize's	Removing oversize's	Without removing oversize's	Removing oversize's	Without removing oversize's	
Gravel (Wood) (%) (2-63mm)	0	4	0	4	0	4	
and (%) (1-0.63mm)	50	46	40	36	50	46	
Silt (%) (0.063 – 0.002 mm)	42	42	52	52	40	40	
Clay (%) (< 0.002 mm)	8	8	8	8	10	10	
Consistency limits (%)							
Liquid limit (%) *	1	08	137		238		
Plasticity limit (%)	8	80	88		128		
Plasticity index (%)	2	9	49		109		
Natural water content (%)	2	35	300		600		
Particle density	2.4	155	2.4		2.133		
Loss of ignition %	11.9	11.9 -13.3		17.5 -17.65		30.6 - 35.1	
Bulk density (g/cm3)	1.36		1.22		1.11		
рН							
Soil Classification	saclSi, sandy clayey Silt.		saclSi, sandy clayey Silt.		saclSi, sandy clayey Silt.		
	High plasticity		High pl	asticity	Very high	plasticity	

Table 1. Engineering properties of tested sediments.

\* Determined by the fall cone test.







*Figure 2 Particle size distribution of test sediments without removing the oversize's.* 



# 2.2 Capping layer (cover layer)

A mixture of bentonite and biochar was used as capping layer to cover the contaminated sea sediments. The amount of bentonite and biochar used in the mixture was calculated according to the surface area.

Table 2 described the amounts of bentonite and biochar that planned to use in kg/m<sup>2</sup> for field and laboratory investigation. The amount applied in field test is doubled to take into account material loss to surroundings.

Parameters	Unit	Laboratory investigation	Field investigation
Bentonite	kg/m²	7	14
Biochar	kg/m²	1.6	3.2

Table 2. Proposed mixture used during laboratory and field investigation.

# 3. Methods and Experimental Program

An extensive experimental program was initiated to determine a mixing recipe, which included tests of consistency limits, density, free swelling, dry and wet sieving, and sedimentation conducted on uncovered sediments as well as the mixture of bentonite and biochar.

# 3.1 Water content and density

Water content is defined as the ratio of the weight of water to the total weight of the solids. It is determined by heating the mixture in the laboratory oven at a temperature of 105 °C for 24 hours.

Bulk density of sediments was investigated by using a cylindrical tube by knowing its weights and dimensions.

# 3.2 Swelling test

A series of swelling tests were conducted to investigate effect of addition salt (NaCl) on the swelling potential of capping layer. The test was done by using bentonite only as capping layer and when the bentonite was mixed with 5% biochar. The salt content was added as a dried material at various ratios ranged from 0 to 4% of bentonite dry mass. The biochar's was added as 5% of bentonite dry mass. The mixture was mixed by hand untile become homogenized in the dry state.

The mixtures were poured in cylindrical glass jars (100 x 300 mm) partially filled with water by using a cone. The swelling for each mixture was measured at different interval times from 0 to 168 hours after pouring the mixture in the cylinder. The tests plan of salinity effects on the swelling behavior of the different mixture are presented in Table 3.



Mixture	Apparatus	Water (ml)	Salt (%)	Time (hours)
Bentonite	Cylindrical glass jars (100 x	1000		0 to 168 at various
only	300 mm)	1000	0, 0, 5, 1, 2, 5, 4	interval
Bentonite	Cylindrical glass jars (100 x	1000	0 0 5 1 2 4	0 to 168 at various
mixed with	300 mm)	1000	0, 0, 3, 1,2,4	interval
5% biochar's	Small bakers 200 ml	300	0, 0,5, 1,2,3,4	IIILEI Val

Table 3. Test program for the salinity effects on swelling behaviors.

# 3.3 Effect of water content

For laboratory investigation, specimens for the sedimentation tests were prepared by calculating the required amount of bentonite, biochar and salt (NaCl) for the circular area with 0.15 m diameter as described in Table 2. The salt content was added as one percent of the weight of bentonite. Different water amounts (see Table 4) were added to the bowl of the mixer and mixed with 1% salt for one minute, then bentonite and biochar were added and mixed for 15 minutes using a laboratory mixing machine (see Figure 3 (a)).

# 3.4 Effect of mixing time and mixer type

Specimens for the effect of mixing time and mixer types were prepared in an identical way to the sedimentation tests specimens for the effect of water content, but only adding a specified water content of 1450 and 1600 millilitre (water to solid ratio (W/S) 7.41 and 6.73), and mixed for various times by using different mixer types (see Table 4), to compare the effect of different mixing times and mixer size. Before sedimentation tests, the mixtures were tested for flowability and slump height by using the flow table test and slump test by using mini and standard cones as well as measuring the water content of the mixture.

The average of two tests represents the flowability and slump height, whilst the water content is based on the average of four tests.



Mixture	Test	ing program	Mixer Type	Mixing time (Minute)	Added Water (mL)	Water /solid (Bentonite + Biochar+salt) ratio	Water /Bentonite ratio
		Flow table test,	cake		1000	4.67	5.15
	Water content,EffectSedimentation	Water content,	mixer		1400	6.50	7.21
		with 7		1500	6.95	7.73	
	of	test,	speeds	15	1600	7.41	8.24
bentonite	water Dimensions content and thickness of mixture and	Dimensions and thickness of mixture and Turbidity	bowl size:3 litter		1800	8.36	9.31
6 of			cake	30		7.41	8.24
) 19		Flow table test,	mixer	60		7.41	8.24
aCl			FIOW LADIE LESL,	with 7	120		7.41
t (N	Z test.	speeds,	180	1600	7.41	8.24	
Salt		Water content,	bowl	240		7.43	8.27
and	Sedimentation	size:3 litter	300		7.41	8.24	
ſm2	r ty	, lesi, Dimensions	Dimensions Harbot 60				
Х Ю	ixe	and thickness	mixer , 3	120	1450	6.73	7.47
1,6	μ	of mixture and turbidity	speed,	180			
Jar	an		bowl	240			
Bioch	Bioch		liter	300			
Bentonite 7 kg/m2,	Effect of mixing	Flow table test, min slump test, standard slump test, Water content, Sedimentation test, Dimensions and thickness of mixture and turbidity	Concrete mixer bowl size: 200 litter	60 and 90	1450	6.73	7.48

Table 4. Overview of the sedimentation tests program.





(a) Cake mixer with 3 litre bowl





(c) Laboratory mixer with 200 litre bowl

Figure 3Different laboratory mixer used in the laboratory investigation.

# 3.5 Flowability and workability

Workable soil from a geotechnical point of view refers to the soil which can be easily handled and compacted homogeneously. Improving the workability of the mixture makes the mixture easy to be mixed, loaded, transported, pumped and self-compacted homogeneously, which subsequently leads to implementing the cover layer.

The workability of clay in the ceramic industry was defined by (Barnes, 2013<sup>1</sup> and 2018<sup>2</sup>) as how much working effort is needed for the clay to be molded or worked together with the ease or difficulty of working, measured by its toughness, T, (kJ/m3). These working efforts (toughness) are mainly controlled by the range of soil water content and subsequently by the plasticity of the clay. (Barnes, 2013) proposed a workability index (Iw), as expressed in equation 1, to measure the workability degree in soils.

$$I_W = \frac{\omega - \omega_P}{\omega_T - \omega_P} \qquad \qquad \text{eq. (1)}$$

Where:  $\omega$  : water content of soil,  $\omega_P$ : plastic limit and  $\omega_T$ : toughness limit, which is defined as the water content of clay at zero toughness.

<sup>1</sup> Barnes (2013). An apparatus for the determination of the workability and plastic limit of clays. Applied Clay Science. Volumes 80– 81, August 2013, Pages 281-290.

<sup>2</sup> Barnes (2018). Workability of clay mixtures. Applied Clay Science. Volume 153, 1 March 2018, Pages 107-112.



#### 3.6 Experimental setup for sedimentation stests

Glass and plastic containers (450\*260\*450 mm) were used for sedimentation tests as shown in Figure 4. A series of sedimentation tests were conducted to find a workable mixture.



Figure 4 Glass container for sedimentation tests

# 4. Results

#### 4.1 Determination of the capping thickness

The aim of swelling tests was to investigate the salinity effect on thickness of capping layer as well as the turbidity in water after adding the mixture in dry state. When only bentonite was used as capping layer, Figure 5 and Figure 6 shows the tests progress after 1 hour and 168 hours from starting the test, respectively. Table 5 shows the average swelling thickness in mm at various salt content and times.

From Figure 5, after adding the mixture of bentonite and salt, it is seen that that main particles were settled down on the bottom of jar and the finer particles remains in water as suspension at various salt contents. Over time, the fine particles start to settle down to the bottom of jars. The effects of salt on



the turbidity were clearly visible after one day from the test with 1% of salt and it is clearly pronounced when 2% of salt was added. No further effect when 3% and 4% of salt was added.

For the effect on salt on swelling of bentonite layer, it was found that the salt has effect on reducing the swelling of bentonite layer and the thickness of layer decreased as salt content increased to 4% as illustrated in Table 5.



Figure 5 Shows the salinity effect on the turbidity in water due to adding bentonite after1 hour from starting the test.



Figure 6 Figure 6. Shows the salinity effect on the turbidity in water due to adding bentonite after 168 hour from starting the test.



Salt content		(mm)			
(%)	Time (Hours) = 0	24	48	120	168
0	32	33	37	50	55
0,5	32	33	36	45	54
1	33	37	40	50	53
2	37	43	40	40	45
3	38	38	40	40	42
4	37	40	40	40	40

Table 5.Effect of salinity on the swelling of bentonite layer

For the mixture of bentonite and 5% biochar, Figure 7, Figure 8, and Figure 9 show the tests progress after 1, 24 and 168 hours from starting the test, respectively. Table 6 shows the average swelling thickness in mm at various salt content and times.

From Figure 7, after adding the mixture of bentonite, biochar and salt, it can be seen the main mixture particles of bentonite were settled down to the bottom of jar and the remains (finer particles) and some of the biochar stay in suspension at various salt content. Over time, the fine particles start to settle down to the bottom of jars. The effects of salt on the turbidity were clearly visible after one day from the test with 1% of salt and it is clearly pronounced when 4% of salt was added (Figure 8). Theses effect is more pronounced with increasing the time up to 168 hours (Figure 9.

For the effect of salt on swelling of mixture of bentonite and biochar layer, it was found that the salt has effect on reducing the swelling of mixture layer and the layer thickness decreased as salt content increased to 4% as illustrated in Table 6.





Figure 7.Shows the salinity effect on the turbidity in water due to adding mixture of<br/>bentonite and biochar after one hour from starting the test.



Figure 8Shows the salinity effect on the turbidity in water due to adding mixture of<br/>bentonite and biochar after 24 hours from starting the test.





Figure 9 Shows the salinity effect on the turbidity in water due to adding mixture of bentonite and biochar after 168 hours from starting the test.

Salt content (%)	Average layer thickness (mm)					
	Time (Hours) = 0*	48	120	144	168	
0		72	75	75	75	
0,5		72	75	75	74	
1		70	70	70	73	
2		70	70	70	72	
4		70	70	70	70	

Table 6. Effect of salinity on the swelling of bentonite-biochar mixture

\*Difficult to estimate due to turbidity of water.

From, Figure 7, Figure 8, and Figure 9, after pouting the mixture in dry state, it can be seen that the biochar was separated from bentonite and settled with time above the bentonite layer. Therefore, small beakers were used instead of cylindrical jars to measure the swelling effects at various amounts of slat as shown in Figure 10 and Table 7. The purpose of using small backer was to ensure of getting homogenized layer of the mixture of bentonite and biochar by mixing the layer after pouring the materials inside the beaker. Water was added in same amount to each beaker at different interval times. It is found that salt has effect on reducing the swelling of mixture layer and the layer thickness decreased as salt content increased to 4% as illustrated in Table 7..





Figure 10 Shows the salinity effect on the turbidity and swelling due to adding mixture of bentonite and biochar after 168 hours from starting the test by using small beakers.

Table 7.	Effect of salinit	v on the swelling of	f bentonite-biochar	mixture by using	a small beaker.
TUDIC 7.	Lifect of Summe	y on the swenning of		minite by using	g sinian beaker.

Salt content (%)	Average swelling thickness (mm)						
	Time (Hours) = 0*	24	96	120	144		
0		50	60	60	60		
0,5		50	50	55	55		
1		45	45	47	50		
2		40	45	47	47		
3		40	43	45	45		
4		40	40	40	40		

\*Difficult to estimate due to turbidity of water.

#### 4.2 Results – Sedimentation of the capping

#### 4.2.1 Effect of water to solid ratio on the sedimentation

To assess the effect of water content, five batches were conducted by adding diffrent water amounts to investigate the effect of water on the workability and homogeneity of the mixture. The water to solid ratio (W/S) was used to express the relation between the total amount of water in the mixture (water added + moisture content of the materials itself) to the total weight of bentonite, biochar and salt that used in the batch. While the water to bentonite ratio was used to express the relation between the added amount of water to the added amount of bentonite in the batch. Table 8 showed the values between calculated and measured water content, water to the solid ratio and water to bentonite ratio for each batch.



# Table 8.Results of calculated and measured water content of the mixture for different<br/>batches mixed with various amount of water.

	Added water	Water co	Water content (%)		Water /Bentonite
Batch Number	(Mililiter)	Calculated	Measured	Biochar+salt) ratio	ratio
Batch 1	1000	467	462	4.67	5.15
Batch 2	1600	741	747	7.41	8.24
Batch 3	1400	650	642	6.50	7.21
Batch 4	1500	695	692	6.95	7.73
Batch 5	1800	836	834	8.36	9.31

Figure 11 to 15 show how sedimentation is affected adding various amount of water on the homogeneity of mixtures and the turbidity in water due to pouring the mixture.

Figure 16 show a comparison between batch four and batch five (Table 8) after 24 hours from the sedimentation test. Figure 17 shows a comparison between batch two and batch three after 30 minutes from the sedimentation test.

From Figure 11, when 1000 ml of water was added and mixed with bentonite, biochar and salt (batch one). The water to soild ratio was (W/S) 4.67. The mixture was homogenized and its fall and settled down at the base of the glass container as one-pieces as illustrated (Figure 11). The mixture was too thick, cohesive and doesn't have any flowability (Flow =0). The mixture has no turbidity effects on the water when it falling down during the test.

Figure 12 shows the sedimentation test for batch two when 1600 ml of water was added and mixed with bentonite, biochar and salt. The W/S ratio was 7.47. the mixture was homogenized and fall down forming a uniform layer covering the bottom of the aquarium. The mixture has turbidity effects on water directly after pouring the mixture inside the container. The turbidity of water decreased with increased time. The mixture covers the base of the aquarium with a layer with an average thickness of 2-3 cm.

Figure 13 shows the sedimentation test for batch three when 1400 ml of water was added and mixed with bentonite, biochar and salt. The W/S ratio was 6.5. The mixture was homogenized and fall down forming a semi-uniform layer covering the base of the container. The mixture has turbidity effects on the water directly after pouring the mixture inside the container. The turbidity of water decreased with increasing time. The mixture covers the base of the container with a layer with an average thickness of 1-3.5 cm. The turbidity effect is less compared to the mixture of batch two.



Figure 14 shows the sedimentation test for batch four when 1500 ml of water was added and mixed with bentonite, biochar and salt. The W/S ratio was 6.92. The mixture was homogenized and fall down to the bottom of the container forming a uniform layer. The mixture has turbidity effects on the water directly after pouring the mixture inside the container. The turbidity of water decreased with increased time. The mixture covers the base of the container with a layer with an average thickness of 0.5-3 cm. The turbidity effect was less compared to the mixture of batch two and almost the same effect for the mixture of batch three.

Figure 15 shows the sedimentation test for batch five when the added water was increased to 1800 ml and mixed with bentonite, biochar and salt. The W/S ratio was 8.36. The mixture was homogenized and fall down and forming a uniform later covering the base of the container. The mixture has high turbidity effects on the water directly after pouring inside the container and it decreased over a long time. The mixture covers the base of the container with a uniform layer with a thickness of 2.5-3 cm. The turbidity effect was higher compared to all previous mixtures as shown in Figure 16.



Figure 11 Sedimentation test for the mixture of batch one (W/S=4.67) after pouring in the aquarium.





(a) Directly after poruring in the aquirum

(b)After three hours from poruring in the aquirum



(c)After 24 hours from poruring in the aquirum

Figure 12 Sedimentation test for the mixture of batch two (W/S=7.47), (a) directly after pouring the mixture, (b) after three hours from pouring the mixture and (c) after 24 hours from pouring the mixture.





(a) Directly after poruring the mixture in the aquirum



(b) After 24 hours from poruring the mixture in the aquirum

Figure 13 Sedimentation test for the mixture of batch three (W/S=6.5), (a) directly after pouring the mixture and (b) after 24 hours from pouring the mixture.





(a) Directly After poruring in the aquirum

(b)After 15 minutes from poruring in the aquirum



(c) After 24 hours from poruring in the aquirum

Figure 14 Sedimentation test for the mixture of batch four (W/S=6.92), (a) directly after pouring the mixture, (b) after 15 minutes from poring the mixture and (c) after 24 hours from pouring the mixture.





(a) Directly After poruring in the aquirum



(b) After 24 hours from poruring in the aquirum

Figure 15 Sedimentation test for the mixture of batch five (W/S=8.35), (a) directly after pouring the mixture and (b) after 24 hours from pouring the mixture.





(a) Batch four (W/S=6.92)

(b) Batch five (W/S=8.35)

Figure 16 Comparison between batch four and five after 24 hours from pouring the mixture



(a) batch two (W/S=7.47)

(b) batch three (W/S=6.5)

Figure 17 Comparison between batch two and batch three after 30 minutes from pouring the mixture



# 4.2.2 Consistency at the bottom

To investigate the disturbance of the layer on the turbidity of water inside the container after 24 hours from the beginning of the sedimentation test, a steel ruler was used to disturb the surface of the layer by hand and observed the time required to settled down the disturbing particles (particles in suspension due to disturbing effect).

Figure 18 and Figure 19 show the disturbing effect on the mixture of batch two and four hours respectively, after 24 hours from the start of the sedimentation test.

The mixtures of batch two and four were chosen because they occurred within the best range of water /solid ratio that gives less turbidity in water, a more uniform layer and a more workable mixture during the sedimentation test.



(a) Directly after taching the surfce the layer

(b) After 2 minutes from taching the surfce layer

Figure 18 Disturbing the surface layer of batch two (W/S=7.47) by using steel ruler, (a) directly after disturb the layer, (b) After three minutes from disturb the surface in the aquarium







(a) Directly after moving the layer



- Figure 19 Disturbing the surface layer of batch four (W/S=6.92), (a) directly after pouring the mixture, (b) after 15 minutes from poring the mixture.
- 4.2.3 Control of the flowability

The consistency of the slurry was assessed using slump test (Figure 20). A flowability between 25 and 35% was judged as acceptable for the slurry to be pumped based on discussion with the contractor (Figure 21).



(a) Slump test with mini slump cone

(b) Field slump test

Figure 20 Measurement of the flowability and slump height by using flow table test and slump test with mini-slump cone (a) and field test with slump cone





*Figure 21 Effect of water to solid ratio on the flowability of each batch mixture.* 

# 5. Proposed recipe

From the sedimentation tests, it can be observed that the suitable amount of water to solid ratio that can give a homogenized, uniform layer and workable mixture as well as lower turbidity, occurs between the range of batch 2 and 3 and 4. The results of the laboratory investigations is presented in Table 9. The recipe was optimised in order to get a slurry with a consistency that enable pumping with as low water addition as possible i.e. as thick as possible.

	Recipe	Blend	Field batch
	kg	%	kg
Bentonite	100	11,5	6 000
Biochar	22,9	2,6	1371
Salt	1	0,1	60
Water	750	85,8	44 834
Acceptable span for water addition	720-825	85,4-86,9	43 300-49 500

 Table 9
 Target recipe for the blend elaborated and acceptable span for water addition