Session: A.5-Eco-engineering

CAPILLARY BARRIER SYSTEMS FROM CONSTRUCTION WASTES TO COVER RED MUD RESERVOIRS

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Keywords: capillary barrier, capillary layer, capillary block layer, red mud, construction waste

Abstract

Covering and revegetation of red mud disposal sites is a complex engineering task, due to the special conditions, such as the high pH and the high exchangeable sodium content of red mud. The capillary barrier system introduced in this paper consists of two layers, a capillary block layer and a capillary layer made from construction waste such as crushed concrete and brick. The capillary block layer consists of a coarse material that prevents upward capillary transport of the highly alkaline and Na^+ containing liquor from the red mud while the capillary layer comprises fine material that withholds and stores the infiltrating water from precipitation. The function and capacity of the capillary barrier system was tested and monitored in scaled-up experimental series: 1. plastic vessels to test the waste materials (1−1.5 kg); 2. microcosms (20 kg) for modelling the processes at a larger scale; 3. open-air lysimeters (1.5 tonnes) for measuring the technological parameters under field conditions. We characterized the waste materials of the capillary system by the integrated application of physico-chemical analyses and environmental toxicity testing. Chemical analysis showed that the applied materials do not contain mobile metals that might pose additional risk. The environmental toxicity tests proved that the applied materials are non-toxic. The results of the microcosm studies showed that crushed concrete of 30−50 mm particle size can be used as capillary block layer, while both the 0−20 mm particle size crushed concrete and the 0−6 mm particle size crushed brick can be used as capillary layer.

1. Introduction

Red mud is the by-product of the Bayer process in which aluminium is produced from bauxite by digestion at high temperature with concentrated caustic soda (NaOH). The main components of red mud are Fe_2O_3 (41%), Al_2O_3 (17%), SiO_2 (10%), TiO_2 (9%), CaO (9%) and Na_2O (5%). Worldwide bauxite residue disposal areas contain an estimated 2.7 billion tonnes of residue, increasing by approximately 120 million tonnes per annum (Klauber *et al.*, 2011). The existing red mud disposal technologies are wet lagooning, dry stacking, dry cake disposal and marine discharge (Power *et al.*, 2011). Wet lagooning is the simplest way of disposal, but its high risk was tragically demonstrated by the Ajka, Hungary dam failure in October 2010 (Gruiz *et al.*, 2012, Klebercz *et al.*, 2012).

Revegetation of red mud may be problematic due to its high pH (pH>10) and high exchangeable sodium concentration (>70%). In addition, concentrations of plant nutrients such as calcium, magnesium, manganese, and phosphorus in the red mud are low and its fine texture impedes penetration of plant roots. The main approaches include amelioration of the surface layer of the red mud with gypsum, manure, compost, sewage sludge, soil etc., or by capping with soil. After the amendment of the surface layer most commonly the grass species are planted. However, there are no indications that the vegetation would have survived on a longer timescale (Wehr *et al.*, 2006). A Hungarian example from Almásfüzitő showed similar failures: the alkalic solute from the red mud saturated the soil capillary system destroying soil structure and function, leading to soil depletion and plant death (Gruiz, 2007).

Mohan *et al.* (1997) suggested a capping method consisting of a 0.61 m layer of fine clayey dredged material (low permeability layer) covered by a 0.31 m layer of coarser dredged material (vegetative layer) with coastal Bermuda grass, alecia Bermuda grass, alkali sacaton, and salt-bush. Wher *et al.* (2006) applied a cap consisting of 75–150 cm clayey subsoil overlain by 15 cm sandy topsoil with naturally occurring vegetation in a 30-year old and still ongoing revegetation program. However, they experienced problems with the soil becoming sodic and alkaline, and suggested the application of a capillary block layer to prevent the capillary rise from red mud.

Capillary barriers typically consist of two (or more) inclined layers of differently grained cohesionless material. The upper layer, called the capillary layer (or moisture retention layer), is constructed of fine material which can be characterized as a compromise between strong capillarity and high hydraulic conductivity, whereas the lower layer, the capillary block layer (or capillary break layer) is made of coarse material with weak capillarity. The textural contrast between the upper fine layer and the bottom coarse layer controls the vertical infiltration through the barrier by capillary forces. In practice a capillary barrier is covered with a layer of soil acting as a damper and reservoir for the precipitation as well as enabling the revegetation of the cover. Capillary barriers can be used as evapotranspirative covers for flat or nearly flat landfill covers, as lateral drains that limit percolation into the landfill and as oxygen barriers to limit the production of acid mine drainage (Harder and Martin, 2001, Parent and Cabral, 2006).

So far mainly natural materials such as sand and gravel have been used in capillary barrier systems. However, a more sustainable solution is the use of waste material, such as recycled building material or waste rock as constituents. This way the rehabilitation of waste disposals is solved in a socio-economically sustainable way. Harder and Martin (2001) investigated the possibility of using recycled building materials instead of material from natural resources as components of capillary barrier systems. They demonstrated the applicability of the crushed sand-lime brick aggregate (0–2 mm fractions) as capillary layer and of the crushed concrete aggregate (4–16 mm fractions) as capillary block layer. Based on their work, we developed a capillary barrier system from construction wastes, such as crushed brick and concrete, to cover the surface of desiccated red mud reservoirs. As capillary block layer, a coarse material (10–50 mm fractions) was used to prevent upward capillary transport of the highly alkaline and Na^+ containing liquor from the red mud and as capillary layer fine material (0–20 mm fractions) was used to withhold the infiltrating water from precipitation. We investigated the suitability and performance of the construction wastes in the capillary system in microcosms and field lysimeters to predict their possible large scale applicability and to validate the geometry (depth of the layers) and optimal particle size of the wastes, suggested by the microcosm test results.

2. Materials and methods

A scaled-up experimental series was carried out to assess the suitability of waste materials in the capillary barrier systems: 1. testing and measuring the hydraulic properties of various waste materials in simple plastic vessels (1−1.5 kg); 2. microcosms (20 kg) for modelling the processes at a larger scale; 3. lysimeters (1.5 tonnes) for measuring the technological parameters under field conditions. In this paper we are focusing on the laboratory experiments.

The red mud originated from Almásfüzitő, Hungary. The construction wastes applied in the capillary system were crushed brick (0−6 mm and 0−50 mm fractions) and crushed concrete (0−20 and 16−50 mm fractions). As top layer waste soil characterized as kiscelli loam from the construction of the metro line No. 4 in Budapest was used. The loam excavated from 30–40 m depth under the surface is yellow due to the oxidation of pyrite, the lower layers are grey.

2.1. Physical, chemical and environmental toxicological assessment of the waste materials

Prior to the start of the experiments we assessed the physical, chemical and environmental toxicological properties of the waste materials based on an integrated approach (Gruiz *et al.*, 2009). The texture was defined based on the <0.02 mm fraction of the soil/waste particles according to the Hungarian Standard (HS) 08 0205-78. The porosity was determined according to Buzás (1993), the water holding capacity according to Öhlinger (1996) and the capillary head according to Barczi *et al.* (1991). The pH was measured according to (HS 21470-2:1981). The mobile metal content was determined from the distilled water extract (HS 21978-9:1998), the total metal content from aqua regia digest (HS 21470-50:2006) both measured by ICP-AES method using a Jobin-Yvon ULTIMA instrument (HS 21470-50:2006).

We also followed the changes in the quality of the water that gets in contact with the wastes in a batch system on longer time scale. We modelled the scenario when the infiltrating precipitation is ponding on the top of the wastes. The pH and the electrical conductivity of the model precipitation (0.16 mM $CaCl_2$ solution) on the top of the red mud (45 mm height) and in contact with the crushed concrete and brick (1:2 ratio) have been measured for 17 days.

For direct toxicity testing three testorganisms from different trophic levels were used: *Vibrio fischeri* in a luminescence inhibition test, *Sinapis alba* (white mustard) in a root and shoot growth inhibition test and *Folsomia candida* (springtail) in lethality test as described for direct contact with soil/waste by Gruiz *et al.* (2001).

2.2. Model experiments with a capillary block layer

We tested the suitability of the materials in the capillary block layer in small and medium size microcosms. The experiments were carried out in used PET bottles (cut to 17 cm height with 8.5 cm diameter). We measured the water suction of the crushed concrete and brick (10−20, 20−30 and 30−50 mm particle fractions) layer placed on top of the red mud and the red mud plus 5 weight% fly ash (from Visonta, Hungary) mixture layers in the PET bottle. First the capillary motion was measured from the red mud with the original water content (~30%). In the second experiment 17 mm model precipitation (0.16 mM $CaCl_2$ solution; modelling an average large rain event based on meteorological data from Almásfüzitő, 2.5% of one year precipitation, which was 668 mm in 2009) was poured on the top of the red mud (under the capillary block layer). The capillary water flow was followed by measuring the height of the capillary fringe zone in the vessels by a ruler after 1 day.

At larger scale in a 10×30×50 cm plastic vessel 28 cm from the larger fraction (30−50 mm) of the waste materials (concrete and brick) was placed on the top of a 13 cm red mud layer. To reduce evaporation, 6 cm grey waste soil was placed on the coarse concrete/brick fraction. The separation of the different particle size materials was done with a thin layer of wheat straw. On the top of the red mud layer 45 mm (1.2 l) model precipitation was poured (pessimistic scenario modelling an extreme high rain event, 6.7% of one year precipitation) and the upward capillary suction of the water has been followed for 1 week in case of concrete and for 3 weeks in case of brick measuring the height of the capillary fringe zone in the vessels with a ruler. At the end of the experiment the microcosms were taken into pieces and the pH and EC of the interstitial water was measured.

2.3. Model experiments for capillary layer

In used PET bottles (cut to 23 cm height with 8.5 cm diameter) 1.5 kg each of the following materials were placed: crushed concrete (<1 mm fraction), crushed brick (<1 mm fraction), concrete and brick at 1:1 ratio, grey waste soil, yellow waste soil. The microcosms were wetted with 11 mm of model precipitation per day for 4 days (a total of 44 mm, modelling a long lasting large rain event) and the speed of water infiltration was followed measuring the water front by a ruler. After 4 days the maximum water holding capacity of the microcosms was measured by wetting to the point, where the wastes could not hold more water. After irrigation, the slow, spontaneous desiccation was followed for 37 days by gravimetry.

To model the processes at larger scale we placed the capillary layer (crushed brick, <1 mm fraction) and the top layer (grey waste soil) of 25−25 cm and 50−50 cm height into a cylinder (9 cm in diameter) and followed the wetting process (11 mm precipitation per day) by measuring the infiltration fringe by a ruler for 8 days.

3. Results and discussion

We determined the physical, chemical and ecotoxicological characteristics of the wastes, and we tested their applicability in the capillary system.

3.1. Metal content and toxicity

The total metal contents of the waste soils and construction wastes measured in aqua regia extract (Table 1) were under the Hungarian quality criteria (HQC) for soil, except for As in the grey soil (16.9 mg/kg, HQC=15 mg/kg).

The red mud contained As, Cd, Co, Cr, Cu, Ni, Pb in a higher amount than the quality criteria for soil, but the amount was under the permissible metal content for wastewater sludge for agricultural applications. In the 1:10 distilled water extract of the red mud only arsenic (16.5 µg/l), chromium (21.3 µg/l) and nickel (6.70 µg/l) was above detection limit.

We assessed the quality of the precipitation ponding on the top of the wastes (Figure 1). The electrical conductivity (EC) of the water has increased strongly during the first four days, but by the 16^{th} day the increase became less intensive and reached an equilibrium stage. The small particle size fractions of brick (0–6 mm) and concrete (0–20 mm) has similar leaching properties as the red mud, reaching EC 1090–1350 µS by day 16. The large particle size fraction of concrete resulted low EC in the model water (28 µS by day 16), but the water in contact with the coarse fraction of brick (16–50 mm) reached two times the EC as compared with the water in contact with the red mud. This means that the use of coarse crushed brick might cause additional leaching of ions, so the coarse concrete is more suitable in the capillary system than brick of the same particle size.

The pH of the water increased with 0.2 units in case of concrete and with 0.4–0.5 untis in case of brick during 16 days, reaching the maximum of pH 8.1. In case of red mud the pH reached a value of 9.0.

Table 1: Total metal content (in aqua regia extract) of the wastes

Metals in aqua regia extract	HQC for soil[1]	HQC for wwt sludge[2]	Red mud	Grey soil	Yellow soil	Concrete	Brick
	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg
As	15	75	61.3	16.9	15.0	7.90	6.56
Ba	250	-	85.9	161	91.2	130	96.6
Cd	1	10	1.52	0.167	0.175	0.375	0.169
Co	30	50	38.0	15.8	10.4	5.56	5.12
Cr	75	1000	236	54.2	31.8	25.1	23.7
Cu	75	1000	85.8	18.7	20.7	35.3	13.4
Hg	0.5	10	<0.120	<0.120	<0.120	<0.120	<0.120
Mo	7	20	0.760	<0.040	0.505	0.524	0.566
Ni	40	200	130	40.7	30.8	16.1	15.2
Pb	100	750	112	17.7	10.9	41.1	14.9
Se	1	100	<0.600	<0.600	<0.600	<0.600	<0.600
Sn	30	-	13.3	2.47	1.83	15.5	2.46
Zn	200	2500	160	94.5	57.4	105	48.3
pH[3]	-	-	10.0	8.6	9.0	8.6	8.3

[1] HQC = Hungarian Quality Criteria for soil based on KvVM-EüM-FVM Joint Decree No. 6/2009. (IV.14.).
[2] HQC = Hungarian Quality Criteria for sludge from waste water treatment for agricultural applications based on Government Decree No. 50/2001. (IV.3.).
[3] pH in water extract

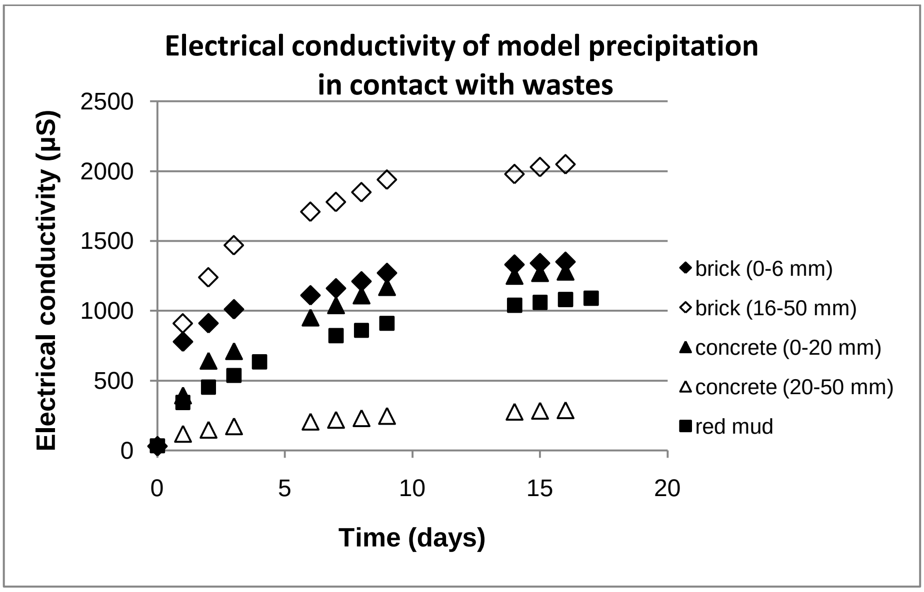

Figure 1: Electrical conductivity of the model precipitation (0.16 mM $CaCl_{2(aq)}$) ponding on the top of the wastes (waste–water ratio: 1:2)

The exotoxicological test results proved that the waste soils and construction wastes are non-toxic on the selected testorganisms (Table 2). The inhibition percentage was under 14% in the *Vibrio fischeri* luminescence test and 20% in the *Folsomia candida* lethality test. The wastes stimulated plant growth as shown by the *Sinapis alba* (white mustard) test results.

Red mud resulted in 92% luminescence inhibition in the *Vibrio fischeri* test. This was not the result of toxicity alone, but also of the light absorption of the small red mud particles. However, 50 time dilution of the red mud resulted in 20% inhibition of luminescence. Red mud caused 41% root and 32% shoot inhibition for *Sinapis alba,* respectively. Abnormal morphology, such as extremely short and curved roots and shoots were observed. The original, wet/alkalic red mud caused 54% lethality for *Folsomia candida*, however, air-dried and re-wetted red mud caused only 39%.

Table 2: Ecotoxicity of the wastes

Waste / Testorganism	Red mud	Grey soil	Yellow soil	Concrete (<1 mm)	Brick (<1 mm)
	Inhibition (%)				
Vibrio fischeri[1]	92%	6%	14%	14%	4%
***Sinapis alba* root**[2]	41%	-20%	-7%	-34%	-44%
***Sinapis alba* shoot**[2]	32%	-14%	-4%	-53%	-73%
Folsomia candida[3]	54%	11%	20%	0%	8%

[1] Inhibition compared to 2% $NaCl_{(aq)}$
[2] Inhibition compared to distilled water.
[3] Inhibition compared to OECD control soil.

3.2. Model experiments for capillary block layer

In small size microcosm experiments (carried out in used PET bottles) we observed low capillary activity by the water suction of the coarse fractions of concrete and brick from red mud and the mixture of red mud and 5 w/w% fly ash (Figure 2). Both concrete and brick can be characterised by low capillarity: 25–35 mm (concrete) and 20–25 mm (brick) from the red mud with the original water content (~30%). The ideal fringe height would be zero in case of the perfect capillary block material, however, 20–35 mm suction can be accepted when the layer thickness is designed properly. The water suction by the crushed concrete and brick is due to the porous structure of the grains themselves.

In case of 17 mm precipitation (average large rain event) 80–90 mm (concrete) and 90–100 mm (brick) fringe height was measured, which means that both materials can suck up 4.7–5.9 times the original height of the water. Therefore in case of a heavy rain event or flood, there is a need for an appropriately designed capillary block layer, which is able to block the upward filtration of water so that it should not reach the capillary layer.

Figure 2: Red mud + concrete (20–30 mm fraction) in PET bottles

Table 3: Water suction from red mud and red mud + 5 w/w% fly ash by large particle size fractions of crushed concrete and brick

Waste material as capillary block	Particle size (mm)	Waste to be covered	Fringe height from original wet red mud after 1 day (mm)	Fringe height from red mud + 17 mm precipitation after 1 day (mm)
Concrete	10–20	red mud	35	80
		red mud + fly ash	30	80
	20–30	red mud	30	85
		red mud + fly ash	25	85
	30–50	red mud	25	85
		red mud + fly ash	30	90
Brick	20–30	red mud	20	90
		red mud + fly ash	20	90
	30–50	red mud	25	95
		red mud + fly ash	20	100

To define the appropriate height of the capillary block layer we assessed the water suction of large particle sized concrete and brick (30–50 mm fractions, 280 mm height) from red mud (130 mm height) at 45 mm model precipitation (modelling an extreme large rain event; pH=5.0, EC=33 μS) After one week the water suction of concrete (with water pH=8.5 and EC=653 μS) was 125 mm (2.8 times the height of the water) (Figure 3), and the water suction of brick was 185 mm (Figure 4). We continued the monitoring of the microcosm with brick. After one week we added an extra 600 ml (22.5 mm) model precipitation. One week later the water suction increased only with 20 mm and remained unchanged for another week. By the end of the third week the water had pH=7.9 and EC=3020 μS.

We can conclude that the 30–50 mm fraction of crushed concrete is recommended for capillary block layer, with the minimum height of 150 mm, as it has low water suction potential. The 30–50 mm fraction of crushed brick could also be used with the minimum height of 250 mm.

Figure 3: Water suction from red mud + 45 mm precipitation by coarse concrete (30–50 mm) after 1 week

Figure 4: Water suction from red mud + 45 mm precipitation by coarse brick (30–50 mm) after 1 week

3.3. Model experiments with the capillary layer

In case of the capillary layer and the top soil cover it is important that the applied materials have active capillarity and optimal water retention. We measured the water holding capacity (WHC) and the capillary head of the materials. The water holding capacity of the small fraction of concrete (0–20 mm) is 46% of the total porosity and 58% in case of brick (0–6 mm) (Table 4). The capillary forces are stronger in case of the brick. However, the WHC decreases with the particle size, meaning that there is an optimal particle size distribution. Also the waste soils have good water holding capacity.

The measured capillary head values indicate that small particle size brick (<1 mm) has stronger potential for the suction of water than the other tested materials. According to Harder and Martin (2001) at least 200 mm capillary head is needed for an ideal material used in the capillary layer, which was fulfilled by all materials (except for the 0–4 mm concrete fraction, where the value was 162 mm).

Table 4: Physical characteristics of the wastes

Waste material / Measured parameter	**Concrete**				**Brick**			**Grey soil**	**Yellow soil**
Particle size (mm)	**<1**	**0–4**	**0–20**	**20–50**	**<1**	**0–6**	**16–50**	**<1**	**<1**
Particles <0.02 mm (%)	-	-	11.5	-	-	11.9	-	36.2	32.8
Total porosity (V%)	-	-	51.8	-	-	54.2	-	54.7	53.6
WHC (%)	31.4	-	24.0	2.6	32.3	31.5	13.4	45.5	36.8
Capillary head[1] (mm)	230	162	-	-	342	230	-	200	260

[1] Measured after 5 hours. -: Not measured

We modelled the wetting and desiccation of the wastes in 1.5 kg microcosms to see the capillary motions of water in the materials. The wastes were wetted for 4 days with 62 ml (11 mm) model precipitation per day and the infiltration of the water was followed. The infiltration was the slowest in case of concrete and yellow soil (Table 5). After 4 days we wetted the microcosms up to their maximum water holding capacity. The result showed that brick can hold the largest amount of water, which is ~50% of the total weight of the microcosms. It indicates that the standard WHC measurement (Table 4) might underestimate the water holding potential of the wastes. There were no differences in the desiccation of the wastes, the water retentions of the different wastes in the microcosms were 88–89% of their water content by day 37.

Table 5: Wetting and desiccation of wastes in microcosms

Process	Time	Unit	Concrete (<1 mm)	Brick (<1 mm)	Concrete: brick = 1:1	Grey soil	Yellow soil
Infiltration (wetting front)[1]	Day 1	mm	40	50	52	40	35
	Day 4	mm	180	195	205	210	185
Water holding[2]	Maximum	ml	685	745	685	625	625
Desiccation[3]	Day 15	%	94.2	93.9	94.2	93.9	93.5
	Day 37	%	88.4	88.5	88.4	89.0	88.0

[1] Wetted with 62 ml (11 mm) model precipitation per day.
[2] Maximum amount of water that can be hold by 1.5 kg waste material.
[3] At room temperature. % of the maximum water.

At larger scale we followed the infiltration (11 mm model precipitation per day) for 8 days into: 1. 25 cm top layer (grey soil) and 25 cm capillary layer (brick) (Figure 5); 2. 50 cm top layer and 50 cm capillary layer. In the first experiment the water front reached the bottom of the top layer by day 7. In the second experiment the water front reached 36.2 cm from the top by day 8. This validated that for the top layer 50 cm should be enough under the modelled weather conditions.

Figure 5: Water infiltration into the top (grey soil) layer (25 cm) after 5 days. Bottom layer: 25 cm brick.

4. Conclusions

In microcosm experiments we tested the applicability of crushed concrete and brick and waste soils in capillary barrier systems to cover red mud reservoirs. The results showed that the applied materials do not contain toxic metals above the quality criteria and they are non-toxic. It was shown that both coarse concrete and brick (30–50 mm fraction) can be used in capillary block layers. We recommend a minimum of 15 cm layer in case of concrete and 25 cm in case of brick, however, thicker layer might be necessary at field conditions. Both crushed concrete (0−20 mm) and crushed brick (0−6 mm) can be used as capillary layers, and the mixture of the two materials is also suitable, having high water retention and capillary activity. As the next step of the experiments we assess the technological parameters under field conditions in 1.5 tonnes lysimeters with built in sensors for continuous monitoring of water fronts and the electrical conductivity of the water.

5. Acknowledgement

The work was carried out as part of the SOILUTIL project (TECH_09-A4-2009-0129) funded by the National Innovation Office, and the scholarship of the TÁMOP 4.2.4.A-1 project financed by the Hungarian State and the European Union, co-financed by the European Social Fund. The chemical analysis was done at the Institute for Soil Science and Agricultural Chemistry, Centre for Agricultural Research, Hungarian Academy of Sciences.

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