LICENTIATE THESIS

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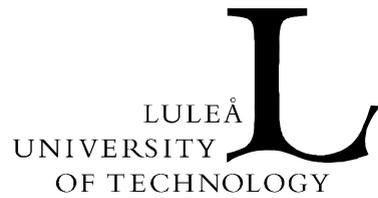
Use of Secondary Construction Material in Landfill Cover Liners

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USE OF SECONDARY CONSTRUCTION MATERIAL IN LANDFILL COVER LINERS

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Paper II   L. Andreas, I. Herrmann, M. Lidström Larsson and A. Lagerkvist (2005), Physical properties of steel slag to be reused in a landfill cover. *Sardinia 2005, Tenth International Waste Management and Landfill Symposium*, Cagliari, Italy.

Paper III   I. Herrmann, M. Svensson, H. Ecke, J. Kumpiene, C. Maurice, L. Andreas, A. Lagerkvist (submitted). Hydraulic conductivity of fly ash - sewage sludge mixtures used in landfill liners. Submitted to *Journal of Hazardous Materials*.

SUMMARY

Recent EU legislation prescribing stricter rules for landfilling is leading to the closure of many landfills in Europe. The utilisation of secondary materials in landfill cover liners decreases the landfilled amounts of these materials while saving natural resources such as clay. The main objective of this thesis is to investigate the applicability of fly ash - sewage sludge and steel slag mixtures in landfill cover liners putting special emphasis on the hydraulic conductivity (HC) of the materials.

The influence of several factors on the HC of the material was investigated using factorial designs. The data were evaluated by means of multiple linear regression (MLR) and multivariate data analysis. The factors investigated in the fly ash – sewage sludge experiment were MATERIAL, ASH RATIO in the mixture, INITIAL WATER CONTENT, COMPACTION ENERGY, FREEZING, DRYING and BIOLOGICAL ACTIVITY. The factors investigated in the steel slag mixtures were the TIME between the addition of water and compaction, the ELECTRIC ARC FURNACE SLAG RATIO in the mixture and the INITIAL WATER CONTENT.

Although mainly affected by the COMPACTION ENERGY used, the HC of the fly ash – sewage sludge samples were also affected by DRYING and the factor interactions MATERIAL×ASH RATIO and ASH RATIO×COMPACTION ENERGY. The models derived using MLR were reliable and could be used to predict an HC between 1.7×10^{-11} m s^{-1} and 8.9×10^{-10} m s^{-1}, if a compaction energy of 2.4 J cm^{-3} was applied, the ash ratio was ≤ 75 % and no drying occurred. However, applying such high compaction energy in the field might cause difficulties.

TIME between the addition of water and compaction mainly affected the HC of the steel slag samples. The HC of the steel slag mixtures was between 7×10^{-8} and 10^{-12} m s^{-1} if they were compacted immediately after water addition. The HC showed a decreasing trend during the measurement period of 53±1 days. It was suspected that cement reactions in the material influenced HC. When steel slags are compacted in landfill liners, the time between the admixing of water and the compaction should be as short as possible.

Low-HC liners are a potential environmental hazard. Monitoring of landfill emissions is prescribed for about 30 years though contaminated landfill leachate is generated for at least one human generation. Because the liner can fail, which might cause a sudden increase of leachate release from the landfill and thus severe environmental pollution, the concept of applying low-HC liners needs to be reconsidered.

SAMMANFATTNING

Nya striktare regler för deponering av avfall enligt EU lagstiftningen leder till att många deponier i Europa kommer att avslutas inom kort. Användningen av sekundära byggmaterial i tätskikt i deponitäckningar minskar deponimängden av dessa material. Samtidigt innebär det en minskad användning av jungfruliga material som t.ex. lera. Syftet med den här avhandlingen är att undersöka om blandningar av flygaska och avloppsslam samt stålslaggsblandningar kan användas som tätskiktsmaterial med hänsyn till materialens hydrauliska konduktivitet (HC (engelska: hydraulic conductivity)).

Ett antal faktorers påverkan på materialens HC undersöktes i två faktorförsök. Försöken utvärderades med multipel linjär regression (MLR) och multivariat dataanalys. Faktorerna som undersöktes i försöket med flygaska-avloppsslam-blandningarna var MATERIAL, KVOT AV INBLANDAD ASKA, UTGÅNGSVATTENHALT, PACKNINGSENERGI, FRYSNING, TORKNING och BIOLOGISK AKTIVITET. Faktorerna som undersöktes i försöket med stålslaggsblandningarna var TID mellan vatteninblandning och kompaktering, KVOT AV INBLANDAD LJUSBÅGSUGNSSLAGG och UTGÅNGSVATTENHALT.

HC för proverna som innehöll flygaska och avloppsslam påverkades mest av PACKNINGSENERGIN, men också TORKNING och faktorinteraktionerna MATERIAL×KVOT AV INBLANDAD ASKA och KVOT AV INBLANDAD ASKA×PACKNINGSENERGI påverkade HC. Modellerna som härleddes med MLR var tillförlitliga och kunde användas för att beräkna HC som låg mellan 1.7×10^{-11} m s^{-1} och 8.9×10^{-10} m s^{-1}, om PACKNINGSENERGIN var 2.4 J cm^{-3}, KVOT AV INBLANDAD ASKA var ≤ 75 % och ingen TORKNING skedde. Det kan dock vara svårt att tillämpa en så stor packningsenergi i fält.

HC för stålslaggsproverna påverkades mest av TIDEN mellan vatteninblandning och kompaktering. HC låg mellan ca 7×10^{-8} och 10^{-12} m s^{-1}, om slaggerna kompakterades direkt efter blandning med vatten. HC visade en tendens att minska under mätperioden som varade 53 ± 1 dagar. HC påverkades troligtvis av cementreaktioner i materialet. Om stålslagg packas i deponitätskikt, bör tiden mellan vatteninblandning och kompaktering vara så kort som möjligt.

Tätskikt med låg HC är en potentiell miljörisk. Övervakningen av deponiemissioner är garanterad i ungefär 30 år, men förorenat lakvatten genereras i minst en mansålder. Om tätskiktet går sönder kan utsläppet av lakvatten plötsligt öka betydligt, vilket kan medföra förorening av miljön. Konceptet med användningen av tätskikt med låg HC borde därför omprövas.

1 INTRODUCTION

Recent EU legislation (EU, 1999) is leading to the closure of many landfills in Europe that have to be covered. A final landfill cover consists of several layers (Travar, Herrmann, *et al.*, in manuscript) and requires several tons of material per m^2. The use of secondary construction materials (SCM) in the cover liner can preserve natural resources and give secondary materials a new field of application, and thus save landfill volume. In landfill cover liners, several secondary materials are conceivable: inorganic wastes such as residues from the steel industry and incineration, and organic wastes such as residues from the paper industry and sludge from water and wastewater treatment. However, SCMs have to fulfil several requirements to be applicable as liner material (Figure 1).
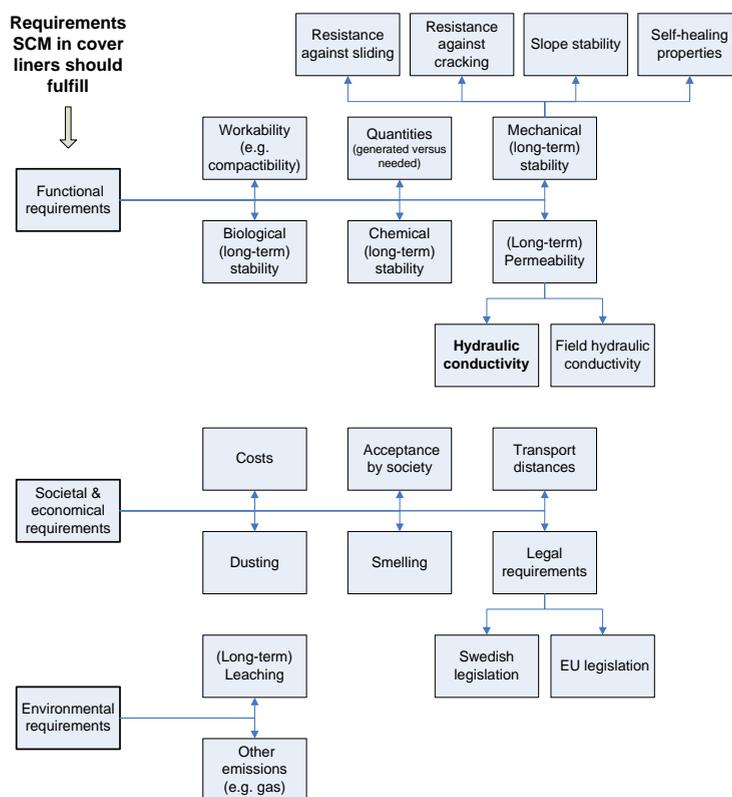
Figure 1   Some issues to consider when assessing the applicability of secondary construction material in landfill cover liners

This thesis emphasises the permeability of SCMs by means of laboratory scale investigations of hydraulic conductivity (HC). As an example of SCMs, steel slag mixtures and fly ash - sewage sludge mixtures are investigated.

The following questions are discussed:

- What factors influence the HC of the investigated SCMs and how?
- How can an acceptably low HC be achieved?
- What is the impact of a low HC on the landfill system and the environment?
- What are other issues regarding the assessment of the material as cover material?

2 MATERIAL AND METHODS

2.1 Material

Two mixtures of SCMs were studied:

1) Digested sewage sludge admixed with fly ash (Herrmann, Svensson*, et al.*, submitted).
2) Ladle slag (LS) admixed with electric arc furnace slag (EAFS) (Andreas, Herrmann*, et al.*, 2005).

2.2 Experiments

To investigate the effect of certain factors on the HC of steel slag and ash - sludge mixtures, two factorial experiments with centre points (Montgomery, 2001) were performed. HC was tested according to the Swedish standard (SIS, 1989) using the constant head method and Darcy's equation for calculating HC.

2.2.1 Fly ash – sewage sludge mixtures

The effect of 7 factors was investigated on the hydraulic conductivity of the fly ash - sewage sludge mixtures, according to a 2^{7-1} fractional factorial design with 8 centre points (Herrmann, Svensson*, et al.*, submitted). The factors investigated were:

- MATERIAL,
- ASH RATIO IN THE MIXTURE,
- INITIAL WATER CONTENT,
- COMPACTION ENERGY,
- FREEZING,
- DRYING and
- BIOLOGICAL ACTIVITY.

Herrmann, Svensson*, et al.* (submitted) describe the performance of the experiment.

2.2.2 Steel slag mixtures

On the steel slag, the effect of 3 factors was studied according to a 2^3 factorial design with 2 centre points (Andreas, Herrmann, *et al.*, 2005). The factors were

- EAFS RATIO in the mixture
- INITIAL WATER CONTENT and
- TIME between water addition and compaction.

The samples were compacted with a hand hammer and a compaction energy of 2.65 J cm^{-3} as suggested in the Swedish compaction standard (SIS, 1994). HC was monitored for a period of 53±1 days. Tap water was used for the tests. HC readings were taken at least two times per week. The measuring period was divided into 5 sub-periods (Periods 1-5), each with a duration of approximately 10 days. In each sub-period, the HC was averaged over n = 5 to 12 readings. The initial water head used was about 1.5 m water column. For samples without permeating water, the water head was gradually increased to a water column of about 6.5 m to be able to take any measurements.

2.3 Statistical evaluation

The data were evaluated using multiple linear regression (MLR) (α = 5 %) and the computer software MODDE (Umetrics, 2003). Multivariate data analysis was performed by partial least squares (PLS) modelling using the software SIMCA (Umetrics, 2004). All response variables were log-transformed before evaluation because they were not distributed normally.

3 RESULTS AND DISCUSSION

3.1 Factors affecting the hydraulic conductivity of fly ash – sewage sludge and steel slag mixtures

3.1.1 Models

To identify the factors influencing HC, reliable models are necessary. The response variables were curved in both fly ash – sewage sludge and steel slag mixtures, i.e. they were not linear. A linear model could not accurately describe the data. Low $R^2_{adj.}$ values characterise a model that fits the data poorly (Montgomery, 2001). To improve model fitting, a quadratic term can be added to the model. In the fly ash – sewage sludge data, the addition of a quadratic term did not greatly improve the model, and $R^2_{adj.}$ were very high in any case (Table 2 in Herrmann, Svensson, *et al.* (submitted)). Hence, the addition of a quadratic term was avoided.

However, including a quadratic term considerably improved the models fitted to the steel slag response variables for Periods 1 to 3, resulting in high $R^2_{adj.}$ values between 0.93 and 0.99 (Table 1). Since the factors that must be squared and added to the model at this stage are unknown, the derived models containing a squared term cannot be used

to predict the HC of steel slag mixtures. Additional observations are necessary to assess which of the investigated factors caused the quadratic behaviour of the response variables. The quadratic effect is very strong (Figure 3) and must be investigated before HC in steel slag can be successfully predicted. In Period 4 and 5 of the steel slag investigation, a quadratic term was not added as it did not improve the model.

3.1.2 Affecting factors: Fly ash – sewage sludge mixtures

The fly ash - sewage sludge mixtures' experiment was conducted in five cycles (Herrmann, Svensson*, et al.*, submitted). The factors affecting the HC differed depending on the cycle. In the last cycle, the material was assumed to have settled well and the results from this cycle were regarded as most relevant. The factors and their interactions influencing the HC of the material in the fifth cycle were

- COMPACTION ENERGY
- MATERIAL × ASH RATIO IN THE MIXTURE
- ASH RATIO IN THE MIXTURE × COMPACTION ENERGY
- DRYING (Herrmann, Svensson*, et al.*, submitted).
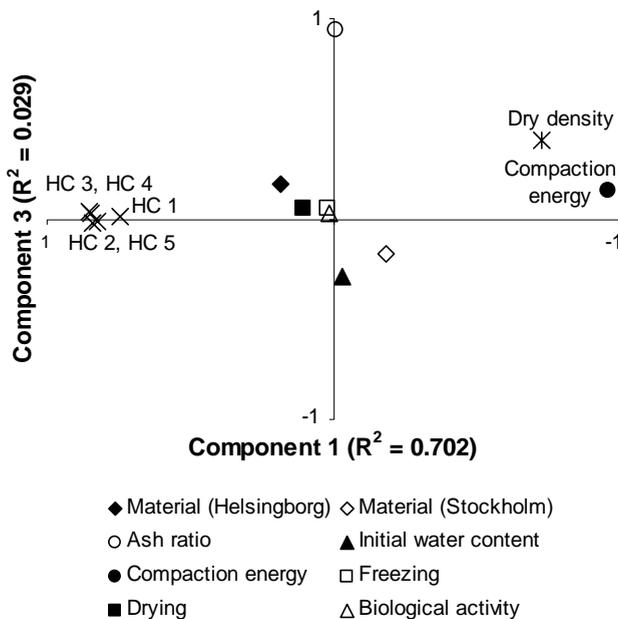
Figure 2   PLS loading plot showing the relations between the investigated factors and responses on fly ash - sewage sludge mixtures

COMPACTION ENERGY clearly affected the HC the most; higher COMPACTION ENERGY led to a lower HC (Figure 2 in Herrmann, Svensson*, et al.* (submitted)). The applied compaction energy is generally known to strongly affect the density of the material. In fly ash - sewage sludge samples, both the dry density and the HC were affected by

COMPACTION ENERGY (Figure 2). Dry density and HC are thus correlated: the higher the dry density, the lower the HC (Figure 2).


3.1.3 Affecting factors: Steel slag mixtures

The factors significantly affecting HC depended on the measuring period (Table 1). Only TIME strongly affected HC in each period as was also shown by a multivariate analysis of the data (Figure 4). The PLS modelling of the steel slag data resulted in only one significant component. However, two components were generated to span a PLS plot (Figure 4). The plot should be interpreted in the direction of Component 1 only since Component 2 is actually insignificant. Because TIME is positioned on the same side of the plot as the response variables, any increase in TIME increases HC. TIME is positioned furthest away from the origin (Figure 4), i.e. it has the strongest effect on HC.

Table 1   Factors and factor interactions affecting HC in steel slag. Positive effect, +; negative effect, -; no significant effect, 0. If a factor has a negative effect, higher levels of the factor decrease the response value and vice versa. $R^2_{adj.}$: adjusted coefficient of multiple determination (Montgomery, 2001).

Factor	Hydraulic Conductivity (HC)				
	Period 1	Period 2	Period 3	Period 4	Period 5
TIME	+	+	+	+	+
WATER CONTENT×TIME	+	0	+	0	0
WATER CONTENT	-	0	-	0	0
EAFS	0	0	+	0	0
Squared term (factor×factor)	+	+	+	0	0
$R^2_{adj.}$	0.97	0.93	0.99	0.79	0.47

The initial water content had a relatively small effect on HC and was observed in Periods 1 and 3 only, i.e. almost at the beginning of the experiment (Figure 3). A similar effect was observed in fly ash – sewage sludge mixtures (Herrmann, Svensson, *et al.*, submitted). During the experiments, a continuous supply of water was forced through the samples. Thus, in terms of water availability, hardening reactions were not restricted. However, water is not continuously available in the field. Hence, the initial water content might be more important and have a more pronounced effect on field HC than on laboratory HC.
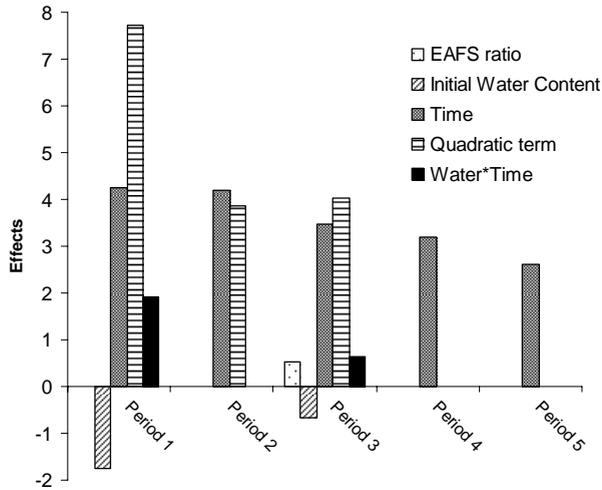
Figure 3   Centred and scaled effects on HC determined using MLR. Positive effect: HC increases with an increase in the respective factor setting. Negative effect: vice versa.

The number of factors contributing to the model shows a decreasing trend from Period 1 to Period 5 (Table 1, Figure 3). Thus, the influences on HC change with time (or with increasing L/S ratio). The model derived for Period 4 was reliable, as indicated by $R^2 = 0.82$ (Table 1), but the model derived for Period 5 was not ($R^2 = 0.5$ (Table 1)). Factors other than those investigated were possibly responsible for the data variation in Period 5. Such factors could be, e.g. changes in water head during the test, preferential flow, chemical reactions or rearrangement of particles. Because some samples were rather impervious and did not show any permeating water, the water head was increased during the experiment to establish a water flow. This might have influenced HC. However, this potential effect can neither be proven nor quantified. Preferential flow (or macroscopic flow) usually causes higher HC. Because the HC of nearly all samples showed a decreasing trend with time (Figure 5), macroscopic flow is not likely to explain the data variation. Steel slag exhibits cementitious properties if it contains C_3S[a] (tricalcium silicate), C_2S[a] (dicalcium silicate), C_4AF[a] (iron aluminium compound) and C_2F[a] (Shi and Qian, 2000). When steel slag is mixed with water, a chemical reaction begins between these compounds and the water (hydration), causing setting and hardening, as described for cement by Murdock and Brook (1979). The major mineral compound of ladle slag is C_2S[*] (Shi, 2002). This compound is mainly responsible for the progressive increase in strength (occurring from 14-28 days and onwards) (Murdock and Brook, 1979). Cement reactions require water and because water was continuously available during the experiment, it can be assumed that the reactions occurring in the material throughout the experiment were responsible for the decrease in HC with time (Figure 5). Regarding the high strength and cohesion of the slag after hydration, a rearrangement of particles seems unlikely to have occurred.

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[a] Cement chemist's notation: C: CaO, S: SiO_2, A: Al_2O_3, F: Fe_2O_3 (Murdock and Brook 1979)

Time ◆ ○ HC Period 2 □ HC Period 3 △ HC Period 4
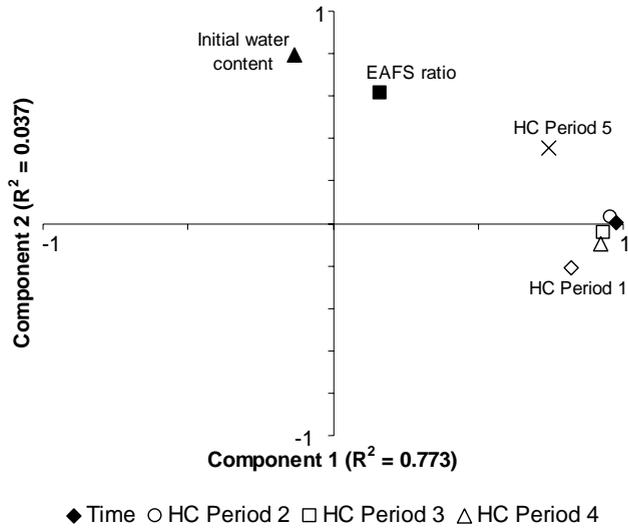
Figure 4 PLS loading plot showing the relations between the investigated factors and responses on steel slag

Dry density of steel slag was observed to decrease with increasing time between the addition of water and compaction (Andreas, Herrmann*, et al.*, 2005). Thus, the factor TIME might reflect the compaction properties of the slag and the compactibility of samples with higher TIME probably deteriorated leading to increased HC.
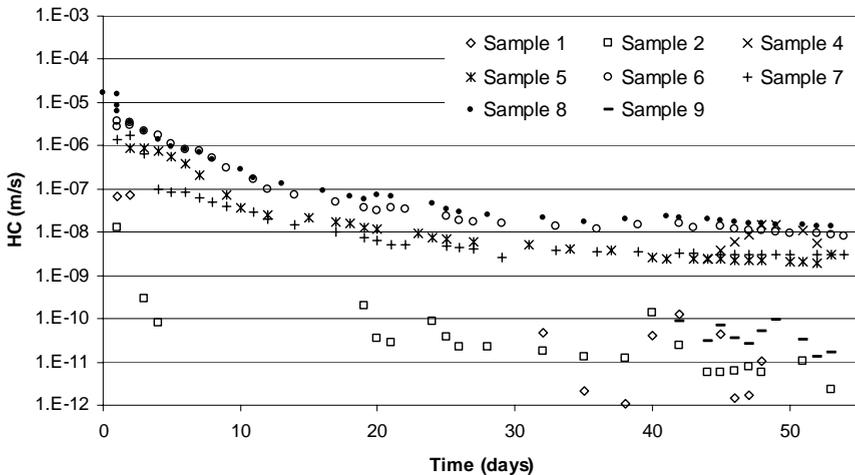
Figure 5 Development of HC of steel slag mixtures over time. Samples 3 and 10 are not shown, since there was no water collected from them. Missing values correspond to measurements where no water had permeated and HC could not be determined.

3.2 How can an acceptably low hydraulic conductivity be achieved?

Swedish legislation prescribes the HC limit values for landfill covers of non-hazardous waste landfills to 50 and of hazardous waste landfills to $5 \, l \, m^{-2} \, yr^{-1}$ (SFS, 2001). These limits are determined for the entire cover construction. For only the liner, a HC value of about $10^{-9} \, m \, s^{-1}$ is usually sought-after for non-hazardous waste landfills. The following two sections outline how this limit value can be met using the investigated material mixtures.

3.2.1 Fly ash – sewage sludge mixtures

For fly ash – sewage sludge mixtures, a reliable model based on the investigated factors was derived (Herrmann, Svensson*, et al.*, submitted) which can be used to predict HC. In one of the MATERIAL mixtures, the lowest HC of $1.7 \times 10^{-11} \, m \, s^{-1}$ was achieved at an ASH RATIO of 20 %, a COMPACTION ENERGY of $2.4 \, J \, cm^{-3}$ and without the occurrence of any DRYING. In the other MATERIAL mixture, the lowest HC of $5.7 \times 10^{-11} \, m \, s^{-1}$ was reached at the same factor settings but an ASH RATIO of 80 %. The HC of both mixtures was between $1.7 \times 10^{-11} \, m \, s^{-1}$ and $8.9 \times 10^{-10} \, m \, s^{-1}$ at an ASH RATIO of up to 75 %. With regards to HC, these materials seem promising for usage in the liner, since they meet the (above discussed) limit value of $10^{-9} \, m \, s^{-1}$ by a comfortable margin. However, the above calculations were based on laboratory data and the results can only be carried over to the field with certain reservations.

As indicated by the model, DRYING of the liner should be avoided to maintain a low HC. The ASH RATIO must be considered, especially if other fly ash – sewage sludge mixtures are tested for usage in a liner construction. Applying a COMPACTION ENERGY of $2.4 \, J \, cm^{-3}$, as required by the above calculation, might be difficult. Mácsik*, et al.* (2006) state that a mixture containing 50 % fly ash has such a low initial strength that it is not possible to drive on the material with bigger machinery such as an excavator. The undrained shear strength of one of the investigated mixtures (tested one day after sample preparation) has been observed as low as 9 kPa at an ash ratio of 25 % and 12.5 kPa at an ash ratio of 57 % (Mácsik, Rogbeck*, et al.*, 2003). The undrained shear strength of consolidated clay formations has been observed to be between 7 and 252 kPa (Cripps and Taylor, 1986), i.e. the shear strength of the investigated mixture is rather small compared to clay. Thus, compacting the material with the appropriate machinery and achieving a sufficient compaction might be difficult. However, Mácsik*, et al.* (2006) compacted fly ash – sludge mixtures with only a scoop and still achieved an acceptably low HC down to $< 14 \, l \, m^{-2} \, yr^{-1}$, corresponding to an HC of $< 4.4 \times 10^{-10} \, m \, s^{-1}$.

When applying fly ash – sewage sludge mixtures in practice, achieving a good blending of the materials can be difficult because of the sticky consistency of the sludge (Mácsik, Maurice*, et al.*, 2006). Mixing fly ash and sewage sludge of one of the investigated mixtures with a screening crusher followed by a compulsory mixer leads to satisfying results regarding shearing strength and permeability of the compacted material (Mácsik, Maurice*, et al.*, 2006).

3.2.2 Steel slag mixtures

As discussed above, the derived models cannot be used to predict the HC of steel slag mixtures. However, recommendations can be given. The TIME between the addition of water and compaction strongly affected HC (Figure 3). Thus, when the material is mixed with water, it should be compacted as soon as possible, since a retardation of 24 hours can lead to unacceptably high HC.

The effect of compaction on HC was not investigated on steel slags and the dry density was not measured. The compaction energy was the same (2.65 J cm^{-3}) for all samples. Decreasing the compactive effort down to 2 J cm^{-3} neither decreases the dry density nor the HC of steel slag (Herrmann, Hamberg, *et al.*, 2006). A further decrease of compaction energy, however, might lead to an increase in HC; hence, a sufficiently high compaction energy should be warranted when steel slags are applied as liner material.

The gradual decrease in HC (Figure 5) is possibly due to the occurrence of cement reactions. It can be concluded that the (desired) cement reactions should not take place before compaction. When the material is stored in a pile outside, however, a crust forms in the outermost layer of the material that prevents water from penetrating into the pile. Thus, outside storage might be possible.

3.3 Impact of a low-hydraulic-conductivity-liner on the landfill system and the environment

Allowing only small amounts of water percolating into a landfill leads to a low leachate generation and hence, to long periods of time until the gradient between landfill and environment is negligible. A landfill needs many centuries to achieve final storage quality (Belevi and Baccini, 1989), i.e. to achieve a state where the leachate produced is fully acceptable in the surrounding environment (Hjelmar and Hansen, 2005). For heavy metals such as Cr and Ni, an L/S ratio of up to 100 is needed to reach certain discharge limits (Lagerkvist, 2003). The time needed to reach such high L/S ratios depends highly on the features of the respective landfill, though it would certainly take more than a human generation (Lagerkvist, 2003).

The advantage of low water infiltration is the small amount of leachate to treat during the after-care phase. In Sweden, after-care (maintenance, monitoring and control) of the landfill is assured for at least 30 years (SFS, 2001). Also beyond the after-care period, the leachate amount is low. Hence, the load of contaminants discharged into the environment per unit time is also low. Furthermore, a liner with low HC can be expected to have low gas permeability that is advantageous for landfill gas collection.

However, the disadvantages of applying low-HC liners are considerable. Leachate is generated over a long time (see above). Beyond the after-care phase, leachate treatment and control of the discharge are not provided. The function of the liner is not monitored and there is a risk of failure through, e.g. cracking, intruding plant roots, earthquakes or failure of the drainage resulting in higher hydraulic gradients so that leachate generation might suddenly increase. In such cases, the leachate would not be treated and directly pollute the environment, groundwater or both with considerable loads of contaminants.

To reliably avoid such incidents, continuous control measurements of leachate amount and quality have to be ensured. Counter measures have to be taken in case the capping or bottom sealing fail. Both control measurements and counter measures are costly. An alternative to landfill capping and infiltration prevention could be enforced and controlled leachate generation with leachate treatment for the time it takes the landfill to reach the final storage quality.

3.4 Other issues regarding the use of fly ash – sewage sludge and steel slag mixtures in landfill covers

One main function of a liner in a landfill cover is to prevent water from seeping through. Thus, when a potential liner material is to be tested, investigating the HC is indispensable. However, measuring HC in the laboratory is insufficient to assess the applicability of a material in a liner. On the contrary, the overall HC of the liner, i.e. the bulk HC (HC_{bulk}), is of interest. HC and HC_{bulk} can differ considerably because HC_{bulk} is dependent on, for example, HC, macroscopic flow (preferential flow) through cracks (Schachtschabel, Blume*, et al.*, 1998), the hydraulic gradient and the degree of saturation of the material. Thus, to assess HC_{bulk}, it seems reasonable to investigate either the resistance to cracking of a material or the performance of the material in the field. The risk for cracking could be assessed by measuring the elasticity, shear strength, tensile strength, shrinkage-swelling behaviour or the crack intensity factor, i.e. the ratio of the surface area of cracks to the total surface area of a soil (Yesiller, Miller*, et al.*, 2000).

The steel slag investigated in this study was observed to be very brittle after compaction. Thus, there is certainly a risk for cracking in steel slag liners, especially when considering that the liner might be exposed to internal stresses due to differential settlements of the subjacent waste. To assess this risk, the elasticity and shear strength of the material and the settlement behaviour of the landfill should be investigated. Field-testing seems to be most suitable in assessing HC_{bulk}. Palmer*, et al.* (2000) confirm the difficulty in reaching the same HC in the field as in the laboratory due to macroscopic flow through the material. Thus, laboratory testing combined with field-testing is necessary to assess HC_{bulk}.

In addition to the HC of the liner, other requirements on the material have to be considered in terms of the liner's function (Figure 1). The material must be workable during the construction process, i.e. mixing with water and compaction must be practicable in the field. The sticky consistency of sewage sludge hampers the applicability of sludge-containing mixtures, and its low strength complicates compaction (Mácsik, Maurice*, et al.*, 2006). Methods for material mixing, admixing of water and compaction need to be developed. Furthermore, the liner has to be mechanically stable for a long time, i.e. it should be resistant against sliding and slope failure. The mechanical stability can be assessed, e.g., by means of the material's shear strength (or friction angle). Biological and chemical degradation and transformation processes might gradually change the stability properties of the material. An investigation of material changes under field conditions seems thus reasonable.

The environmental issues that should be considered when using SCMs in landfill liners are mainly problems relating to dusting and smelling during the construction process, and environmental pollution through leachate or drainage water. In fly ash – sewage sludge mixtures, dusting does not seem to be a problem (Mácsik, Maurice, *et al.*, 2006), but the mixing of steel slag might cause severe dusting. However, smelling does not occur in steel slag whereas the release of ammonia from fly ash – sewage sludge mixtures may cause smelling problems during handling (Mácsik, Maurice, *et al.*, 2006).

Other issues that have to be considered when applying SCM in landfill liners are societal, economical and environmental issues such as legal and quantitative requirements, transport distances and hence costs, and leaching behaviour of the materials (Figure 1). The leaching of fly ash and steel slags is discussed in Travar, *et al.* (in manuscript).

Not only the liner is responsible for the function of a landfill cover. The entire cover construction with its interacting layers has to be considered. A sufficiently thick and well-functioning protective layer, an effective drainage and a stable grading layer will support the overall function of the cover. Fly ash, sewage sludge and steel slag can possibly be also used in other parts of the cover. Since EAFS has a high strength as indicated by an impact value of 18 wt.-% and crushing value of 13 wt.-% (Motz and Geiseler, 2001), it can be used in, e.g. the grading layer. Steel slags can also have grain size distributions similar to well-graded gravelly sand (Travar, Herrmann, *et al.*, in manuscript), making them potentially suitable for use in drainage. The admixture of fly ash, sewage sludge or steel slag to the protective layer is another possible application.

4   CONCLUSIONS

The applicability of fly ash - sewage sludge and steel slag mixtures in landfill cover liners was assessed. Emphasis was put on investigating the hydraulic conductivity (HC) of the materials.

The HC of fly ash sewage sludge mixtures varied between 1.7×10^{-11} m s^{-1} and 8.9×10^{-10} m s^{-1}, if a COMPACTION ENERGY of 2.4 J cm^{-3} was applied, the ASH RATIO was ≤ 75 % and no DRYING occurred. The HC of the steel slag mixtures was between 7×10^{-8} and 10^{-12} m s^{-1}, if they were compacted immediately after the addition of water. Thus, with regards to their HC, the investigated materials seem promising for an application in landfill liners, since they can meet the limit value of 10^{-9} m s^{-1}.

The HC of fly ash - sewage sludge mixtures was mainly influenced by the COMPACTION ENERGY applied. Furthermore, DRYING, the MATERIAL mixture and the ASH RATIO should be considered. Because the COMPACTION ENERGY had by far the strongest effect on HC, the application of high compaction energy should be ensured when the material is compacted on landfills.

The HC of steel slag mixtures was mainly affected by the TIME between water addition and compaction. Steel slag should thus be compacted directly to achieve a sufficiently low HC.

In this study, the materials were investigated on a laboratory scale. The HC detected in the laboratory does not necessarily correspond with the HC in the field. Full scale experiments could give important information on the performance of the material under field conditions.

Low HC of landfill liners leads to a low amount of generated leachate. The generation of contaminated leachate, however, could last for at least a human generation. When the after-care phase of a landfill terminates, i.e. after at least 30 years according to Swedish legislation, no leachate control is assured. If the liner fails by, e.g. cracking, there is a risk for a sudden increase of leachate release from the landfill that might lead to severe environmental pollution. Thus, the application of low-HC liners should be reconsidered.

Other issues that should be considered before applying a secondary material in landfill liners are, e.g. mechanical, biological and chemical stability, compactibility in practice, quantitative and legal requirements, dusting, smelling, leaching and gas emissions.

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6 LIST OF ABBREVIATIONS

EAFS	Electric arc furnace slag
EU	European Union
HC	Hydraulic conductivity
J	Joule [kg m^2 s^{-2}]
LS	Ladle slag
L/S ratio	Liquid to solid ratio
MLR	Multiple linear regression
$R^2_{adj.}$	adjusted coefficient of (multiple) determination
SCM	Secondary construction material
SFS	Svensk författningssamling (Swedish statute collection)
SIS	Swedish Standards Institute

Paper I

USE OF INORGANIC SOLID WASTES IN
LANDFILL COVER LINERS – LITERATURE REVIEW

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Use of Inorganic Solid Wastes in Landfill Cover Liners - Literature Review

Igor Travar, Inga Herrmann, Lale Andreas, Holger Ecke, Anders Lagerkvist

INTRODUCTION

The aim of this literature review is to investigate if inorganic solid wastes are applicable in landfill cover liners. The materials discussed are fly ash (FA) and bottom ash (BA) from municipal solid waste incineration (MSWI) and bio fuel combustion, as well as electric arc furnace slag (EAFS) and ladle slag (LS) from steel production. These wastes are generated in increasing amounts. In Sweden, the annual production is estimated at 447 000 tons of waste incineration ash (Bjurström, 2002) and 1.3 million tons of steel slag (Jernkontoret, Stockholm, Sweden, in 2002, including blast furnace slag). Today, these residues are often landfilled which is costly and leads to the occupation of landfill volume. An alternative to landfilling is to use these materials as secondary construction material (SCM), e.g. in landfill covers. Recent EU legislation (EU, 1999) prescribing stricter rules for landfilling leads to the closure of many landfills that have to be covered in the near future resulting in an increased need for cover materials. The use of SCM saves natural resources and is economically advantageous. The economical benefit achieved by using secondary materials instead of virgin materials is about 100 € per m^2 landfill cover (Tham*, et al.*, 2005).

A landfill cover (Figure 1) serves as a barrier between the landfill and the environment. It prevents the emission of landfill gas into the atmosphere and the infiltration of precipitation into the waste and thus reduces the generation of landfill leachate. The cover construction consists of several layers (Figure 1). The liner is made of impervious material, traditionally using synthetic materials (e.g. geomembranes), natural mineral materials (e.g. clay) or a combination of natural and synthetic materials (bentonite/textile liners). Recently, efforts have been undertaken to use inorganic wastes such as ashes and steel slags as a material for landfill cover liners (Andreas*, et al.*, 2005;Travar*, et al.*, 2005).
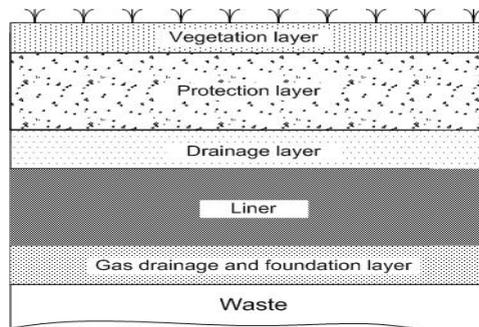
Figure 1   Components in a final landfill cover with a mineral liner

However, there are barriers for the use of SCM in Sweden (Lidelöw, 2004): Unless the environmental impact may be regarded as minor, an environmental permit is required by the supervisory authority. Then, it is necessary to demonstrate that the utilization of SCM is not harmful to humans and the environment, but a general procedure for such environmental assessments is lacking. The permission process for the use of SCM in constructions can be lengthy and costly which decreases the interest in these materials. Furthermore, there is still a lack of experience of handling SCM among users.

The aim of this study is to

- review the state of knowledge in respect of ashes and slags for use in landfill cover liners,
- assess the applicability of the materials in liners,
- and to help to overcome the barriers for using these materials in landfill cover liners.

First, the origin of the materials is explained and a survey of their chemical composition is presented. Then, an overview of the relevant technical properties of the materials for the liner construction is given. The environmental impact is assessed on the basis of leaching data. Finally, legislative aspects are considered.

Subject to this review were in the first instance journal articles, books, standards and legislative texts. In cases where information was not available from these sources, reports, conference articles and theses were resorted to.

MATERIAL

Origin of the investigated inorganic wastes

The combustion of municipal waste and bio fuels (e.g. paper, cardboard, wood) results in the generation of BA and FA. BA accounts for 20 to 30 wt-% of the MSW incinerated and 85 to 95 wt-% of all the waste produced during the combustion process (Chandler, *et al.*, 1997). It comprises material retained on furnace grates (grate ash) and material that passes through the grates (grate siftings). BA is a heterogeneous mixture of slag, ferrous and non-ferrous metals, ceramics, glass and other non-combustibles (Kosson, *et al.*, 1996;Wiles, 1996).

FA is defined as the particulate matter that is carried over from the combustion chamber to the heat recovery units and mechanically separated before any chemical treatment of the flue gas stream (Chandler, *et al.*, 1997;Kosson, *et al.*, 1996). The amount of FA that is produced during the combustion process accounts to about 1 to 3 wt-% of the waste input mass (Sabbas, *et al.*, 2003).

EAFS and LS are by-products of steel production. There are principally two steelmaking technologies: basic oxygen steelmaking (using raw iron as feed) and the electric production of steel in an EAF (using steel scrap as feed). Both processes are usually followed by the second steelmaking step, i.e. ladle metallurgy (Manso, *et al.*, 2005). Since the early 1960s, the adoption of the EAF process has gradually increased,

eroding the market share of the basic oxygen process (Crompton, 2001). Steelmaking using the EAF process has been described by Crompton (2001) and Beskow, *et al.* (2004): In an electric arc furnace, scrap steel and small quantities of raw iron are melted with the help of an arc of electricity produced by electrodes. The molten steel is tapped into a ladle and transferred to the ladle station where it is deslagged (EAFS is drawn off). At the ladle station, the steel is deoxidised and alloys and slag formers are added. Per ton of steel produced, an average of 30 kg LS is generated (Manso, *et al.*, 2005).

Chemical composition

Ashes and slags consist predominately of inorganic matter (Table 1). The most abundant elements in ashes are: Si, Fe, Al, Ca, Na, K with the addition of Cl, Mg, Zn, S and Pb in fly ash. The most abundant elements in steel slag are Ca and Si, with the addition of Fe in some EAFS. The majority of metals in ashes and slag are present as oxides e.g. CaO, SiO_2, Al_2O_3 and MgO (Chandler, *et al.*, 1997;Drissen and Arlt, 2000;Manso, *et al.*, 2005;Shi, 2002). In ashes, metals are also present in form of chlorides, sulphates, and carbonates (Ecke, 2003;Wiles, 1996).

Organic matter in waste incinerator ashes can be as much as 5 wt-% of the total mass (Arvelakis and Frandsen, 2005;Chandler, *et al.*, 1997) and it can be even more in wood ashes. Organic matter in ashes originates from unburned fuel (e.g. cellulose, plant fibre, plastic). Steel slag does not contain any organic substances (EUROSLAG, 2006).

The water soluble fraction of FA is between 21 and 60 wt-% (Ecke, *et al.*, 2002;Kosson, *et al.*, 1996) while the soluble fraction of BA is often less than 1 wt-% (Hjelmar, 1996).

Table 1   Total contents and availability leaching data of ashes, slags and natural rock material, **bold:** availability of elements exceeding the one from natural rock, nd = not detected.

Element	Total Content					Availability			
	Rock[1]	BA[2]	EAFS[3]	FA[2]	LS[6]	Rock[1]	BA[2,7]	EAFS[4,8]	FA[2]
Main elements [g kg⁻¹]									
Si	217 - 346	91 – 308	39.9 - 152.7	95 - 210	125	nd	nd	10	nd
Ca	9.6 - 87	50 – 90	172.3-324.4	50 - 200	407	nd	20-70[2]	100	50 - 100
Mg	3.5 - 47	10 – 30	23.6 - 91.5	10 - 30	19.3	nd	1 - 6[2]	4	4 - 15
Na	0.24 - 29.7	2.9 – 42	0.37[4] / 0.4[5]	15 - 57	nd	nd	nd[2]	0.15	nd
K	2.3-35	7 – 20	0.46[4] / 0.5[5]	20 - 40	nd	nd	1 - 4[2]	0.2	10 - 25
Cl	nd	1 – 3	nd	8 - 60	nd	nd	1 - 3[2], 4.6[7]	nd	8 - 60
F	nd	Nd	nd	nd	30	nd	nd[2]	nd	nd
S	0.0865 - 2	1 – 5	0.6 - 3.31	11 - 45	6.8	0.0241-0.0929	nd[2]	**0.5**	nd
Fe	6.6 - 85	4 – 150	32.7 - 312	12 - 44	11.2	0.163 - 0.894	nd[2]	**3**	nd
Al	69.9 - 85.2	22 – 72	14.1 - 71.6	49 - 90	27.5	0.055 - 0.173	nd[2]	0.03	nd
Mn	0.24 - 1.45	0.08 - 2.4	18.9 - 63.8	0.8 - 1.9	7.75	0.0187-0.0714	nd[2]	nd	nd
Ti		2.6 - 9.5	2.8[4] / 2.8[5]	6.8 - 14	1.8	nd	nd[2]	nd	nd
Other elements [mg kg⁻¹]									
As	0.3 - 104	5 - 40	0.5 - 5.8	30 - 100	nd	0.08 - 3.29	0.3-0.5[2]	<1.5	1 - 2
Ba	368 - 857	500 - 1800	160 - 1800	100 - 3000	nd	4.1 - 52.5	**50-200[2]**	**300**	**30 - 80**
Cd	0.06 - 0.28	2 - 25	0.1 – 19	100 - 900	nd	0.01 - 0.04	**0.5 - 5[2]**	0.07	**100 - 900**
Co	3.7 - 19.2	6 - 350	2.5 – 11	13 - 87	nd	0.1 - 0.6	nd[2]	**0.7**	nd
Cr	20.3 - 331	200 - 1000	320 - 6200	100 - 800	nd	0.1 - 0.33	**2 - 10[2], 9.5[7]**	**7**	**5 - 50**
Cu	11.5 - 62.8	1200 - 2500	62 – 540	300 - 3000	nd	0.16 - 0.76	**50-200[2], 1500[7]**	**1**	**1 - 20**
Hg	0.053-0.198	0.5 - 1	0.1 - 0.1	5 - 20	nd	0.003 - 0.179	0.01-0.1[2]	nd	**4 - 10**
Mo	nd	5 - 30	1.6 – 81	15 - 150	nd	nd	1 - 4[2]	nd	nd
Ni	2.8 - 38.8	10 - 4300	5.2 – 310	60 - 260	nd	0.4 - 2.0	nd[2]	1	nd
Pb	3.3 - 10.7	1500 - 3000	4.5 – 220	4000 - 20000	nd	0.03 - 1.82	**50 - 300[2], 500[7]**	<0.1	**100 – 5000**
Sn	nd	2 - 380	3.2 – 34	550 - 2000	nd	nd	nd[2]	nd	nd
Sr	72 - 760	85 - 1000	178	40 - 640	nd	0.9 - 8.3	nd[2]	nd	nd
Se	nd	0.05 - 10	7.5 – 36	0.4 - 31	nd	nd	nd[2]	nd	nd
Ag	nd	0.3 - 37	1.3 – 100	2.3 - 100	nd	nd	nd[2]	nd	nd
Sb	nd	30 - 200	1.1 – 18	50 - 950	nd	nd	1 - 2[2], 2.5[7]	nd	0.5 - 1
V	17 - 282	nd	170 - 1500	29 - 150	nd	nd	nd[2]	100	nd
Zn	38.5 - 114	2000 - 4000	31 – 690	5000 - 40000	nd	2.4 - 8.0	**50 - 500[2], 2750[7]**	**30**	**4000 – 20000**
Zr	nd	nd	55.2	nd	1480	nd	nd[2]	nd	nd

[1] minima and maxima of n = 9 natural rock materials (Tossavainen and Forssberg, 1999)
[2] Chandler, *et al.*(1997)
[3] minima and maxima of n = 45 samples (Proctor, *et al.*, 2000)
[4] means of duplicates (Fällman and Hartlén, 1994)
[5] Yan, *et al.* (2000)
[6] Shi (2002)
[7] Todorović (2004)
[8] values are approximate

LEGISLATION

Ashes and slags are listed in the European Waste Catalogue (EU, 2000) which means that they are generally referred to as wastes. A material defined as waste usually has to be disposed in accordance with EU legislation (EU, 2002). However, being listed in the European Waste Catalogue does not mean that the materials are considered wastes

under all circumstances (EUROSLAG, 2006). If they are not wastes, their application in geotechnical constructions becomes possible. This application is not regulated but requires an environmental permit according to the Swedish Environmental Protection Law (Thurdin, *et al.*, 2006).

Both EU and Swedish legislation deal with the application of landfill top covers. According to the EU (1999), the application of a surface sealing can be prescribed by the competent authority. If it is installed, it should contain an impermeable mineral layer (EU, 1999). Swedish legislation, however, demands landfill covers. Their permeability for water must not exceed $5\,l\,m^{-2}\,yr^{-1}$ on hazardous waste landfills and $50\,l\,m^{-2}\,yr^{-1}$ on non-hazardous waste landfills (SFS, 2001). As these limit values aim at the entire cover construction, the permeability requirement for only the liner is difficult to define. Liner materials exhibiting a hydraulic conductivity of 10^{-10} m s^{-1} and 10^{-9} m s^{-1} for hazardous and non-hazardous waste landfills, respectively, are expected to meet the Swedish limit values. Other properties of material to be used in cover liners, e.g. regarding stability, are not regulated.

TECHNICAL PROPERTIES OF ASHES AND SLAGS

Relevant technical properties

One prerequisite for reusing SCM is that they meet the same requirements as the materials they shall replace. As described above, nothing but the hydraulical conductivity is regulated with regard to the construction of landfill covers in Sweden. Suggested properties relevant to describe the technical suitability of materials for their use in a cover construction are the material's particle size distribution, hydraulic conductivity, plasticity, compaction, shrinkage and strength characteristics (Kumar and Stewart, 2003;Prashanth, *et al.*, 2001). In this chapter, information on these properties is compiled for the waste materials discussed.
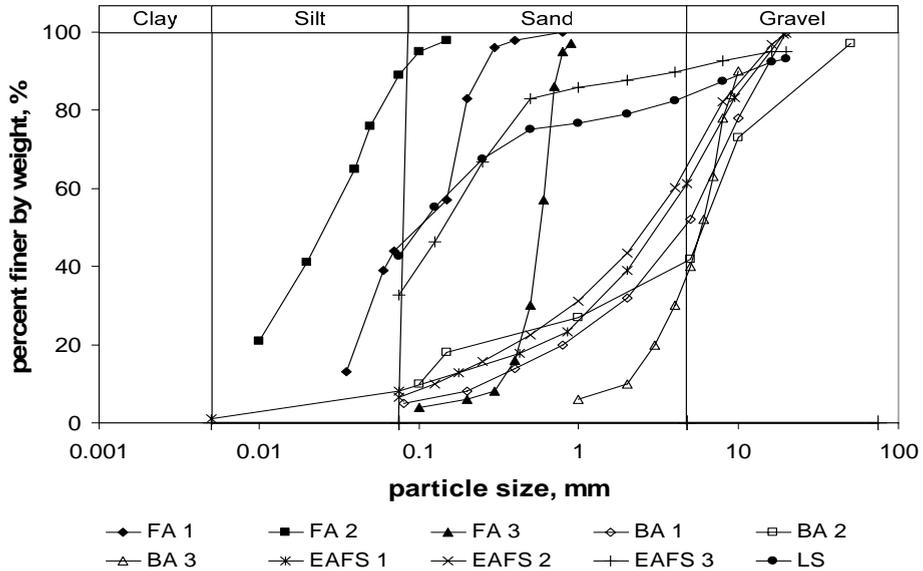
Particle size distribution

The particle size distribution of BA covers the whole range between sand and gravel (Figure 2). The uniformity coefficient (ratio D_{60}/D_{10}) for BA is higher than 6, thus BA can be classified as well-graded gravely sand (Chandler, *et al.*, 1997;Forteza, *et al.*, 2004;Izquierdo, *et al.*, 2002). The percentage of fines (defined as particle size passing through 63 μm sieve) in BA is in the range from 1.9 to 7.4 wt-% (Chandler, *et al.*, 1997).

Particle size distribution curves of FA cover the range between silt and sand size (Figure 2). Factors such as the proportion of fines in the waste, the type of incinerator, changes in operating conditions and the presence of a heat recovery system strongly influence the particle size distribution of FA (Chandler, *et al.*, 1997).

The particle sizes of LS range between 2-200 μm with a prevalence of sizes between 50-60 μm (Manso, *et al.*, 2005), i.e. silt size. EAFS covers the whole range between silt

and gravel with a prevalence of sand particle size (Figure 2). According to the uniformity coefficient, EAFS can be classified as well-graded gravelly sand.

The particle size distribution of ashes and slags can be used to estimate other physical properties such as shear strength or hydraulic conductivity.

Figure 2   Particle size distributions of ashes and slags; FA 1 and BA 1 Chandler, *et al.* (1997), FA 2 Mangialardi, (2003), FA 3 and BA 3 Muhunthan, *et al.*, (2004), BA 2 Izquierdo, *et al.*, (2001), EAFS 1 Proctor, *et al.* (2000), EAFS 2, EAFS 3 and LS Andreas, *et al.*, (2005)

Hydraulic conductivity

A low hydraulic conductivity of the liner is a precondition for a low amount of precipitation infiltrating into the landfill and thus low leachate generation.

The use of BA solely as liner material is not suitable because it is 1-3 orders of magnitude too permeable (Table 2). The same is most probably the case for EAFS having a similar grain size distribution as bottom ash (Figure 2). In order to reduce the hydraulic conductivity of BA, it can be mixed with fine grained material e.g. bentonite or Friedland clay (Kumar and Stewart, 2003;Tham, *et al.*, 2003). It is important to determine the optimal content of the fine material in the mixture with regard to both hydraulic conductivity and geotechnical stability. A relatively low portion of bentonite e.g. 5-15 wt-% mixed with BA or crushed rock can reduce the hydraulic conductivity sufficiently for a utilization as liner material (Keto, 2004;Kumar and Stewart, 2003). Kumar and Stewart (2003) found that the addition of 15 wt-% of bentonite to BA resulted in a hydraulic conductivity close to 10^{-9} m/s. However, adding more than 15 wt-% of bentonite to the mixture may lead to a situation where ballast grains float in the

bentonite matrix having no contact between each other (Pusch, 2001). As a result, the stability of the landfill cover slope may deteriorate.

Higher compaction of ashes is associated with lower hydraulic conductivity. Laboratory tests showed that well compacted FA has a hydraulic conductivity close to that of fine grained till or clay (Table 2). However, it is not always possible to achieve sufficient compaction under field conditions, especially at landfill slopes (Rollings and Rollings, 1996). A stabilization of FA with e.g. lime or bentonite may increase the workability during compaction and hence reduce the hydraulic conductivity of the liner. Mollamahmutogly and Yilmaz (2001) reported that a mixture of FA and 20 wt-% of bentonite is suitable as cover material with regard to its hydraulic conductivity of 10^{-9} m/s. According to Nhan*, et al.* (1996), the hydraulic conductivity of FA can be reduced one order of magnitude by adding lime with CaO > 15 wt-%. Prashanth*, et al.*, (2001) explained this melioration with the formation of gelatinous compounds which block the pores.

A disadvantage with using ash-bentonite mixture in landfill liners is the increase of the hydraulic conductivity over time due to pore water quality. Increase of the pore water salinity due to the release of easy leachable salts from the ash can result in a decreased swelling pressure of the bentonite which leads to an increase of the hydraulic conductivity (Pusch, 2001). Nhan*, et al* (1996) recommended the use of calcium bentonite rather than sodium bentonite in the mixture with ash. Calcium bentonite tends to swell less than e.g. sodium bentonite, thus it is less sensitive to constituents leached from the ash (Nhan*, et al.*, 1996). However, according to Pusch (2001), the hydraulic conductivity is not affected by the pore water chemistry if the liner is compacted sufficiently. In this study, the density of the bentonite at water saturation was higher than 2000 kg/m^3.

Andreas*, et al.* (2005) reported that a mixture of EAFS and LS slag has a hydraulic conductivity of 3.5×10^{-9} m/s. Investigations on the hydraulic conductivity of EAFS and LS slag solely are not reported in literature.

Compaction

Compaction is the simplest and most effective way to improve a material's engineering properties (Rollings and Rollings, 1996). A well compacted material improves its strength and decreases its hydraulic conductivity.

Compaction properties of incineration wastes are not well documented in the literature. BA has similar compaction properties to gravely sand (Table 2) (Chandler*, et al.*, 1997). Field tests showed that it is fully possible to achieve a degree of BA compaction of over 90 % determined in the laboratory related to modified Proctor (Rogbeck and Hartlen, 1996).

Volume stability (Shrinkage and expansion)

The shrinkage of a material can cause cracks and serious damage of the liner. Shrinkage is caused by loss of moisture which results in a volume change of the material. The equilibrium moisture content at which a loss of moisture results in no further volume change is defined as the shrinkage limit. The shrinkage limit of fly ash is higher than clay (Table 2) which indicates that fly ash has a lower shrinkage potential than clay. Low shrinkage potential results in low volume change, thus liners built of a material with lower shrinkage potential have higher resistance to cracks. FA might be a better choice than clay as liner material in dry or cold climates.

In EAFS, expansion might occur (Montgomery and Wang, 1993).The volume stability of LS is unknown.

Plasticity, specific gravity, and shear strength

Plasticity, specific gravity, and shear strength are important properties for the evaluation of slope stability and settlements of the landfill cover.

Settlements occur in all deposits. A common problem with landfills containing degradable material is that the settlements are quite considerable and they tend to occur unevenly, both spatial and over time. Differential settlements of the deposited waste can cause cracks in the landfill liner. The risk for cracks is lower if the liner has plastic properties.

Clay is plastic within a certain water ratio interval (e.g. from 35 to 60 wt-% for illite) (Das, 1998;Lagerkvist, 2003) Therefore, a liner built of clay has a high resistance against cracking as long as it is moist.

Ashes are non-plastic materials (Forteza*, et al.*, 2004). However, a stabilization of fly ash with lime and bottom ash with clay may improve the plastic properties of these materials and hence, enhance their resistance to cracking. Nhan*, et al.* (1996), found that FA mixed with lime is more deformable and less susceptible to cracking than pure FA. In the same study, FA with higher lime content developed higher strength due to pozzolanic reactions.

The shear strength of a material controls the slope stability of a landfill cover. Clay has a very low friction angle (Table 2) which may result in instability of the cover at landfill slopes. Slippage of landfill cover at steep slopes can be avoided by using ashes. The shear strength of EAFS and LS slag is not reported in literature but is expected to be in the range of BA for EAFS and in the range of FA for LS due to the pozzolanic properties of the LS.

Table 2   Technical properties of ashes, slag and clay

		Material			
		Bottom ash	Fly ash	Steel slag	Clay
Specific gravity		$1.8 - 2.2$ [1,7]	1.6 [2]	$3.2 - 3.6$ [10]	$2.5 - 2.9$ [4]
Compaction	max. density, t/m³	$1.5 - 1.8$ [1,5,6,7]	$1.22 - 1.43$ [1]		1.8 [8]
	opt. moist. content, wt-%	$13 - 16$ [1,5,6]	$30 - 40$ [1]		14 [8]
Hydraulic conductivity, m/s		$10^{-6} - 10^{-8}$ [1,7]	$10^{-7} - 10^{-10}$ [1,2]		$< 10^{-9}$ [8]
Shrinkage limit, wt-%			$38 - 52$ [9]		$8.5 - 29$ [8]
Shear strength	cohesion, kPa	7.7 [2]	34.1 [2]		56 [3]
	fiction angle, °	50.2 [2]	29.3 [2]		9.9 [3]

[1] Chandler, *et al.*(1997), [2] Muhunthan, *et al.*(2004), [3] Keto (2004), [4] Rollings and Rollings (1996), [5] Rogbeck and Hartlen (1996), [6] Forteza, *et al.*(2004), [7] Wiles (1996), [8] Das (1998), [9]Prashanth, *et al.*(2001), [10]Montgomery and Wang (1993)

Influence of ageing on technical properties

Ageing is a process, which occurs as a consequence of non equilibrium conditions between the material and the environment. It results in the occurrence of slow mineralogical changes over time which may have an influence on the technical properties of incineration wastes.

Chandler, *et al.*, (1997) reported that BA improves its strength probably due to the formation of new mineral phases that increase the particle interlocking within BA. In the same study, one year aged bottom ash showed much better compaction characteristics than fresh BA. In FA, on the other hand, it has been observed that the process of carbonation can decrease the hydraulic conductivity of the material by up to two orders of magnitude (Meynaud, 2001) which may be due to deteriorated compaction properties.

A siliceous component in FA chemically reacts with calcium hydroxide in the presence of moisture forming a material that exhibits cementitious properties (Palmer, *et al.*, 2000). As a result, FA shows a gain in strength and a decrease in hydraulic conductivity. A FA stabilized with 9 wt-% lime showed a decrease in hydraulic conductivity of three orders of magnitude after 256 days compared to the initial value (Palmer, *et al.*, 2000). Similar reactions can be anticipated when LS is used in the liner.

ENVIRONMENTAL IMPACT OF ASHES AND SLAGS IN LINERS

The environmental impact of liners built with ashes and slags can be of different nature. This chapter deals with the leaching of contaminants that might occur from the surface of the liner when it comes into contact with water.

Leaching conditions in the field

Rain water that percolates through the upper layers of the landfill cover either drains off in the drainage layer or seeps through the liner into the landfill body. The part of the water seeping through the liner leaches constituents from the liner into the landfill. If the hydraulic conductivity of the liner is as low as 10^{-9} m s^{-1}, this amount of water does not exceed 32 mm yr^{-1}. As in Sweden, the annual precipitation amounts to between 400 and 1100 mm (SMHI, 2005), only 3-8 % at most of the precipitation seeps through the liner and this path will not be discussed in this study. The drainage water moves through the drainage layer on top of the liner into a ditch and is usually discharged into a local recipient. Assuming that 50 % of the precipitation turns into drainage run-off, this water can amount to up to 550 mm yr^{-1}. As the drainage layer is located directly above the liner, there is a potential for release of contaminants from the liner surface into the drainage water. This release is influenced by e.g. particle size and leaching conditions such as pH, solvent quality, redox conditions, temperature and liquid to solid ratio (L/S) (Todorović and Ecke, 2006).

Compacting ashes in a liner leads to a development of a monolithic structure of the material (Tham*, et al.*, 2005). The same is probably the case for slags. Therefore, leaching from the liner surface is assumed to be diffusion controlled (Kosson*, et al.*, 1996). This means that the particle size does not affect leaching and surface accessibility is generally low. Leaching is to a great extend influenced by pH. The quality of the drainage water, especially in terms of pH, is highly influenced by the layers it has been percolating through. pH conditions on the liner surface are therefore difficult to predict as they depend largely on the material used and may change over time. However, pH values between 6 and 8 seem realistic to assume. Redox conditions are unknown but might be anaerobic. The temperature on the liner surface depends e.g. on climate, season and thickness of the protective layer. In the soil cover of a Northern Swedish landfill test cell, temperatures between 2 and 16°C were measured in 1.2 m depth (Maurice and Lagerkvist, 1997). The L/S (liquid to solid) ratio increases continually with precipitation.

Critical elements

To identify the environmentally harmful elements (critical elements) that could be released from ashes and slags, their mobility (or concentration) would usually be compared to limit values. As there are no limit values available from legislation, they are compared to natural material using data from leaching tests (Table 1). As the leaching from the liner surface is diffusion controlled (Kosson*, et al.*, 1996), data from diffusion leaching tests would be appropriate for the comparison. However, diffusion test data are neither available for natural material nor for slags. Another reasonable

leaching procedure is the availability leaching test which is designed to provide an estimate of the maximum mass of material that could be leached under aggressive leaching conditions (Kosson*, et al.*, 1996). In this test, the material is leached at L/S 100 in two steps: first at pH 7 for three hours and then at pH 4 for 18 hours (Nordtest, 1995). The sample is furthermore milled to a small particle size (95 % < 125 µm) (Nordtest, 1995). The availability test shows the potentially leachable amount of a constituent but does not provide results whether or not this maximum release will ever be fulfilled or the time intervals during which this release may occur (Todorović and Ecke, 2004). The availability test is a solubility controlled test and does not reflect the leaching conditions in the field, but it represents a "worst case" scenario. Data from availability testing are available for both natural material and ashes and slags (Table 1).

EAFS and BA are reused as substitutes for natural rock. Thus their leaching is compared to the leaching of rock (Table 1). FA and LS are used as substitutes for clay in the liner and should thus be compared to clay. However, availability leaching data from clay and LS are not reported in literature. The leaching from FA is therefore also compared to the leaching from natural rock (Table 1).

The results of the comparison are shown in Table 3. Most striking is the mobility of **Pb** (up to 150 000 times higher in FA compared to rock) and **Cd** (up to 90 000 times higher in FA) and **Cu** (up to 9000 times higher in BA). In BA, FA as well as EAFS, the mobility of **Cr** (up to 500 times higher in FA) and **Zn** (up to 8000 times higher in FA) is remarkable. Also the mobility of **Hg** from FA exceeds the one from rock. Furthermore, in FA and BA, **Cl**, **SO₄** and **Mo** are regarded as major pollutants (Chandler*, et al.*, 1997;Comans*, et al.*, 2000;Meima and Comans, 1999). In BA used in road construction, also Al and Sb have been identified as critical (Todorović, 2006). In FA, the initial mobility of salt formers, such as **K**, **Na**, **Mg** and **Ca** (Chandler*, et al.*, 1997;Sabbas*, et al.*, 2003) might be problematic. In EAFS, the mobility of **V** might cause environmental pollution.

Table 3   Critical elements in BA, FA and EAFS and LS. In BA, FA and EAFS, the critical elements were identified by comparing data from availability leaching to natural rock material. Some constituents (marked in the table) were described as critical in literature but were not critical according to the comparison above. Critical elements in LS were identified by comparison with EAFS.

BA	FA	EAFS	LS
Cd^0	Cd^0	Cd^0	Ti^0
$Cu^{0,3}$	Cu^0	Cu^0	Zr^0
$Cr^{0,3}$	Cr^0	Cr^0	
Zn^0	Zn^0	Zn^0	
Ba^0	Ba^0	Ba^0	
Pb^0	Pb^0	Co^0	
$Cl^{1,3}$, $SO_4^{1,3}$, $Mo^{1,3}$	Cl^{-1}, SO_4^1, Mo^1	V^0	
Al^3	Hg^0		
Sb^3	K^2, Na^2, Mg^2, Ca^2		

[0] critical according to above described comparison, [1] Chandler*, et al.* (1997), Comans*, et al.* (2000), Meima and Comans (1999), [2] Chandler*, et al.* (1997), Sabbas*, et al.*, (2003), [3] Todorović (2006)

Leaching of the critical elements

The leaching of the critical elements from ash and slag as measured in the availability test (solubility controlled leaching) certainly overestimates the availability of these constituents under field conditions (diffusion controlled leaching). When leaching is diffusion controlled, the mobilization of contaminants can be expected to be low. Nevertheless, leaching of the identified critical elements shall be discussed in the following.

Ecke and Åberg (2006) investigated the influence of the factors pH, L/S, leaching atmosphere and time on the leaching of Cd, Cr, Cu, Ni, Pb, and Zn from BA. They concluded that the factors pH and L/S are the most significant factors affecting leaching.

In BA and FA, the minimum release of **Pb** is observed at pH between 8 and 10, while lowest concentrations of **Zn** are at about pH 10 (Hjelmar, *et al.*, 2001;Wiles, 1996). The leachability of Pb is primarily caused by chloride complexation. At pH 6-9, the release of Pb is lowered due to the formation of less soluble carbonates (Ecke, *et al.*, 2002) which suggests that Pb leaching from the liner surface is marginal. For steel slag, Pb and Zn leaching has not been investigated further. The amounts of Pb and Zn leached from ashes and slag under landfill conditions must be studied before it is possible to assess the environmental impact of the materials with regard to these metals.

At alkaline pH, leaching of **Cd** from ash is influenced by chlorides. The release of Cd from ash is increased with decreasing chloride content and pH at pH less than 7 (Hjelmar, *et al.*, 2001;Wiles, 1996). Further decreasing of pH in the long run may increase the leachability of Cd dramatically (Hjelmar, *et al.*, 2001). These data suggest that Cd might be available to a quite high extend on the liner surface. It was observed that Cd concentrations are higher in combined ash leachate than in bottom ash or fly ash leachate (Hjelmar, 1996). It is probably caused by the high content of chlorides and the presence of organic acids produced by the biodegradation of unburned matter in bottom ash. Thus, a liner comprising both materials is more susceptible to Cd leaching.

The leachability of **Cr** is of special interest because of its toxicity in the hexavalent form (Pillay, *et al.*, 2003). The availability of Cr increases under oxidizing conditions (Fällman and Hartlén, 1994). As oxygen availability cannot be ruled out, this means that Cr leaching could occur on the surface of the liner and contribute to the pollution of the drainage water. Leaching of Cr is different for fly ash and bottom ash at pH > 5 probably due to the presence of Cr as Cr(III) in BA and Cr(VI) in FA (Hjelmar, *et al.*, 2001). Cr (VI) is highly soluble over the complete pH range (Cohen and Patrie, 1997) whereas Cr(III) in BA has the lowest release at neutral and slightly alkaline pH (Hjelmar, *et al.*, 2001). In EAFS, Cr is present in both oxidation states (Proctor, *et al.*, 2000). Cr(III) in EAFS can, in the presence of calcium oxide, oxidize to hexavalent Cr (Pillay, *et al.*, 2003). Pillay, *et al.* (2003), reported that 0.1 -1% of the Cr in EAFS in ambient conditions (e.g. on a deposit) oxidizes to Cr(VI) within 6-9 month when exposed to oxygen. However, the reaction ceases after 12 months. Assuming the oxidation of Cr(III) to Cr(VI) to occur not only in EAFS but also in BA, leaching of toxic Cr(VI) can be expected from all three materials. To address this problem, the Cr contaminated drainage water should be treated at least in the first year.

Leaching of fresh ash results in high initial concentrations of **Cu** (Chandler, *et al.*, 1997). Dissolved Cu in ash leachate is bound to organic matter to a significant extent (Meima and Comans, 1999). This is confirmed by Olsson (2005) who interpreted a decrease in Cu^{2+} activity in BA leachate to be caused by Cu-DOC complex formation. It was observed that the release of Cu from bottom ash may be reduced by reducing the organic matter (Hjelmar, *et al.*, 2001) or by forming ternary complexes of Cu with DOC (dissolved organic carbon) bound to the precipitate (Meima, *et al.*, 2002). Another option for lower release of Cu from ash may be treatment with Fe (III) and Al (III) acidic salt solutions which leads to a pH decrease from about 10-11 to about pH = 7-8 and in-situ precipitation of the metal(hydr)oxides (Comans, *et al.*, 2000). The main effect of this treatment is the binding of Cu to the ash matrix.

The mobility of **Hg** in FA can be up to 3300 times higher than in natural rock (Table 1). The leachability of Hg from FA is lowest at pH 8 to 10 with a strong increase below pH 8 (Chandler, *et al.*, 1997) but only limited data regarding Hg leaching from FA are available (Chandler, *et al.*, 1997). In coal fly ash, however, Hg leaching has been investigated. Hg in coal fly ash is tightly bound and not released in significant amounts to water under mildly acidic leaching conditions (Gustin and Ladwig, 2004). The data above suggest that Hg leaching from FA is a minor problem in terms of environmental pollution. However, as the difference in Hg mobility between FA and rock is pronounced, further investigations should be undertaken.

The leachability of **Mo** in FA and BA appears to be constant between pH 7 and 13 and decreases towards pH 4 (Hjelmar, *et al.*, 2001). Leaching tests showed that Mo is almost completely washed out within L/S = 2 l/kg which indicates that Mo is an example of availability controlled leaching (Hjelmar, *et al.*, 2001). Meima, *et al.* (2002) have shown that Mo leaching from BA also remains largely unaffected by carbonation. Mo can thus be expected to be quickly released from the materials.

The release of **V** from BA in the pH dependence test showed that the minimum is at pH 6 increasing towards pH 12 (Hjelmar, *et al.*, 2001). In EAFS, V is a critical element. As it is leached under neutral conditions from ash, it might also be mobile from EAFS under these conditions and thereby be of potential harm for the environment.

Initial leachate from ashes shows relatively high content of (earth) alkali metals such as **K, Na, Ca** that are present in a form of salts (Hjelmar, 1996). Leaching of most alkali metals from ashes is independent of the pH and they are released rapidly as a function of L/S (Kosson, *et al.*, 1996; Sabbas, *et al.*, 2003; Wiles, 1996). Concentrations of most alkali metals in the leachate have a tendency to decrease as the leaching progresses. Kosson, *et al.* (1996) reported that about 60 % and 90 % of the salt (e.g. KCl, NaCl, $CaCl_2$) availability was released by LS of 1 l/kg for bottom ash and fly ash, respectively.

The leaching properties of LS have not been investigated. However, total contents can be compared to those of natural rock. The total content of Mn exceeds natural rock. Ti and Zr contents in rock are unknown, but compared to EAFS, both Ti and Zr in LS are elevated. Zr has been shown to be of low toxicity (Couture, *et al.*, 1989; Fodor, *et al.*, 2005). However, as Zr concentrations in LS can be 1480 mg kg^{-1} (Shi, 2002) and the

lethal concentration is LC 50 > 20 mg L^{-1} (Couture*, et al.*, 1989), the environmental harmlessness of Zr in steel slags still has to be proved.

Influence of ageing on leaching behaviour

Ageing has a significant influence on the element leaching from ashes (Meima and Comans, 1999). The most important ageing processes in ashes are carbonation and formation of minerals that can bind contaminants (Comans*, et al.*, 2000;Wiles, 1996). Carbonation is caused by the uptake of carbon dioxide (CO_2) by the material which results in a decrease of the pH from alkaline towards neutral (Meima*, et al.*, 2002). Possible CO_2 sources for the carbonation of material are atmosphere (before use in construction) and landfill gas.

The leaching of trace elements such as Cd, Pb, Zn, Cu and Mo from bottom ash is significantly lower for carbonated samples than for fresh samples (Meima and Comans, 1999). Ecke*, et al* .(2003), found that carbonation of fly ash reduced the availability of Pb and Zn by two orders of magnitude.

The lower release of contaminants from aged ash is related to the decrease of pH values of ash leachate (Meima and Comans, 1999). Another effect that is assumed to control leaching from aged ash is the sorption of contaminants onto neoformed minerals. Elements such as Cd, Pb, Zn, Cu and Mo in BA have high affinity for Fe/Al-hydroxides and amorphous aluminosilicates that can be formed as a result of ageing (Meima and Comans, 1999). In aged fly ash, the formation of calcium aluminosilicate hydrates (C-A-S-H) has the potential to retain metals through both bonding and absorption (Ecke*, et al.*, 2002).

Zevenbergen*, et al.*(1998) investigated the influence of ageing on glassy constituents of bottom ash. In this study, a 12 year old bottom ash showed a rapid formation of well-ordered primitive clay (illite) from glasses. Clay mineral formation is also observed in 8 year old fly ash samples (Zevenbergen*, et al.*, 1999). It is found that heavy metals like Zn and Cu were incorporated into clay minerals, thus the latter may play an important role in the immobilization of heavy metals and other toxic species (Zevenbergen*, et al.*, 1999;Zevenbergen*, et al.*, 1998).

CONCLUSIONS

Ashes can be beneficially used in landfill cover liners with regard to their technical properties investigated on laboratory scale. However, bottom ash (BA) cannot be applied solely in the liner but mixed with bentonite, it reaches hydraulic conductivities down to about 10^{-9} m s^{-1}. Well compacted fly ash (FA) can be used solely in the liner with regard to its hydraulic conductivity. The use of ashes in the liner has an advantage compared to clay regarding their lower shrinkage potential and also their higher friction angle which results in a good slope stability. Further research on technical properties of ashes should be performed in full scale paying special attention on hydraulic conductivity and resistance to cracks due to differential settlements.

There is a lack of information on technical properties of electric arc furnace slag (EAFS) and ladle slag (LS) in the literature. The technical performance of slags in liner constructions can therefore not be assessed for neither EAFS nor LS. However, in terms of grain size distributions, EAFS and LS are similar to BA and FA, respectively. Hence, it can be assumed that EAFS cannot be used solely in the liner. Mixtures of EAFS and LS seem suitable liner materials as they reach a hydraulic conductivity of 3.5×10^{-9} m s^{-1}. This, however, has to be confirmed by additional laboratory and field investigations because it is not certain that other technical properties of EAFS and LS are similar with ashes.

When utilizing ashes and slags in landfill cover liners, there is a risk for Pb, Cd, Cu, Cr, Zn, Ti, Zr and Hg to be leached from the liner surface by the drainage water and released into the environment. Furthermore, Cl, SO_4, Mo, salt formers (K, Na, Mg, Ca) and V might be problematic. The leaching of trace metals from BA and FA has been investigated quite thoroughly whereas leaching data for EAFS are rare. LS have not been investigated at all in terms of leaching. From the data compiled in this study, a significant release of Cd, Cr, Mo, salts and possibly Cu is to be expected from the landfill liner containing ashes or slags. A liner comprising both BA and FA is more susceptible to Cd leaching than a liner built of FA only. The leaching of some contaminants are short-term processes, which applies to Mo and salts and possibly also to Cr. The problem of their release could be addressed by a temporary treatment of the drainage water.

Ageing of ashes reduces the release of contaminants (salts and heavy metals) into the environment. It is expected that rapid formation of primitive clay minerals in aged ashes will reduce the release of heavy metals but also improve technical properties such as hydraulic conductivity. The influence of ageing on leaching of EAFS and LS has not been investigated.

The leaching of critical elements from ashes and slag under landfill conditions should be subject to further research as it is difficult to assess the environmental impact of the materials used in liners on the basis of the available data. The influence of factors such as pH, redox conditions, temperature, L/S, time and buffer capacity has been investigated only rarely. Data from leaching of constituents depending on pH are available for BA and FA but not for EAFS and LS. More thorough investigations on all four materials are needed especially for toxic trace metals like Pb, Cd, Cu, Cr, Zn and Hg. Leaching should also be investigated in full scale.

Other metallurgical by-products that were not reviewed are e.g. blast furnace slag or basic oxygen furnace slag. Also these materials might be interesting to investigate towards reuse in a landfill liner.

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Paper II

PHYSICAL PROPERTIES OF STEEL SLAG TO BE REUSED IN A LANDFILL COVER

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PHYSICAL PROPERTIES OF STEEL SLAG TO BE REUSED IN A LANDFILL COVER

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SUMMARY: Many landfills in Sweden as well as in Europe have to be closed in the near future. Apart from material costs in the order of tens of billions Euro, this puts a strain on the environment through the exploitation of virgin materials. Many landfill operators are considering alternative cover constructions in order to reduce resource spending. In this study, the authors are looking at a construction including slags from a steel mill. Four electric arc furnace (EAF) slags and one ladle slag from Uddeholm Tooling AB at Hagfors, about 270 km Northwest of Stockholm, were investigated with regard to their physical properties. A full scale field test on an area of about 5,000 m^2 will be started at the local municipal landfill in Hagfors in August 2005.

1. INTRODUCTION

Many landfills in Europe do not meet the requirements stipulated in recent EU legislation and thus have to be closed and covered in the near future. Roughly 2,000 hectares of landfill area in Sweden have to be covered, equivalent to almost one hundred million tonnes of construction material. In order to minimize the exploitation of virgin materials, secondary materials are often considered to be a sensible alternative. Steel industry slags could be a conceivable possibility if they are cheap and easily available. However, no studies are known looking at the behaviour of steel slags in this special area, e.g. what kind of problems may occur due to the different properties of the slags compared to conventional construction material, if the slags will meet the requirements for landfill cover materials, and how they will perform in the long run.

In co-operation with Uddeholm Tooling AB and Hagfors municipality, the Division of Waste Science & Technology and MiMeR (Mineral and Metals Recycling Research Centre) at Luleå University of Technology, Sweden, perform a project investigating the potential use of steel industry slags in landfill cover constructions since autumn 2004. Parts of the municipal landfill in Hagfors – a landfill for non hazardous waste – are to be covered and the slags are a potential constituent of the cover construction. Electric arc furnace (EAF) slags and ladle slag from Uddeholm Tooling AB, Hagfors, Sweden, are investigated.
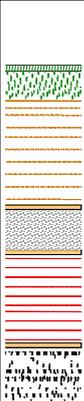
		Possible materials	Water balance
	• ~ 0,3 m **Vegetation layer**: plant colonization, water magazine, protection against erosion	Soil, compost	100 % precipitation
	• ≥ 1.5 m **Protection layer**: water magazine, protection against desiccation, freezing, root penetration, erosion of the layers below	Soil, digested sewage sludge mixed with the finer fraction of slag	70 % evaporation and root uptake
	• *Geomembrane (~ 0,5-5 mm):* layer separation • ~ 0.4 m Drainage layer: collection and discharge of percolation water, prevent water standing on liner • *Geomembrane*		
	• ≥ 1.0 m **Barrier layer, liner**: minimize gas and water transport	Coarse slag (EAF), crushed construction debris	25 % drainage water
	• *Geomembrane*		
	• ≥ 0.5 m **Gas drainage, foundation layer**: surface adjustment, load distribution, gas transport	Ladle slag mixed with EAF slag	
		Coarse slag (EAF)	5 % leachate

Figure 1. Layers and their function in a cover construction with a mineral liner; examples for possible alternative materials and water balance for the different layers.

2. CONSTRUCTION OF A LANDFILL COVER SYSTEM

A final cover construction is a system of components that all contribute to achieve the desired function. The legal requirement in Sweden is directed towards the maximum amount of leachate generated: < 5 and < 50 l $(m^2*a)^{-1}$ for landfill class 1 and 2, respectively (SFS 2001:512). The functions of the single layers within the cover are described in Figure 1.
The figure also gives examples of possible alternative construction materials and of how the different layers influence the water balance.
The field test at the Hagfors landfill will include a number of alternative cover constructions with varying design of the barrier layer. The construction activities will start in August 2005 and be finished during 2006. A similar project investigating an alternative cover construction including ashes from the incineration of refuse derived fuel (RDF) and biofuel, is performed at the Tveta landfill outside Stockholm (Tham et al., 2003 & 2005; Travar et al., 2005).

3. METHODS AND MATERIALS

The evaluation of the function and long-term durability of the cover construction has to be made in both lab and full scale. Swedish legislation prescribes only elementary analysis and short-time leaching tests for material characterization (NFS 2004:10). Simple leaching tests can indicate if the material contains substances that leach under the chemical conditions of the used test. They are not pH- or redox controlled and hence do not reveal information about the interaction of the tested materials with other materials. The short time of the test prevents the observation of processes governed by biological reactions in the sample. Though some of the tests are performed at high L/S[1] ratios the significance for the long-term behaviour is reduced to the leaching of e.g. salts and other components that do not change their behaviour over time and/or depending on the environment (Andreas, 2000).

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[1] L/S ratio – liquid to solid ratio

Table 1. Annually output of the different slags from the Uddeholm Tooling steel mill

slag	EAF 1	EAF 2	EAF 3	EAF 4	LS
amount [ton/a]	5,200	2,000	2,500	800	1,300

In order to make more realistic predictions a combination of different assays must be used (Kylefors et al. 2003). Also, predictive models must be validated through field observations. Many detail issues are to be studied in the over all evaluation of the landfill cover such as:

- How do the different parts of the structure affect the water balance?
- How are landfill gas emissions affected by and interacting with the liner?
- How is the construction affected by landfill settlements?
- What reactions take place as water percolates through the different layers?
- How should the water above the liner be treated and for how long?
- What are the impacts of external and landfill internal processes on the liner?

During the field test, also mixing procedures and different standard contractor equipment (compacting technique, lay-out procedures etc) are tested. The field tests are the predominant part, including monitoring and sampling from the experimental area (water quantity and quality, gas quality, temperature, settlements; compare Tham et al. 2003 and Travar et al. 2005).

The lab tests include material characterization (elemental analysis; compliance, availability and percolation leaching tests), grain size distribution, curing properties, proctor density, hydraulic conductivity and field capacity. For the assessment of the future development of both function and emissions, also the long-term behaviour will be investigated using physical models (e.g. 80-liter-vessels). A mineralogical characterization of the materials using X-ray diffraction as well as SEM and IR-techniques will give answers regarding reactions of the liner materials with gas and percolation water. How the materials interact within the liner after being mixed with water will be studied with the same type of techniques, in both short time and long time perspective.

Four electric arc furnace (EAF) slags and one ladle[2] slag from Uddeholm Tooling AB were tested. The slags are referred to as slag 1 to 5 whereas 1 to 4 are EAF slags and 5 is the ladle slag. The annually output of the different slags is listed in Table 1.

Slag 1 and 2 are produced under reducing conditions; slag 1 under addition of silicon (FeSi) and recycling steel, slag 2 under addition of FeSi and a chromium substance with high carbon content to the EAF. Furthermore, slag 2 is decarburized with oxygen before slag separation. Slag 3 is produced under oxidizing conditions and addition of iron, molybdenum oxide, oxygen and carbon. After the separation of slag 3, FeCr and FeSi are added to the EAF whereupon slag 4 is drawn off. The steel is treated further at the ladle station and new slag former is added. The treatment at the ladle station occurs under reducing conditions. Slag 4 and 5 disintegrate into fine powder because of the high ratio of Ca/Si.

4. RESULTS AND DISCUSSION

4.1 Grain size distribution

Figure 2 shows the grain size distribution of the tested materials. The EAF slags 1, 2, 3 were crushed to a size smaller than 20 mm at Uddeholm Tooling AB. The sieving shows that they have a very similar grain size distribution. Moreover, they are much coarser than EAF 4 and

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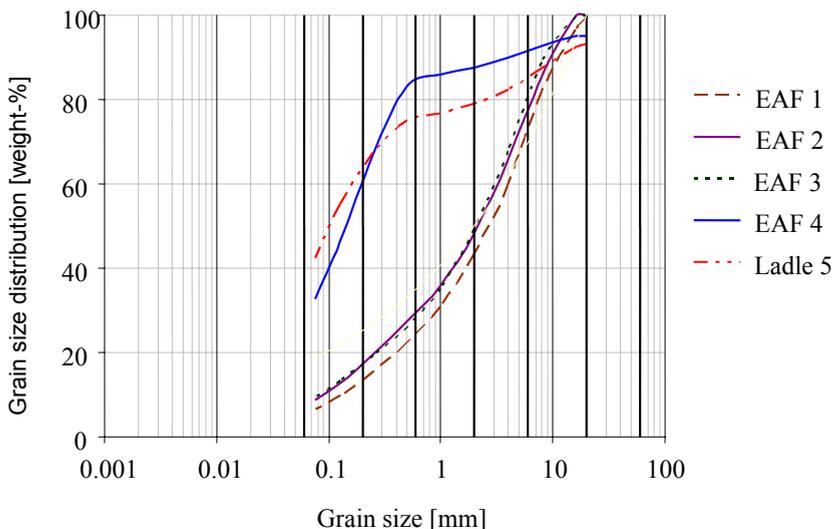[2] Ladle: a vessel for transporting molten metal in a foundry

Figure 2.   Grain size distribution of the tested materials.

ladle slag 5 with only 16 to 20 weight-percent of the particles being smaller than 0.25 mm and 50 % smaller than 2 mm. Even though slag 4 and 5 are different in terms of their generation process they have a number of similarities, to begin with they have similar grain size distributions with two thirds of the particles in the fraction < 0.25 mm. This makes them to a first choice material for the liner within the cover system since the fine structure is a basis for good compaction properties and low permeability values.

The EAF slags 1-3 are intended to be used in the drainage layers. For this purpose the crushing mesh size may be chosen bigger than 20 mm, for example 16/32 mm. However, as described further down, a part of the material is also needed for admixture to the fine slags in the liner in order to decelerate the curing process. The finer fraction generated during crushing seems to be useful for this.

The laboratory tests described below were performed with the fraction < 8 mm of the different materials.

4.2   Curing and compaction properties

The curing behaviour and the proctor density of the slags and different mixtures to be used in the liner were tested.

Curing (after mixing with water) is a special feature of ashes and slags that have cementitious properties which differentiates them from materials conventionally used in landfill covers. Slag 1, 2 and 3 can be described as gravel-like, i.e. they are quite coarse (compare Figure 2) and do not cure. Slags 4 and 5 have curing properties: Without compaction slag 4 is pliable for some time (days if not desiccated) whereas slag 5 hardens to a rock-like consistency after a few hours. The curing of the slags contributes to an increased mechanical stability and a low hydraulic conductivity of the materials. Yet, it should not proceed too far before the material is in place and compacted. For that reason, slags with different properties were mixed in order to achieve a slower and less powerful curing process. Admixtures of 35 to 55 % of EAF slag to the ladle slag resulted in satisfying sample consistency after up to 24 hours.
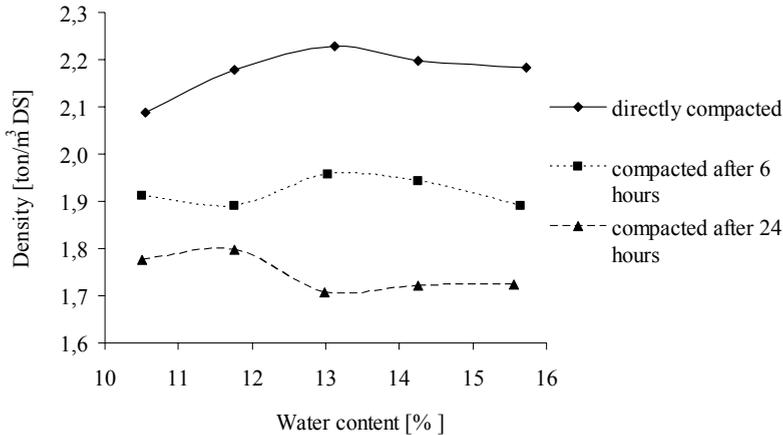
Figure 3.   Variation in Proctor density and optimum water content depending on the point of time of compaction related to the addition of water for ladle slag + 35 % EAF 1

The curing properties of the materials have an impact on the compaction properties. Different optimum water contents and maximum densities were expected depending on how long time passes between the addition of water to the samples and the compaction. In practice, a period of about 8 hours will probably be the maximum, assumed that the material for a whole working day will be prepared at once e.g. in the morning. Therefore, the proctor tests were performed at three different points of time after the addition of water: (1) directly after the admixture of water, (2) 6 hours, and (3) 24 hours later. The proctor densities were in the range of 1.9 to 2.3 t/m^3 TS, depending on time and water content. Figure 3 shows the variation of the compaction properties instancing the sample "Ladle slag + 35 % EAF 1". Longer time between water addition and compaction seems to result in lower densities and the highest density for each curve is achieved at a lower water content compared to direct compaction.

When higher admixtures of EAF (45 and 55 %) were applied the achievable maximum densities increased by about 5-10 % which basically depends on the higher density of the EAF. The optimum water content decreases with increasing admixtures. Compared to soils, the range of possible water content is quite narrow.

4.3   Hydraulic conductivity

The tests for the hydraulic conductivity of the material mixtures were performed according to a factorial design. The investigated factors and their levels are listed in Table 2.

Table 2.   Investigated factors and their levels

Level	Factors		
	A	B	C
	share of admixture of electric arc furnace slag	water content during compaction	time between adding water and compacting
Low	35 %	9 %	0 h
Centre	45 %	11 %	6 h
High	55 %	13 %	24 h

The experimental equipment did not allow more than 10 runs at the same time. Therefore, the experiment was run as an unreplicated full factorial design with two center points which results in $2^3+2 = 10$ runs. The run order was fully randomized. The running time of the tests was extended to about 12 weeks compared to about two to three weeks for soil or clay samples. Yet, for some of the samples the flow did not become stable. Four samples had a permeability smaller than the lowest measurable value, i.e. $< 2.2*10^{-11}$ m/s but for the statistical evaluation all the four values were set to $2.2*10^{-11}$. The hydraulic conductivity of the other samples was the range of $1.4*10^{-8}$ and $4.5*10^{-11}$ m/s, on average $3.5*10^{-9}$ m/s.

Due to these experimental difficulties and due to the lack of replicates, the resulting statistical model was very weak. Yet, it could be seen that an increase of the factors A and C leads to an increase of the permeability. Hence, EAF should be admixed moderately and the time span between mixing and compacting should be kept as short as possible.

According to Swedish legislation, the hydraulic conductivity for a cover system for a landfill for non hazardous waste must not exceed 50 litres per square meter and year. As described above, the cover system consists of a number of different layers that all contribute to achieve the desired function. As can be seen in Figure 1, the largest reduction of potential leachate formation occurs in the layers above the liner, depending on design aspects like the relative permeability of the different layers, their slope and the pipe and trench system design. The covers used are designed to allow only one percent or less of the yearly precipitation to form leachate.

5. SUMMARY AND CONCLUSIONS

Research work on steel work slags as secondary construction materials is in its initial phase. The available results show that:

- It is possible to use the tested steel slags in a landfill cover construction. They can be used in all layers apart from the vegetation layer.
- Only EAF slag 4 might be used as a single material in the liner while the ladle slag needs to be mixed with one (or a mixture) of the EAF slags 1, 2, 3 in order to decelerate the curing process and thereby make it workable.
- The possible range of optimum water content for compaction is quite narrow which makes it necessary to adjust the water content closely.
- However, the water content does not seem to affect the hydraulic conductivity to the same degree.
- The compactibility deteriorates with increasing time between the addition of water and the compaction.
- The cover construction is expected to fulfil the regulations with regard to permitted leachate generation. The full scale test will give knowledge about how the materials work under field conditions.
- Much research is still required in order to show the impact of the slags in the long term.

ACKNOWLEDGEMENTS

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Paper III

HYDRAULIC CONDUCTIVITY OF FLY ASH – SEWAGE SLUDGE MIXTURES USED IN LANDFILL LINERS

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Hydraulic conductivity of fly ash - sewage sludge mixtures used in landfill liners

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Abstract

The effect of several factors on the hydraulic conductivity (HC) of fly ash – sewage sludge mixtures was investigated using a 2^{7-1} fractional factorial design. The aim was to identify the factors that influence HC, to quantify their effects and to assess how a sufficiently low HC can be achieved. The factors "compaction energy" and "drying", as well as the factor interactions "material×ash ratio" and "ash ratio×compaction energy" affected HC significantly ($\alpha = 0.05$). The effect of the compaction energy was very large compared to the other factor effects. The HC data were modelled using multiple linear regression. The derived models were reliable as indicated by R^2 values between 0.76 and 0.88. Independent on the ash ratio and the material, HC was predicted to be between 1.7×10^{-11} m s^{-1} and 8.9×10^{-10} m s^{-1} if the compaction energy was 2.4 J cm^{-3}, the ash ratio ≤ 75 % and drying did not occur. Thus, the investigated materials met the limit value for non-hazardous waste landfills of 10^{-9} m s^{-1}. As the compaction energy affected HC the most, sufficient compaction should be ensured when the material is used in landfill liners.

Keywords: Hydraulic conductivity, Fly ash, Sewage sludge, Landfill liner, Permeability

1. Introduction

Recent EU legislation prescribing stricter rules for the landfilling of waste leads to the closure of many landfills that consequently need to be covered. In Sweden, the permeability of a cover construction must not exceed 50 l m^{-2} yr^{-1} on non-hazardous landfills and 5 l m^{-2} yr^{-1} on hazardous landfills [1]. To meet these limit values, a well

sealing liner should be included in the cover. A commonly used liner material is clay. Recently, efforts have been made to substitute clay by secondary materials such as ashes and slags which are generated in increasing amounts. In Sweden, the annual production is estimated at 447,000 tons of waste incineration ash [2] and 1.3 million tons of steel slag (Jernkontoret, Stockhom Sweden, in 2002). These residues could receive a new field of application while natural clay resources could be saved.

In this study, mixtures of incineration fly ash and sewage sludge are investigated towards their suitability as liner material in terms of hydraulic conductivity (HC), considering various conditions that may occur in a cover liner. The aim is to (1) identify the factors that influence the HC, (2) quantify the factor effects on HC and to (3) assess how a sufficiently low HC can be achieved.

2. Material and Methods

2.1 Material

The fly ashes investigated in this study were sampled from the combined power and heating plants "Mälarenergi" in Västerås and "Öresundskraft" (Västmalmsverket) in Helsingborg, Sweden. The incinerator at Öresundskraft is a dust firing steam boiler treating wood and coal powder. The fly ash from Öresundskraft is composed of both electric filter ash and desulfurization residues from the bag filters. It consists of coal and wood ash in equal shares. At Mälarenergi, the fly ash was sampled from the circulating fluidized bed boiler P5 where wood is used as fuel. The flue gas cleaning is carried out by bag filters and flue gas condensation.

Two types of digested sewage sludge were used for the experiments: one from Nordvästra Skånes Renhållnings AB (NSR) in Helsingborg which was composed of sludge from four different municipal waste water treatment plants, and the other one from Stockholm Vatten's treatment plant in Bromma.

For the experiment, fly ash and sewage sludge were mixed as follows:
- fly ash from Mälarenergi and sewage sludge from Stockholm Vatten, hereafter referred to as **Stockholm** mixture,
- fly ash from Öresundskraft and sewage sludge from NSR, hereafter referred to as **Helsingborg** mixture.

2.2 Experiment

HC was tested according to the Swedish standard [3] using Darcy's equation for calculating HC. The experiment was designed according to a 2^{7-1} fractional factorial design [4] with 7 factors and 8 center points (Table 1). With this design, the effect of the factors and their interactions on the response (HC) could be investigated. 72 samples composed of mixtures of fly ash and sewage sludge were compacted into columns that were connected to a water reservoir (Figure 1). The amount of the permeated water was monitored.

Table 1   Factors and their levels investigated in the experiment

Factor	Level		
	low	center	high
Material	Stockholm	Stockholm/ Helsingborg[b]	Helsingborg
Ash ratio in the mixture [g g^{-1}]	0.2	0.5	0.8
Initial water content [wt.%][a]	20	30	40
Compaction energy [J cm^{-3}]	0.2	0.8	2.4
Freezing	No	No/Yes**	Yes
Drying	No	No/Yes**	Yes
Biological activity	No	No/Yes**	Yes

[a] Water content: mass of water divided by mass of dry solids

[b] The qualitative factors did not have a center level. For the center points, these factors were set to either low or high level.

The experiment was conducted in 5 cycles resulting in 5 readings of HC per sample. Each reading is a mean value of three measurements. The duration of each cycle had to be adapted to the samples with the slowest water flow. Measurements for samples with faster water flow were taken at the beginning of each cycle, then, the samples were disconnected. Due to the practical accomplishment of the experiment, cycle 1 lasted 105 days, cycle 2 25 days, cycle 3 36 days, cycle 4 37 days and cycle 5 75 days. The whole experiment lasted 10.5 months.
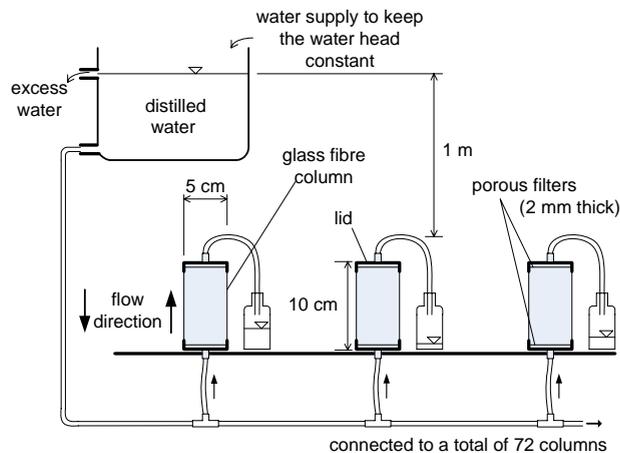
Figure 1   Experimental set-up

The COMPACTION ENERGY was varied to obtain different levels of compaction. High level (i.e. 2.4 J cm^{-3}) was obtained by filling the columns with three layers of material and compacting each layer with ten blows using a Proctor hand hammer. For the center point (i.e. 0.8 J cm^{-3}), two layers were compacted with five blows each. At low level (i.e. 0.2 J cm^{-3}), the material was compacted merely by hand, making three layers with

ten hand shoves each. The achieved wet densities were determined by weighing. Dry density was calculated.

The samples that were exposed to FREEZING (24 hours) or DRYING (72 hours at 50-55°C) were frozen or dried between the cycles, i.e. four times (starting after the first cycle). For FREEZING, the top lid was removed, for DRYING, both lids were removed. To evaluate the impact of biological degradation of organic matter on HC, to half of the samples, the double lethal dose (identified by luminescence inhibition on the bacteria *Vibrio fischeri*) of sodium azide (NaN_3) was added to the permeating water to inhibit microbial activity.

2.3 Statistical evaluation

The data of each cycle were evaluated using multiple linear regression (MLR) [5] (α = 5 %). The correlation between the factors and the response variables was assessed by multivariate data analysis using partial least squares (PLS) modeling. As the response variables were not normally distributed, they were log-transformed before the statistical evaluation.

3. Results

3.1 Factors influencing HC

HC was affected by different factors depending on the cycle (Table 2). COMPACTION ENERGY influenced both HC and dry density the most (Figure 2 and 3). Higher COMPACTION ENERGY led to higher dry density and lower HC. The factor interaction MATERIAL×ASH RATIO had the next biggest effect in cycles 2-5 (Figure 3). The Stockholm mixture generally showed a lower HC and was better compactable than the Helsingborg mixture (cycles 2-4) (Table 2). The effect of the INITIAL WATER CONTENT was also considerable, but only in cycle 1 (Figure 3). The factors FREEZING and BIOLOGICAL ACTIVITY did not show any effect (Figure 2), but DRYING influenced HC in cycle 2 and 5 (Table 2).

Table 2   Factors and factor interactions affecting dry density and HC. Positive effect, +; negative effect, -; no significant effect, 0. If a factor has a negative effect, higher levels of the factor decrease the response value and vice versa. R^2, coefficient of multiple determination; $R^2_{adj.}$, adjusted R^2 [4].
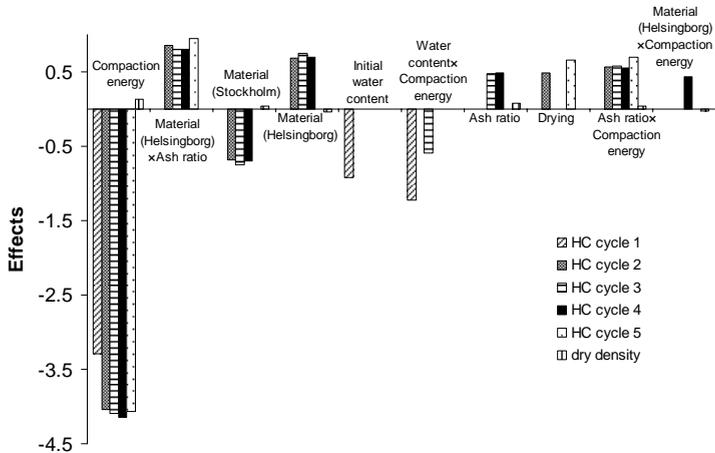
Factor	Hydraulic Conductivity (HC)					Dry Density
	Cycle 1	Cycle 2	Cycle 3	Cycle 4	Cycle 5	
Material (Stockholm)	0	-	-	-	0	+
Material (Helsingborg)	0	+	+	+	0	-
Ash ratio	0	0	+	+	0	+
Water Content	-	0	0	0	0	0
Compaction energy	-	-	-	-	-	+
Drying	0	+	0	0	+	0
Material (Stockholm)× Ash ratio	0	-	-	-	-	0
Material (Stockholm)× Compaction energy	0	0	0	-	0	+
Material (Stockholm)× Drying	0	0	0	0	0	-
Ash ratio× Compaction energy	0	+	+	+	+	+
Water Content× Compaction energy	-	0	-	0	0	0
R^2	0.76	0.86	0.88	0.87	0.84	0.82
$R^2_{adj.}$	0.75	0.84	0.86	0.85	0.83	0.80

3.2 Models

The MLR models obtained for the five cycles were good and reliable as indicated by R^2 values between 0.76 and 0.88 and $R^2_{adj.}$ values between 0.75 and 0.86 (Table 2). Plotting the model residuals against the factors, both the COMPACTION ENERGY and ASH RATIO plot showed curvature in the residuals, i.e. the residuals of the centre points deviated from a straight line. In the PLS analysis, three principal components were identified as significant according to the criterion "Eigenvalue > 1". The Eigenvalue represents the amount of variance accounted for by a component [6]. The three components explained 76 % of the data variability. The first and third component are visualized (Figure 2).
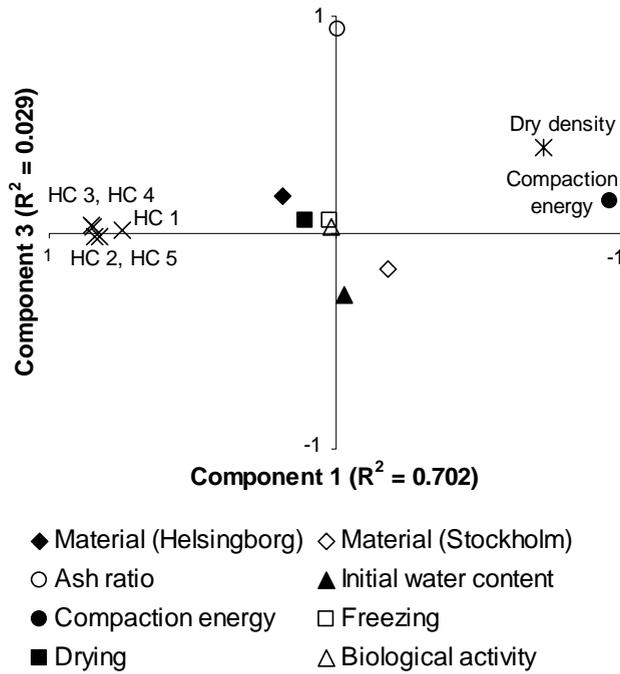
Figure 2 PLS loading plot showing relations between the investigated factors and the responses

Figure 3 Centered and scaled effects on hydraulic conductivity and dry density determined using MLR. Positive effect: HC and density increase with an increase in the respective factor setting. Negative effect: vice versa.

3.3 Quantification

The effects of the factors significantly influencing HC (Table 2, Figure 3) were quantified for cycle 5 (Eq. 1), i.e. for the last HC measurements that were taken in the experiment. The equation represents the model that was derived using MLR. It can be used to calculate HC under different conditions. To obtain a low HC, a high COMPACTION ENERGY of 2.4 J cm^{-3} must be applied, the ASH RATIO must not be higher than 75 % and DRYING must not occur. Then, the HC of the two mixtures was between 1.7×10^{-11} m s^{-1} and 8.9×10^{-10} m s^{-1}. However, the absolutely lowest HC of 1.7×10^{-11} m s^{-1} was achieved using the Helsingborg mixture with an ASH RATIO of 20 %. The Stockholm mixture reached its lowest HC of 5.7×10^{-11} m s^{-1} at an ASH RATIO of 80 %.

$\log_{10}$ (HC) [m s^{-1}] =
- 4.791
+ 0.581×MATERIAL
− 0.958×ASH RATIO
− 2.373×COMPACTION ENERGY
+ 0.327×DRYING
− 1.571×MATERIAL×ASH RATIO
+ 1.047×ASH RATIO×COMPACTION ENERGY;   (Eq. 1)

where

MATERIAL (Stockholm) = 1,
MATERIAL (Helsingborg) = -1,
DRYING (yes) = 1,
DRYING (no) = -1,
the other factors in their units specified in Table 1.

4. Discussion

According to Swedish legislation, the HC of a landfill cover must not exceed 5 and 50 l m^{-2} yr^{-1} on landfills for hazardous and non-hazardous waste, respectively [1]. If cover liners exhibit a HC $<10^{-9}$ m s^{-1}, they are expected to meet the limit value for non-hazardous waste landfills. As a HC of between 1.7×10^{-11} m s^{-1} and 8.9×10^{-10} m s^{-1} could easily be obtained with the investigated ash - sludge mixtures, the limit value is met with wide margin. Muhunthan, *et al.* [7] found the HC of pure fly ash to be 2.2×10^{-9} m s^{-1}. Thus, the admixture of sewage sludge to fly ash could decrease HC by up to two orders of magnitude. Lim*, et al.* [8] found the HC of sewage sludge mixed with lime and fly ash to be between 10^{-6} and 10^{-10} m s^{-1} which also affirms that the admixture of sewage sludge can decrease the HC of fly ash.

4.1 Discussion of factor effects

Depending on the cycle, the factors significantly affecting HC differed (Table 2). Only the COMPACTION ENERGY affected HC in every cycle. In the first cycle, HC was conditioned by COMPACTION ENERGY and WATER CONTENT only, whereas the WATER CONTENT had no effect in the subsequent cycles. The first cycle thus behaved clearly different, maybe because of start-up difficulties and initially unstable measurements. DRYING affected HC only in cycle 2 and 5, and the interaction MATERIAL×COMPACTION ENERGY only in cycle 4. One reason for such factor variations may be that data variability changed from one cycle to another. However, the effects of all factors except COMPACTION ENERGY were rather small (Figure 3) so that they might be regarded to be of less importance. COMPACTION ENERGY, however, affected HC the most and its significance could be proved in all 5 cycles. This confirms the finding of Nhan *et al.* [9] who observed that higher compaction forces lead to a decrease in HC of coal fly ash. Thus, it is crucial to apply a high compaction energy when compacting ash - sludge liners on a landfill.

In cycles 2-4, the HC of the samples depended on the MATERIAL mixture that was used (Table 2). Additionally, in cycles 2-5, the interaction MATERIAL×ASH RATIO (and in cycle 4 also MATERIAL×COMPACTION ENERGY) affected HC which means that the materials behaved differently. Although the influence of the MATERIAL on HC was low (Figure 2), the different behaviour of fly ash – sewage sludge mixtures must be taken into account when such mixtures are to be used in landfill cover liners. A general guideline for handling any fly ash – sewage sludge mixture cannot be outlined on the basis of these results. On the contrary, as the properties of others than the investigated mixtures cannot be predicted, every material mixture must be tested on HC before it is used in a liner.

The ASH RATIO did not influence the HC (Figure 2) since the factor is positioned close to zero on Component 1. As Component 1 explains a larger part of the data variability than Component 3 ($R^2_{Component\ 1}$ = 0.702, $R^2_{Component\ 3}$ = 0.029), the PLS plot (Figure 2) should be evaluated in the direction of Component 1. It was chosen to span the plot over Component 1 and 3 because in this way, it shows very clearly the correlations between the factors and the responses. However, the ASH RATIO was significant in cycles 3 and 4 and is also involved in the factor interactions MATERIAL×ASH RATIO and ASH RATIO×COMPACTION ENERGY (Table 2) which contradicts the findings from the PLS analysis. Howsoever, the ash ratio should be considered when fly ash – sewage sludge mixtures are applied in landfill liners.

A commonly used liner material is clay. As clay is susceptible to freezing and desiccation, it is general practice to overlay landfill cover liners with a sufficiently thick protective layer so that the liner is at frost-free and desiccation-free depth. FREEZING did not show any effect on the HC of fly ash - sludge mixtures (Figure 2), but DRYING affected HC in cycles 2 and 5 (Table 2) and should thus be avoided so that it is important to apply a sufficiently thick protective layer.

The factor BIOLOGICAL ACTIVITY did not affect HC (Figure 2), i.e. there was no difference in HC between the samples where biological activity was allowed and those

where it was inhibited. As the occurrence of biological activity was not measured, it is unknown if biological degradation occurred in either of the samples. Mácsik*, et al.* [10] investigated the biological degradation of the Stockholm mixture with ash ratios of 40 and 60 wt.-% by measuring the formation of biogas. Samples containing 60 wt.-% fly ash did not form any biogas during the measuring period of more than two years. Samples containing only 40 wt.-% fly ash generated biogas after a lag-phase of two months, but only if the leachate (containing e.g. salts) was removed. It is thus possible that biological activity was established in the samples that were investigated in this study and had a low ASH RATIO. However, it is unlikely that biological activity took place in the samples with high ASH RATIO. It is not possible to assess the effect of biological degradation because it is uncertain which samples exhibited biological activity. To investigate the occurrence of biological activity in each sample, the analysis of (total) organic carbon in the solid phase and in the leachates could be a suitable measure. Even if biological activity and therewith decomposition of the material occurs, it does not necessarily increase HC. Mäkelä and Höynälä [11] investigated the HC of fibre sludge and found that a decrease of organics in the sludge by e.g. decomposition decreases HC because the material becomes more consolidated. During the tests of Mácsik*, et al.* [10], no carbon dioxide generation was observed as carbon dioxide precipitated directly as calcium carbonates in the alkaline material. In those samples, the calcium carbonate precipitated in the pores which reduces the porosity and possibly the HC.

Not all factors that might affect HC in fly ash - sludge mixtures were investigated in this study. The HC of fly ash e.g. decreases with CaO content in the ash [9]. Furthermore, the process of carbonation can increase HC in fly ash by up to two orders of magnitude [12]. The ashes received from Stockholm and Helsingborg were possibly carbonated to a different extend and had different CaO contents which may have contributed to the differences in HC between the two material mixtures. Carbonation is expected to occur when the material is used in the cover of a biologically active landfill. As described above, the calcium carbonate formed probably precipitates in the pores and decreases HC. In the tests of Meynaud [12], the samples were carbonated before they were compacted and HC was tested. Thus, fly ash should not be highly carbonated before the liner is installed as this would increase HC [12], but carbonation occurring after liner installation probably decreases HC. A longer curing time also decreases HC [9]. Curing (or hardening) is the process of the reaction between fly ash compounds and water (hydration) leading to an increased strength of the material. In this study, the materials were not cured before HC was measured. However, some curing has certainly occurred during the experiment but its effect was not investigated.

4.2 Time aspect

14 of the 72 samples showed a quite stable and acceptably low HC of $<10^{-9}$ m s^{-1} during all cycles. One of these samples shall be taken as an example for the subsequent calculation as its HC was in average as low as 4.7×10^{-10} m s^{-1} but it still reached a comparably high L/S ratio of 1.8 during the experiment. In a 0.5 m thick landfill liner with a dry density of 0.954 t m^{-3} (as obtained in aforesaid sample), it would take about 116 years to reach this L/S assuming aforementioned HC. It can thus be presumed that

the HC of the material in the landfill liner will be stable at least for this time period. Comparing the complete HC data sets of the 5 cycles, no statistically significant difference in HC could be detected between the cycles, which indicates that HC was generally stable throughout the experiment.

4.3 Correlation HC and dry density

HC was strongly correlated to the material's density: the higher the density, the lower the HC (Figure 2). COMPACTION ENERGY was positioned furthest away from the origin of the PLS plot which shows that both the density and the HC were mostly influenced by this factor (Figure 2). However, the influence of the investigated factors on dry density was generally small (Figure 3). In the 14 samples with continuously low HC, the average dry density was 0.893 t m^{-3} with a standard deviation of 0.158 t m^{-3} resulting in a variation coefficient of 18 %. All of these 14 samples were compacted with high COMPACTION ENERGY. The small standard deviation and variation coefficient indicate that it was relatively easy to obtain a sufficient density when applying high COMPACTION ENERGY, regardless the settings of the other factors.

4.4 Models

Plotting the model residuals against the factors COMPACTION ENERGY and ASH RATIO showed a strong curvature in the data which can be addressed by adding a quadratic term to the model. However, on the basis of the available data, it could not be detected which of the two factors was responsible for the quadratic effect. As COMPACTION ENERGY had the biggest influence on the responses, it can be assumed that mainly this factor caused the curvature. To separate the (quadratic) effects, more experiments would be necessary. However, adding the quadratic term would improve the model only marginally and hence it was not added to the models in this study. This limitation to the models might have led to the incidence that the factor interaction MATERIAL (STOCKHOLM)×DRYING was identified to significantly affect dry density (Table 2). As the material was dried after the dry density was measured, this observation cannot be true. When the significance level was lowered to $\alpha = 1$ % this factor was not significant any more. However, the model was generally good and reliable as indicated by R^2 values between 0.76 and 0.88 and R$^2_{adj.}$ values between 0.75 and 0.86 (Table 2).

4.5 Environmental Impact

Sewage sludge and fly ash contain potential contaminants such as heavy metals that possibly are released from the material when put on a landfill. Sewage sludge often contains large concentrations of trace elements, including Cu and Pb [13]. Fly ash contains, among others, Cd, Cu, Cr, Zn, Ba, Pb, and Hg [14]. However, the leaching of heavy metals from sludge - lime - fly ash mixtures has been found to be small [8]. Wang and Viraraghavan [15] report that sewage sludge - fly ash mixtures can be used for land application without groundwater contamination to occur. Fly ash is rich in Ca compounds and clay-like minerals that are generally efficient for the immobilization of

metals [16]. The mobility of Cu and Pb is usually lowest at slightly alkaline pH while highly alkaline conditions can have a reverse effect on Cu and Pb stability due to the amphoteric nature of these metals [17]. Soluble hydroxide complexes formed at pH >10 can increase the mobility of Cu and at pH >11-12 of Pb [17,18]. However, mixing organic matter and fly ash can be beneficial for enhancing metal sorption on clay-like mineral surfaces through ternary metal-mineral-humic acids complexes [19,20]. Hizal and Apak [20] noted that clay minerals in the presence of humic acids can behave more like chelating ion-exchanger sorbents for metals rather than being simple inorganic ion exchangers.

5. Conclusions

The hydraulic conductivity (HC) of fly ash - sewage sludge mixtures was between $\leq 1.2 \times 10^{-11}$ m s^{-1} and 1.0×10^{-4} m s^{-1}. However, for the two investigated fly ash – sewage sludge mixtures, the HC could be kept between 1.7×10^{-11} m s^{-1} and 8.9×10^{-10} m s^{-1} if the compaction energy was ≥ 2.4 J cm^{-3}, the ash ratio was ≤ 75 % and drying did not occur. The material mixtures are very promising for use in liners as they meet the HC limit value for non-hazardous waste landfills of 10^{-9} m s^{-1}.

Modelling ($\alpha = 5$ %) predicted that the absolutely lowest HC of 1.7×10^{-11} m s^{-1} can be achieved using the Helsingborg mixture at an ash ratio of 20 %, a high compaction energy of 2.4 J cm^{-3} and without any desiccation occurring. The Stockholm mixture reached its lowest HC of 5.7×10^{-11} m s^{-1} under the same conditions but at an ash ratio of 80 %. If other fly ash - sewage sludge mixtures are to be used in a landfill cover liner, their HC should be tested depending on the ash ratio.

The compaction energy that was applied on the material influenced HC the most and thus, special care has to be taken when compacting the material on a landfill. It should be tested how a sufficiently high compaction energy can be applied in practice. Drying increases HC and should be avoided. Freezing and thawing did not affect HC. The effect of biological activity could not be assessed because it was uncertain if the samples really exhibited biological activity. To investigate if biological degradation of organic matter has occurred in the samples, the analysis of (total) organic carbon in the solid phase and in the leachates could be a suitable measure. The water content of the mixture influenced HC only initially. HC is dependent on the material. The Stockholm mixture showed a lower HC than the Helsingborg mixture during most of the experiment. Not all factors that may affect HC were investigated in this study. The effect of carbonation, CaO content and curing time on HC in fly ash - sewage sludge mixtures should be subject to further research.

This study focused on the HC of fly ash - sewage sludge mixtures. The chemical, biological and mechanical stability of the material is still unknown and could not be assessed on the basis of the results from this study.

6. Acknowledgements

7. References

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