Abstract

According to the state of technology any waste deposition has to be secured by a multibarrier encapsulation system. Surface liners and base liners have to be constructed in different ways because they serve different purposes. Surface liners have to prevent precipitation water to infiltrate into the waste and base liners seal the waste from the underground rock resp. groundwater. Standard combined systems consist of mineral barriers and geomembranes. A drainage system at the base for seapage control and a gas collection installation at the surface in case of municipal waste complete the system.

1. Clay Liners and waste disposal

As populations grow and technologies advance, ever-more wastes are produced in ever-growing quantities. Of the many and varied environmental problems, encapsulation of wastes in order to restore sites and protect the environment is the topic of this chapter. Encapsulation means sealing the waste body by geological and engineered liner systems which in most cases partly consist of clay liners in different modifications. Encapsulation systems are as varied as the environments in which they are built, and the components of an encapsulation system are as multiple and complex as the wastes therein. For all waste types, encapsulation is the only option: permanent isolation from the accessible environment (Caldwell & Reith, 1993). The requirements for an encapsulation system are basically the same, whether the waste is municipal refuse in a landfill, hospital debris in a low level waste dump, or mixed wastes of diverse industrial productions or construction activities. This leads us to the need to classify wastes because encapsulation systems consist of engineered liner components according to the magnitude of the risks originating from the waste.

From the beginning: sophisticated engineered liners serve two purposes. They have to guarantee practical imperviousness to prevent leachates from infiltrating the environment and secondly have to prove retention or at least retardation properties to prevent contaminant migration by convection and diffusion (Drescher, 1997). In many cases - but not absolutely - the surface barrier might be designed as slightly permeable layer, because further decomposition of sanitary wastes by precipitation moisture could be achieved. Because of the different functions of the surface and the base encapsulation barriers, different systems are state of the art. In any case, for most of the required properties, clay liners are optimal.
Different waste compositions require different sealing units. E. g. it is not economical to use a multilayer system for inert construction wastes or to design the same systems for sanitary landfills and toxic industrial wastes (Bradshaw et al., 1992). Therefore all regional waste repository regulations have to classify waste categories not by means of the input of waste components but mainly by leaching the waste and determining quantitatively the contaminant content (Czurda, 1992). The European Union, most of the European states, the USA, Canada, Japan etc. follow this system and ended up with similar waste categories. Taking radioactive waste into account as well we roughly have to follow the scheme:

- inert construction and industrial waste
- domestic waste
- toxic industrial waste
- incineration ashes and slags
- radioactive waste

Nuclear waste repositories have to follow special national regulations and are not further treated in this chapter.

As an example for waste assignments some threshold values according to German regulations (TA Abfall, 1991. TA Siedlungsabfall, 1993, Deponie- verordnung, 2002) are shown in table 1. They are close to European Union values (EU-Richtlinie, 1999)
As mineral barriers within engineered sealing layers, but as well as constituents of in situ geological barriers - which means the waste deposit location - clay rocks, clay mineral admixtures and zeolite admixtures are the most important and widely used natural materials. Alternative materials like amorphous silica, fly ashes, fly ash zeolites, clay remnants from coal flotation etc. are not treated in the following chapters.

### 1.2 Mineral barriers

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#### 1.2.1 Clay rocks and clay minerals

In using clays and zeolites and other fine grained material for sealing purposes, two main issues are persued: leachate retention by low hydraulic conductivities and toxic constituent retention or retardation by retention mechanisms like adsorption, precipitation, redox processes and others. Soil barriers, containing enough and function adequate clay minerals to provide low permeability, are used extensively to prevent rapid advective migration by various leachates from waste disposal sites (Hiltmann & Stribny, 1998). The clayey barriers vary from thin geosythetic clay liners (GCL) with 1–3 cm thickness, to compacted clay liners (CCL) up to 300 cm thickness, to natural undisturbed clayey barriers up to 30 m or more in thickness. The hydraulic conductivity of undisturbed clayey deposits depend on the mineralogy, the environment of deposition and the stress history of the deposits. The same is valid for GCL's and CCL's.

The outstanding properties of clay minerals are to be seen in their extremely small grain size (by definition < 2 µm), their negatively charged surface properties of the basal planes, the positively charged broken ends of the platelets and the large surface areas. Most clays in natural settings have a phyllosilicate or sheet structure. Clay minerals have a common structure but they are almost always the result of chemical changes or thermal variations in the range of near surface conditions. Clays attract water, other polar liquids and cations. A dried out clay will expand greatly as it adsorbs water between its layers when attached by aqueous solutions. If toxic ions are constituents of the solution an ion exchange on charged surface sites will result. That is to say, they can accept or release ions depending upon the concentration of the ions in solution relative to that of the clays. This ions, e. g. from the leachate, are not finally fixed but are at disposal for further exchange process depending on the chemical environment. Table 2 gives an overview on cation exchange capacities (CEC) and specific surfaces.

<table>
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<tr>
<th></th>
<th>I</th>
<th>II</th>
<th>III</th>
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<tr>
<td>conductivity</td>
<td>6000 µS/cm</td>
<td>50000 µS/cm</td>
<td>100000 µS/cm</td>
</tr>
<tr>
<td>uniaxial strength</td>
<td>50 kN/m_</td>
<td>50 kN/m_</td>
<td>50 kN/m_</td>
</tr>
<tr>
<td>TO C</td>
<td>20 mg/l</td>
<td>100 mg/l</td>
<td>200 mg/l</td>
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<tr>
<td>phenole</td>
<td>0,2 mg/l</td>
<td>50 mg/l</td>
<td>100 mg/l</td>
</tr>
<tr>
<td>mercury</td>
<td>0,005 mg/l</td>
<td>0,020 mg/l</td>
<td>0,100 mg/l</td>
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<tr>
<td>cadmium</td>
<td>0,050 mg/l</td>
<td>0,100 mg/l</td>
<td>0,500 mg/l</td>
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<tr>
<td>lead</td>
<td>0,200 mg/l</td>
<td>1,000 mg/l</td>
<td>2,000 mg/l</td>
</tr>
<tr>
<td>sulfate</td>
<td>500 mg/l</td>
<td>1400 mg/l</td>
<td>5000 mg/l</td>
</tr>
<tr>
<td>soluble part</td>
<td>3 %</td>
<td>6 %</td>
<td>10 %</td>
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Table 1: Assignment criterion for waste categories according to the German regulations TA-A and TA-Si. Selected examples. Waste category I means inert waste, II domestic waste and III toxic industrial waste. The leachable parts are listed as examples for very common toxic waste constituents.
Of decisive influence on sorption potentials are the primarily fixed cations, e.g. from the marine environment, on the crystallographic basal surfaces. According to the diameter of hydrated cations and their valency they are differently sorbed to the clay surface and are therefore to different quantities exchangeable. For base liner clays as an example Na-bentonites are especially suitable because of their high swelling potential and sorption capacities and therefore fulfil the high degree of imperviousness and the high contaminant retention potential required.

### Zeolites

Zeolites are tectosilicates with 3-dimensional aluminosilicate structures containing water molecules, alkali and alkaline earth metals in their structural framework. They have proven a high potential as contaminant sorbents due to their high exchange capacity and their selectivity for certain constituents such as NH₄, Pb, Cd, Sr and others, especially when they are activated by sodium chloride. The selectivity of certain zeolite minerals for specific chemical compounds is defined by pore size and

<table>
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<th>mineral</th>
<th>CEC millimol equiv./100g</th>
<th>spec. surface m²/g</th>
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<tr>
<td>allophan</td>
<td>50 - 100</td>
<td>500 - 700</td>
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<tr>
<td>kaolinite</td>
<td>3 - 15</td>
<td>10 - 20</td>
</tr>
<tr>
<td>illite</td>
<td>20 - 50</td>
<td>90 - 100</td>
</tr>
<tr>
<td>smectite</td>
<td>70 - 130</td>
<td>750 - 800</td>
</tr>
<tr>
<td>bentonite</td>
<td>95 - 100</td>
<td>800</td>
</tr>
<tr>
<td>vermiculite</td>
<td>150 - 200</td>
<td>750 - 800</td>
</tr>
<tr>
<td>Fe- + Al-hydroxide (pH 8,0)</td>
<td>3 - 25</td>
<td>25 - 42</td>
</tr>
<tr>
<td>humic material</td>
<td>150 - 250</td>
<td>800</td>
</tr>
</tbody>
</table>

Table 2: CEC (cation exchange capacity) and specific surfaces for different clay mineral species and other materials

Fig. 3a: 3-layer clay mineral illite with tetraeder/octaeder/tetraeder arrangement and interlayer space between the O-planes of the tetraeders.

Fig. 3b: Illite aggregate with hexagonal clay flakes, scale: 330x
charge properties of the structure. The unbalanced substitution of Si$^{4+}$ by Al$^{3+}$ in the crystal lattice leads - like in clay minerals - to a net negative charge and, subsequently, to the high cation exchange capacity of most natural zeolites.

Natural zeolites occur in sediments, lava vesicules, deuteric altered plutonic rocks and hydrothermal systems associated with alkaline volcanic rocks. In summary, natural zeolites generally originate from a volcanic glass precursor and are therefore very common in nature.

In Fig. 4, in order to show the retention potential, sorption isotherms are presented (Huttenloch et al., 2001). The example shows the sorption of Cu form
Fig. 6: base liner systems in comparison. The multibarrier system consists of two clay units: an adsorbing bentonite unit and a sealing kaolinite unit.

Two layer minerals
- low swelling / shrinkage
- low sorption
- low self healing
- high permeability

Fig. 7: REM image of kaolinitic clay, scale 5000x. High non communicating microporosity, electrically neutral two layer mineral without swelling potential. Preferable mineral phase for surface sealing.

Fig. 8: Crystal structure of two layers clay mineral (e.g. kaolinite).
dionised water and Cu from a 0.01 M Ca Cl2 matrix solution on clinoptilolite (a very common natural zeolite spezies) and Na-clinoptilolite respectively. The isotherms indicate a non-linear sorption behaviour between sorbed and aqueous concentration of the contaminant, that can be described by the Freundlich equation: 
\[ c_s = K_f \cdot c_w^{N_f} \]
where \( c_s \) designates the amount of the solute sorbed per unit mass of the sorbent, \( c_w \) = the equilibrium solute concentration, \( K_f \) and \( N_f \) are empirical parameters specific to the sorption material used.

### 1.3 Waste deposit multibarrier system

Waste deposits can in principle be constructed as underground storages and at the surface as slope storage, slope dump or depression storage. The common domestic waste, incineration remnants, inert construction wastes etc. are stored at the surface. Underground storage as a special deposition mode is not treated in this chapter.

#### 1.3.1 Base liners

Base liner systems have to prevent leakage from the waste to infiltrate into the subsoil and in addition to guarantee a high potential in toxicant retention by sorption, precipitation and/or redox processes (Rowe et al., 1995). Sorption on mineral surfaces means ion exchange according to chemical environment changes and surface charge properties. In many cases toxicants can be retarded during their migration through the sealing layers.

As an example Figs. 5 a and b show the base liner construction according to German regulations for inert wastes and domestic wastes. Essential are compacted clay layers and in case of domestic wastes a geomembrane in addition to the mineral layers and of course the geological barrier. The basal system contains a leakage collecting layer, connected with a leakage purification plant. There are different leakage detection systems on the market.

#### 1.3.2 Surface liners

The functions of the surface liner systems have exclusively to be seen in preventing the precipitation water from infiltrating into the waste. In case of household wastes the capping system in addition has to have a gas drainage system. Capping layers for all types of waste of course are constructions with a drainage layer (usually gravel 16/32 mm) in case of leaks in the system.

Like in the case of basal systems, compacted clay liners (CCL), and geomembranes are the prevailing sealing elements. But there is an important differ-
ence in the clay mineral composition of the CCLs. Whereas the base CCL-clay should contain 3-layer-minerals as index minerals, e.g. montmorillonite, vermiculite etc., the surface CCL-clay should contain 2-layer-minerals as index minerals, e.g. kaolinite, etc. The 3-layer-clays enable retardation by adsorption and guarantee a high degree of impermeability and the 2-layer-clays of the surface sealing unit combined with sand/silt-matrix are practically impermeable as well but are weak in their adsorption potential. The latter is not required for the surface.

Fig. 7 shows the crystal lattice scheme of kaolinites, a two layer mineral, which combines an aluminium octahedron layers and silica tetrahedron layers. The center, and by their valency electrically balancing ion, is $\text{Al}^{3+}$ respectively $\text{Si}^{4+}$. Because of the lack of negative surface charges the ion adsorption potential is extremely low. Because the particle is - like all other clay mineral particles - extremely small (< 2 µm), a sand-silt-kaolinite admixture for the mineral surface sealing can gain very low permeability values ($k_f$ 10^{-8} to 10^{-12} cm/sec).

**Conclusion**

For hazardous industrial wastes and toxic sanitary landfills we have to locate a site which is primarily a geologic barrier, $k_f < 10^{-6}$ m/sec and at least of 3 m thickness. In case of inert (non toxic) wastes a geologic barrier is not necessary. Essential however is to follow the multibarrier concept and to add on top of the geologic barrier a system of engineered barriers and drainage layers. The engineered barriers comprise as a core unit the combined CCL and geomembrane double layer. A similar multibarrier system has to be constructed for the cover sealing. The difference is expressed in the type of GCL and the drainage layers. The GCL should not contain expanding three layer minerals like montmorillonite or vermiculite. They tend to dry out, forming desiccation cracks. Therefore non swelling two layer minerals like kaolinite or tectosilicates like zeolites should be the index minerals for the sand-silt-clay surface GCL. In case of untreated household waste, a gas drainage layer has to be foreseen to divert the methanol, developing during the waste decay.

**References**


