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► **To cite this version:**

Florent Hénon, Gerald Debenest, Xavier Lefebvre, Sébastien Pommier, Damien Chenu, et al.. Simulation of Transport and Impact of Moisture Content on the Biodegradation. HPM4 2011, Apr 2011, Santander, Spain. pp.0. hal-03617735

HAL Id: hal-03617735

<https://hal.science/hal-03617735>

Submitted on 23 Mar 2022

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To cite this version : Henon, F. and Debenest, G. and Lefebvre, X. and Pommier, S. and Chenu, D. and Quintard, M. *Simulation of Transport and Impact of Moisture Content on the Biodegradation*. In: HPM4 2011, 27-28 Apr 2011, Santander, Spain.

SIMULATION OF TRANSPORT AND IMPACT OF MOISTURE CONTENT ON THE BIODEGRADATION

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SUMMARY: A large scale averaging method is used to study and simulate the influence of water on the solid waste biodegradation kinetics in landfill context. The basic process of interest is a two-phase flow transport model in homogeneous porous medium coupled with a hydrolysis biological reaction. The hydrolysis kinetic is weighted by a Heaviside function which depends on the local saturation level linked to the global moisture content. The shape of the large scale weighting function is in correlation with the experimental linear relationship described in literature for many solid wastes.

1. INTRODUCTION

In Landfills, water content is a major ecological parameter of the anaerobic biodegradation of the solid waste into biogas (Farquhar and Rovers, 1973; Rees, 1980; Kasali et al., 1990; Gurijala and Sufliata, 1993). The value of water content under which no biodegradation activity is observed ranged from 0.15 to 0.5 expressed as g water/g dried waste (Buivid et al., 1981; Barlaz et al., 1990; Yang and Chang, 1998; Rodriguez et al., 2001). In practice, the fresh refuse

moisture ranges from 0.25 to 0.65 (Owens and Chynoweth, 1993; Aguilar et al., 1999; Barina et al., 2003; Lefebvre et al., 2003). Therefore, in a standard situation, biological reactions in landfills may be strongly water limited. Besides, biodegradation improvement by increasing the waste water with leachate injection has been widely demonstrated (Barlaz et al., 1990; Metha et al., 2002, Benbelkacem et al., 2010). In the landfill context, the cellulosic materials in paper or cardboard form are mainly concerned since initially very deficient water and concentrating the major part of the biodegradable organic matter.

The optimisation of the leachate recirculation monitoring need a good description of both the water transport inside such porous system and the water mediated biological reactions. At the scale of the landfill, the water transport are generally considered in the macro-porosity defined by the space between the waste particles (Rosqvist and Destouni, 2000; Hartmann et al, 2004; Rosqvist et al., 2005). However, the biological reaction scale is mainly at the surface and above all inside the organic particles that are micro-porous (especially paper, cardboard). Also, the water transport inside an organic particle needs to be introduced.

At the laboratory experimental observation scale ($L \approx 10\text{cm}$), a linear dependency of both the biodegradation kinetic rates and the amount of biodegraded organic waste with the global water content is generally proposed. This linear correlation is defined between a minimal moisture content and an optimal one (Bogner, 1990; Aran et al., 1999; Pommier et al., 2007). These observations consolidate the assumption of a waste activation controlled by the local moisture content: the solid needs to be wet enough to be available for biological reactions. At larger scale, this concept of activation has been modeled by a linear factor describing the waste bioavailability and applied to the amount of solid matter accessible for biodegradation, as reported by Pommier et al. (2007). At local scale this activation concept amounts to segregating the solid matter into active matter and inactive matter, the state of a local elementary volume being governed by its moisture content (or its saturation level). For that, a critical moisture content could be defined, below which the solid matter is considered as inactive, and above which the solid matter is considered as active.

In this paper, transport and biodegradation reaction at the particle dimension are focused. A cotton paper strip is used as cellulosic matter. Mechanisms are modelled at the local-scale (Darcy's scale) both for the unsaturated flow and water transfer into a waste particle and for the biological hydrolysis reaction. Results of simulations are discussed and compared to the global experimental relationship between water content and biodegradation yield, proposed in literature.

2. THEORETICAL ELEMENTS

For an effective medium, the flow is considered governed at low Reynolds values by a Darcy's equation. For an unsaturated flow, two main methods are usually used: two Darcy's equations for liquid and gas phase and the Richards's equation. The first method has often been discussed in the literature (Withaker, 1986; Auriault, 1987; Lasseux et al., 1996; Hassanizadeh and Gray, 1997; Hilfer, 1998; Wang, 2000). Here, the second approach Richards equation is preferred because the processes are slow enough to neglect pressure gradients in the gas phase (γ -phase). Assuming Darcy's law is valid, Richards equations can be used to describe the water flow in unsaturated porous medium (Richards, 1931). This assumption has been developed in Chenu, 2007, and the validity of Richards equation is discussed herein. Therefore, for a single porosity domain, the water transport equation is given by the following equation (1)

$$\varepsilon \partial_t S_\beta + \nabla \cdot \left[-\frac{\mathbf{K} k_{r\beta}(S_\beta)}{\mu_\beta} \rho_\beta \cdot \nabla \left(\frac{p_c(S_\beta)}{\rho_\beta} + gz \right) \right] = 0, \quad (1)$$

where S_β is the local water saturation and ε the total porosity of the medium. In eqn (1), \mathbf{K} represents Darcy’s law permeability tensor (Whitaker, 1986), $k_{r\beta}$ is the classic relative permeability for the water phase (β -phase) and g is gravity. To describe $p_c(S_\beta)$, different models exist. In this work, the capillary pressure $p_c(S_\beta) = p_\beta - p_\gamma$ is linked with S_β and $k_{r\beta}$ through the classic Van Genuchten Relationship [Eqns (2), (3)],

$$Se_\beta = \frac{S_\beta - S_{ir}}{S_{sat} - S_{ir}} = \left[1 + (\alpha |p_c|)^n \right]^{-m}, \quad p_c < 0, \quad (2a)$$

$$Se_\beta = 1, \quad p_c \geq 0, \quad (2b)$$

$$k_{r\beta} = Se_\beta^l \left[1 - (1 - Se_\beta^{1/m})^2 \right], \quad p_c < 0, \quad (3a)$$

$$k_{r\beta} = 1, \quad p_c \geq 0, \quad (3b)$$

where Se_β is the local effective water saturation, S_{sat} is the saturated saturation, S_{ir} is the irreducible saturation, α is the inverse of a specific pressure, n is the pore size distribution index which determines the slope of the retention curve, $m = 1 - 1/n$, l is the tortuosity parameter. In equation (1), the term, $-\nabla p_c(S_\beta)/\rho_\beta$ represent the capillary mass force tending to produce a motion of water in the porous medium. The velocity and acceleration of the capillary water are everywhere zero when the capillary mass force just balances gravity. This configuration is called capillary equilibrium. As the capillary equilibrium is not reached, there is a resultant water-moving force and then a capillary flow.

The cellulose anaerobic degradation is known to be controlled by its hydrolysis into monomers. Such a biological mechanism is commonly described by a first order reactive scheme relative to cellulose content. Also, hydrolysis reaction at the local-scale is given by Eqn (4),

$$\partial_t Xs(t) = -k_h h(S_\beta(\mathbf{x}, t)) Xs(t), \quad (4)$$

where Xs is the quantity of hydrolysable matter present in the medium and k_h is the hydrolysis constant. The first order kinetics is weighted by a Heaviside function $h = h(S_\beta(\mathbf{x}, t))$ with threshold depending of a local specific activation saturation, S_{act} (Figure 1).

Insert Figure 1

Figure 1. Weighting function $h = h(S_\beta(\mathbf{x}, t))$ of the hydrolysis reaction. The weighting function is equal to zero for $S_\beta < S_{act}$ and equal to 1 for $S_\beta > S_{act}$.

The matrix zone where the local saturation is greater than the local activation saturation is

referred as activated volume since the hydrolysis reaction can start while it is completely bypassed elsewhere.

In the paper, the smallest pores are often less than $0.1\mu m$ and larger pores may be of the order of $1mm$ according the manufacturing process. The smallest bacteria have a size of 0.1 to $0.2\mu m$ and the larger ones can reach a size of $0.75mm$. In this work, the transport of bacteria is considered totally coupled with the water transport and the sizes of meso-pores and micro-pores are not an obstacle for the migration of bacteria. It means that bacteria are transported in the water phase without any coupling with viscosity and density. It is considered as dilute species.

The local specific activation saturation depends upon both the physicochemical properties of the porous medium and to the micro-organisms growth capacities in such limited water environment. At this point, it is more convenient to describe what is called by local-scale and large-scale. Local-scale is a sufficient number of micro-pores and meso-pores so that Darcy’s law is valid [Figure 2 (b)]. From a reactive point of view, since the reaction rate is closely related to the water content, the radius of the volume characterizing the local-scale should be large enough in relation to the variations of the water content at the pore-scale. Large-scale is a volume containing a large number of volumes characterizing the locale-scale and can be represent an individual particle of waste of volume V_{∞} [Figure 2 (a)].

Insert Figure 2

Figure 2. (a) Particle of waste, slip of paper (large-scale). (b) Large number of meso and micro-pores of the paper (local-scale). (c) Micro-porosity, primary fibers of the paper. Photos: Coste, G., INP-Pagora, Grenoble, <http://cerig.efpg.inpg.fr>, 02/2011.

Following this description, it is possible to introduce the large-scale spatial average of a local-scale generic function $\psi(\mathbf{x}, t)$ by

$$\psi^*(t) = \frac{1}{V_{\infty}} \int_{V_{\infty}} \psi(\mathbf{x}, t) dV . \quad (5)$$

The large-scale spatial average of the hydrolysable matter and the effective weighting function (Xs^* and h^*) including the effect of saturation heterogeneities on a volume corresponding to the volume V_{∞} , shown in Figure 2(a), are sought.

The large-scale spatial average of saturation is related to the global water content by applying Eqn (6).

$$w^* = \frac{1000}{\rho_{DM}} \varepsilon S_{\beta}^* \quad (6)$$

Then, the value of the large-scale spatial average of weighting function can be expressed in terms of the global water content for the water content initially injected into V_{∞} [Figure 2(d)]. For each initial amount of water injected into V_{∞} , this approach is repeated to obtain the shape of the large-scale spatial average of the weighting function depending on the global water content of V_{∞} [Figure 2(d)].

3. BIOCHEMICAL AND PHYSICAL PROPERTIES FOR SIMULATION

The system taken into consideration is a cellulosic strip whose dimensions are $1\text{cm} \times 5\text{cm} \times 0.7\text{cm}$ see pictures below. This paper was manufactured from cotton without additives. The biochemical and physical properties used for simulations are given in **Erreur ! Source du renvoi introuvable.**

Insert Picture 1

Picture 1. Cotton paper strip of dimension $1\text{cm} \times 5\text{cm} \times 0.7\text{cm}$.

In such matrix, the micro-pores are formed by the entanglement of the primary fibers [Figure 2 (c)] while the meso-pores result from the manufacturing process or from mechanical stresses undergone during the life of the waste [Figure 2 (b), Picture 1]. As described previously, the water transport dynamics inside the porous solid are controlled both by the permeability K and, especially, by the capillary properties. The Darcy’s law permeability tensor is considered as isotropic [Eqn (7)]

$$\mathbf{K} = K \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad (7)$$

where K is the absolute permeability. This is an assumption which could be discussed, but our samples do not show any anisotropic behaviour. They seem to be homogeneous as there is no preferential orientation. Given the wide variety of existing paper, it is not easy to define a value for K . The values listed in literature range from $1 \times 10^{-12} \text{m}^2$ to $1 \times 10^{-15} \text{m}^2$ depending of the variety of paper (Lindsay, 1988; Nilsson and Stenström, 1997). In Lindsay (1988), transverse permeabilities are measured for different linearboard handsheets compressed to porosities between 0.6 and 0.9. Permeability values are reported as a function of the porosity through an exponential function. For sheets with a porosity between 0.85 and 0.9, the value of transverse permeability is of the order of $K = 1 \times 10^{-12} \text{m}^2$. Although the studied paper is really different, simulations were made with a value of $K = 1 \times 10^{-12} \text{m}^2$.

The capillary properties of this paper have been approximated by an inverse numerical method from the characteristic scale of the capillary fringe observed during the imbibition experiments. The capillary curve obtained by this method is given in Figure 3.

Insert Figure 3

Figure 3. Imbibition curve of the cotton paper.

The irreducible saturation S_{ir} is considered as immobile water quantity highly bounded to the cellulose fibers. Here, a value is deduced from the measurement of the sorption isotherm curve given in the Figure 4. Experimental data are compared to literature data (Parker et al., 2006). Similar profiles are obtained whatever the kind of paper or cardboard. The irreducible local-scale water content, w_{ir} is supposed to be near the water content when the water activity a_w is equal to 1. For our cellulose matter, the value is measured at 0.28 g/g. S_{ir} is also calculated by the relationship between moisture content and saturation Eqn (8).

$$S_{ir} = \frac{w_{ir}\rho_{DM}}{\varepsilon}, \quad (8)$$

where ρ_{DM} is the density of the dried matter (expressed in $g \cdot cm^{-3}$) and ε its porosity.

Finally, in biological reaction point of view, value for the S_{act} parameter is necessary for simulation. No experimental values are available. Indeed, the local reaction scale is very smaller than the scale that can be directly and experimentally observed. Consequently, the value of S_{act} used here for simulations was chosen to be consistent with the experimental results obtained at the large scale. S_{act} is fixed at 0.22 which corresponds to a local-scale water content of 0.95 g water / g DM.

Insert Figure 4

Figure 4. Sorption isotherm curve of the cotton paper comparison to literature data (from Parker et al., 2006).

Table 1. Experimental data for model parameters

Experimental data	Unit	Value
Water content, w (1)	%, g_water / g_DM	3.5
Organic matter (1)	%, DM	99.00
Mineral matter (1)	%, DM	1.00
COD / DM (1)	g / g	1.09
BMP (2)	NmL CH4 / g_COD	321
Biodegradability (2)	%, DCO	92
Kinetic rate, k_h (2)	s^{-1}	8.10^{-7}
Porosity, \square (3)	-	0.85
Absolute permeability, K	m^2	1×10^{-12}
Density of dried matter, ρ_{DM} (1)	$g.cm^{-3}$	0.197
w_{ir}	g_water / g_DM	0.28
S_{ir}	-	0.065
α	Pa^{-1}	4.5×10^{-3}
l	-	0.5
m	-	0.6
n	-	2.5
S_{act}	-	0.22

(1) Standard methods (2) according Pommier et al. (2007) (3) Imbibition method

4. NUMERICAL RESULTS

For the numerical simulations, a 2D cotton paper strip is considered. The dimensions are $1cm \times 5cm$. This strip is submitted to gravitational field. Starting from water saturation near the irreducible saturation [Figure 5(a)], a defined water amount is injected into the medium [Figure 5(b)]. The capillary equilibrium is reached by solving Eqn (1) [Figure 5(c)] and is achieved in short time ($\approx 10d$) compared to the biological degradation time of the hydrolysable matter (100d). The water spatial distribution dynamic can be considered as quasi decoupled from the biodegradation process kinetics and the local-scale fields obtained [Figure 5(d)] can be used

to obtain the large-scale spatial averages fields with Eqn (5).

Insert Figure 5

Figure 5. (a) Initial local water saturation field. (b) Local water saturation field after an injection of water of 5h 33mn. (c) Local water saturation field after capillary equilibrium. (d) Hydrolyzed quantity of hydrolysable matter after 100 days (in %).

Then, successive similar water additions are simulated up to reach the value of S_{sat} of the cotton paper strip. For each w^* , the simulation gives a value for h^* and $1 - Xs^*/Xs_{ini}^*$ which is the large-scale spatial average of the hydrolyzed quantity of hydrolysable matter. In Figure 6, h^* and $1 - Xs^*/Xs_{ini}^*$ are plotted versus w^* .

Insert Figure 6

Figure 6. Large-scale spatial average of the weighting function of the hydrolysis reaction, h^* , and large-scale spatial average of the hydrolyzed quantity of hydrolysable matter, $1 - Xs^*/Xs_{ini}^*$, in terms of global water content w^* in a strip of paper measuring $1cm \times 5cm$. Normalized maximum biogas production for paper/cardboard (Pommier et al., 2007) $CH_{4 \max}/CH_{4 \text{opt}}$, in terms of global water content w^* .

Starting at the local scale with the local-scale weighting function, h , the large-scale spatial average of the weighting function, h^* , and the large-scale spatial average of the hydrolysed matter, $1 - Xs^*/Xs_{ini}^*$, are directly calculated from Eqn (5). The shapes of h^* and $1 - Xs^*/Xs_{ini}^*$ are in good agreement with the experimental relationship between water content and biodegradation yield, listed in literature. The analogy may, for example, be made by comparing $1 - Xs^*/Xs_{ini}^*$ to the normalized maximum biogas production for paper/cardboard, $CH_{4 \max}/CH_{4 \text{opt}}$, obtained by Pommier et al., (2007). It appears that these relationships are not completely linear because of capillary effects, which are not either. In Figure 6, h^* is different from $1 - Xs^*/Xs_{ini}^*$ for values of w^* between 0 and 1 because, for these values, the capillary equilibrium time become not negligible compared to the biological degradation time of the hydrolysable matter. A relatively large amount of matter is then hydrolysed during the capillary equilibrium time in the area where $S_{\beta} > S_{sat}$ and $1 - Xs^*/Xs_{ini}^*$ is decoupled from h^* . The capillary equilibrium time mostly depend of the absolute permeability of the medium and pilot the coupling or decoupling between the water spatial distribution dynamic and the biodegradation process kinetics.

Moreover, a sensitivity analysis on different parameters show that the dominant parameters on the shape of h^* and $1 - Xs^*/Xs_{ini}^*$ are the Van Genuchten parameters, l, m, α , and the local-scale activation water saturation, S_{sat} . For a fixed spatial characteristic scale of the paper strip, Van Genuchten parameters control the slope of the curve, while the local-scale activation water saturation controls the position of the curve on the global water content axis, that is to say, the values of w_{\min}^* and w_{opt}^* .

From this model, it is possible to simulate the evolution of a cellulose particle biodegradation with time in function of its global water content w^* . The curves in Figure 7 have similar profiles

corresponding to a first order reaction but are different from the maximal amount of degraded substrates.

Insert Figure 7

Figure 7. Simulated hydrolysable matter production kinetics at different water contents.

This points out the relevance of the model developed and especially the mechanisms considered. It is possible to take into account the physical properties of the porous matrix, the heterogeneity of the water inside this matrix and consequently the local bio-accessibility of cellulose substrate. One of the critical parameter is the value of S_{act} . No data are available, because the experimental scale is always larger than the reaction scale. Here, an inverse method was adopted. Indeed the value was approximated from the value of w_{min}^* and w_{opt}^* measured at the experimental large-scale.

The model shows also the importance of the solid dimension L on the slope of the relationship at the large-scale. Figure 8 gives an example for two cellulose strips whose L are 1cm and 5cm . As the capillary fringe is large compared to L , the heterogeneities of saturation are reduced into the cellulose and the profile of h^* comes closer to a Heaviside function local form. In contrast, when L is large enough compared to the capillary fringe, the profile of h^* is independent of L . However, whatever the dimension L , all the curves $h^* = f(w^*)$ have a same and unique intersection point corresponding to the activation saturation S_{act} . From two dimensions L , it should be possible to obtain an experimental estimation of S_{act} by the identification of the intersection point.

Insert Figure 8

Figure 8. Large-scale spatial average of the weighting function of the hydrolysis reaction, h^* , in terms of global water content of the strip of paper, w^* , for strips of paper with a spatial characteristic scale of 1cm and 5cm .

5. CONCLUSIONS

The relationship between the local water content and the biological activity is modelled and simulated at the particles scale. The model couples two-phase flow and reactive transport. The reaction rate is controlled by the water saturation at each scale. The main assumption is that, at the local-scale, the reaction is weighted by a Heaviside function which depends on the local saturation level.

The determination of the limitation law for hydrolysis reactions at the large-scale, have showed that it depends on several parameters: capillary properties, S_{act} and transport properties. All these parameters are identifiable from experimental measurements.

If a general law is used in landfills biodegradation, this reaction scheme and the limitation could be estimated using the approach developed in this paper. Simulations are in good agreement with the experimental data. The model succeeds to predict the impact of water content on waste anaerobic biodegradation at the particle scale. The effects of scale and especially the downscaling to investigate the local scale effects (i.e dimensions less than 1cm) will be made in

a forthcoming paper in order to conclude on the result showed in Figure 8.

ACKNOWLEDGEMENTS

This work was supported by the French National Research Agency (ANR) and the French National Agency for Energy and Environment (ADEME). The Authors wish to thank Mr. Gérard Coste from INP-Pagora Grenoble (France) for photos of paper obtained by electron microscopy.

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NOMENCLATURE

a_w	water activity
DM	dry matter
g	gravity, $m \cdot s^{-2}$
X_s	hydrolyzable matter, % DM

X_s^*	large-scale spatial average of hydrolyzable matter, % <i>DM</i>
h	local-scale weighting function of the hydrolysis reaction
h^*	large-scale spatial average of the weighting function of the hydrolysis reaction
K	Darcy's law permeability tensor, m^2
K	absolute permeability, m^2
k_h	local-scale hydrolysis constant, s^{-1}
$k_{r\beta}$	relative permeability for the water phase
L	Spatial characteristic scale of the strip of paper, m
l	tortuosity parameter
n	pore size distribution index
p_c	local-scale capillary pressure, Pa
p_β	local-scale water pressure, Pa
p_γ	local-scale gas pressure, Pa
S_{act}	local-scale activation water saturation
S_{ir}	irreducible saturation
S_{sat}	maximal saturation
Se_β	local-scale effective water saturation
S_β	local-scale water saturation
S_β^*	large-scale spatial average of water saturation
V_∞	large-scale averaging volume, m^3
w	local-scale water content, $g\ water / g\ DM$
w^*	global water content, $g\ water / g\ DM$
w_{ir}	irreducible locale-scale water content, $g\ water / g\ DM$
w_{min}^*	large-scale minimum water content, $g\ water / g\ DM$
w_{opt}^*	large-scale optimum water content, $g\ water / g\ DM$
z	height from reference level, m
α	Van Genuchten parameter, Pa^{-1}
ε	total porosity
μ_β	dynamic viscosity of water, $kg \cdot m^{-1} \cdot s^{-1}$
ρ_{DM}	density of dried matter, $kg \cdot m^{-3}$
ρ_β	density of water,