Combustion within Porous Waste

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1 Introduction

Flammable gases (primarily hydrogen and nitrous oxide but also ammonia and methane) are continously being generated within the waste contained in the tank farms at Hanford Site. Some portions of the waste are porous and conceivably, a combustion event could occur within the waste due to accidental ignition. This has been postulated as a potential hazard since deflagrations and detonations are observed in laboratory experiments to propagate through combustible gases in porous materials, or through interconnected flammable gas voids. The waste in Hanford storage tanks are mainly in three different forms, a: salt cake, b: sludge, c: supernatant. Formation of a crust layer on the top of the waste is also observed in some tanks. The salt cake waste and crust resemble porous materials while sludge and supernatant looks like highly viscous fluids retaining flammable gas as bubbles or inclusions. Although laboratory experiments showed the possibility of propagation of deflagration or detonation in waste-like porous materials filled by flammable gases, the relevance of this issue to safety evaluations at Hanford is a matter of contention.

In order to clarify this issue, we have reviewed the relevant data on laboratory experiments related to combustion in porous material. In doing this, we have concentrated on the flame literature rather than the detonation literature, since Makris et al. (1995) have already examined that. Further, significant mechanisms for the initiation of detonation (i.e., geometries resulting in strong flame acceleration within the dome) have not been identified therefore making flames a much more likely outcome of accidental ignition than detonation. Ignition of flammable waste gases in the waste or the dome space of a tank can occur during intrusive operations into the waste or dome. External events which are not foreseeable such as lightning can also ignite the flammable gas retained in the waste. The present report only examines the basic issues in propagation of deflagration or detonation within waste. The process or probability of combustion ignition and other combustion events such as burns in the dome are not considered.

After our review of the literature, some simple estimates of the potential for flame and detonation propagation are given. We conclude with a discussion of the uncertainties and measurements required to resolve this issue.

2 Literature Survey

In gases, there generally are two possible modes of combustion: a low-speed (1 to 100 m/s) flame or deflagration and a high-speed (1500 to 3000 m/s) detonation. In porous media, the same categories are used but the distinctions are blurred.

2.1 Detonation

Detonations can directly be initiated (through a jet of hot combustion products or through very high ignition energies) or a transition of a deflagration to a detonation can occur. Detonations propagate at supersonic velocities within detonable gas mixtures and can also propagate at high velocities in porous materials (Lyamin and Pinaev 1985). An upper bound for the propagation velocities can be calculated by using the Chapman-Jouget model (CJ) model of a gaseous detonation. The sensitivity of an explosive gas to detonation strongly depends on gas-composition, thermodynamic conditions and possible dilution and can be estimated by using detailed chemical kineticWestbrook and Urtiew (1982) schemes for gases. The influence of mixture sensitivity and pore size on propagation of gaseous detonations in porous media are discussed by Makris (1993) and Makris et al. (1995).

2.2 Deflagration

More likely than a detonation initiation in the tanks of Hanford Site is the initiation of a deflagration. Gas deflagrations propagate at much lower subsonic velocities. The heat transfer from the combustion zone to the porous medium itself is very effective because of the very large surface between them. Ceramic pebbles, ceramic foams (Alvin 1996; Howell et al. 1996) or metal foils have been used as porous media in laboratory studies. The porosity ε (void fraction) of the porous medium (PM) is defined as:

$$\varepsilon = \frac{volume\ of\ pores}{total\ volume}\ . \tag{1}$$

Typical values for an uncompacted bead or pebble bed are $0.36 < \varepsilon < 0.45$ independent of pebble diameter δ. Smaller porosities are found in consolidated or compacted materials. The porosity of the waste in the Hanford tanks is unknown and varies within the waste in a tank and between wastes stored in different tanks. The specific surface a per unit volume of the inner PM depends on the particle size, morphology and packing fraction. For example, in a packed bed formed of spheres with a diameter δ:

$$a = \frac{6(1-\varepsilon)}{\delta}\ . \tag{2}$$

The hydraulic diameter d_h of the pores is for a bed of packed spheres:

$$d_h = \frac{\varepsilon\ \delta}{3(1-\varepsilon)}\ . \tag{3}$$

The waste stored in underground storage tanks consists of salt crystals. The size and shape of these crystals vary considerably depending on the specific type of waste and also spatially within a tank. Based on the range of sizes observed in waste samples taken from tank SY-101, the characteristic dimension of crystals is on the order of 10-100 μm. Representing these salt crystals with a spherical geometry is an oversimplification. In the absence of real data, this assumption, perhaps, is the only choice that we have to address the combustion of flammable gases in the waste. The alternative to this choice is performing experiments with real or simulated wastes.

Flame propagation through porous media has been investigated by numerous researchers. Korzhavin et al. (1982), Kauffman et al. (1982), Lyamin and Pinaev (1986), Lyamin and Pinaev (1987), Pinaev and Lyamin (1989), Babkin et al. (1991) and Korzhavin et al. (1997) did mainly experimental work, while Sathe et al. (1990), Yoshida et al. (1990), Tong and Sathe (1991), Hanamura and Echigo (1991), Hanamura et al. (1993), Hsu et al. (1993) and Aldushin et al. (1996) performed numerical studies and tried to model the porous medium combustion.

The experimental work was performed in tubes filled with porous material and the homogeneous fuel-oxidizer mixture. The propagation of the flame front, temperature and pressure were recorded at several places in the tube after ignition at one end of the tube. The main result of Babkin et al. (1991) was that five steady combustion regimes (steady propagation velocity of the flame front through the tube after a short entrance length) exist for combustion within porous material with different flame propagation mechanisms. An intermediate rapid combustion regime (RCR) between high velocity regime (HVR) and sound velocity regime (SVR) was identified by Lyamin and Pinaev (1986), giving the six regimes presented in Table 1.

Regarding propagation of deflagration through the porous waste in the Hanford tanks the HVR and RCR are of greatest interest. The following flame propagation mechanism for these regions was proposed by Babkin et al. (1991):

“A positive feedback between flame acceleration and turbulence production (the flame acceleration causes turbulence, which leads to flame acceleration) is damped by local quenching of the chemical reaction due to intense heat exchange in the turbulent flame zone. If the characteristic time of thermal relaxation becomes less than that of chemical conversion the flame will be quenched. Since turbulent flow contains a spectrum of instantaneous gas velocities, those parts of the flame moving with the maximum velocities will be quenched, resulting in a stable velocity of flame propagation”.

It must be noted that the frictional resistance in the porous material may be greater by several orders of magnitude than that in smooth or rough pipes (Dullien 1979; Shepherd and Begeal 1988; Abbott 1998). Regardless of such high resistance and heat losses, steady

Table 1: Flame propagation regimes in porous media (Babkin et al. 1991; Lyamin and Pinaev 1986).

Regime	Wave velocity [m/s]	Flame propagation mechanism
Low velocities (LVR)	0 - 10^{-4}	heat conductivity, interphase heat exchange
High velocities (HVR)	0.1 - 10	convective, uniform pressure
Rapid combustion (RCR)	10 - 100	convective, smooth pressure gradient
Sound velocities (SVR)	100 - 300	convective pressure gradient
Low velocity detonation (LVD)	500 - 1000	self-ignition under shock wave interaction
Normal detonation (ND)	1500 - 2000	detonation under heat and pulse losses

combustion and detonation regimes are proven to be possible.

The transition point between flame-propagation and -quenching is very often given by a critical Peclet number Pe^*, where:

$$Pe = \frac{u\ d}{\kappa} \qquad \text{(tube)}, \tag{4}$$

$$Pe = \frac{S_L\ d_m\ c_p\ \rho}{k} \qquad \text{(porous material)}, \tag{5}$$

and u is the flow or flame velocity (depends on the experiment), d the tube diameter, κ the thermal diffusivity, and S_L is the laminar burning velocity, d_m the equivalent porous cavity diameter, c_p the specific heat capacity, ρ the density, and k the heat conductivity of the unburned gas mixture. A limit generally accepted for flame propagation through a tube is $Pe^* = 85$ for the minimum value or minimum tube diameter (Lyamin and Pinaev 1985; Babkin and Laevskii 1987), whereas for propagation through porous material also smaller values were measured: $Pe^* = 65$ (Trimis and Durst 1996).

3 Scaling Criteria

Using the existing data developed on idealized materials, it is possible to make a preliminary evaluation of the potential for flame and detonation propagation in the Hanford waste. It is important to keep in mind that the previous studies have the following features:

1. simple porous materials, usually beds or sintered compacts
2. simple fuel-air mixtures (usually hydrocarbon-air mixtures)
3. dry (water-free)

The situation at the Hanford Tank Farm is very different than in the typical idealized experimental studies. The most significant differences are:

1. the porous waste material is very complex and probably quite dissimilar from the simple artificial beds of beads or uniform particles used in the previous lab studies
2. the combustible gases will be primarily a mixture of hydrogen, nitrous oxide and nitrogen
3. the waste contains a substantial amount of water
4. size and shape of salt crystals are not uniform within the waste
5. gas bubbles (therefore gap size) may not be distributed uniformly
6. existence of water may prevent the formation of interconnected gas networks or cause isolation of bubbles during deflagration

Other differences of secondary importance are:

1. there is heat generation within the waste

2. there is dissolved gas in the liquid phase
3. the conditions vary throughout a tank

3.1 Detonation

It is found that a detonation can transmit into a inert porous medium filled with detonable gas and continue to propagate as a quasi-steady combustion wave (Makris 1993). There exists a continuous spectrum of averaged combustion wave velocities ($0.3 \leq V/V_{CJ} \leq 1$, where V is the wave speed in the porous material and V_{CJ} is the Chapman-Jouquet detonation velocity) spanning the lean and rich propagation limits and exhibiting a maximum value at the most sensitive composition. A decrease in the pore size results in narrowing the detonability range and reducing the propagation velocity for a given mixture. The effect of a reduction in pore size is a decrease in the dimension of the openings and the linear separation between obstacles. This is equivalent to a closer spacing of obstacles in rough tubes or a decrease in the tube diameter. The controlled variation of the pore size for several different fuels at $\phi = 1.0$ indicates that V/V_{CJ} correlates with d_c/d_p: $V/V_{CJ} = [1 - 0.35 log(d_c/d_p)] \pm 0.1$ (d_c: critical tube diameter; d_p: average pore diameter defined as (spherical particle diameter)/3). When $d_c/d_p \leq 1$ the detonation suffers no deficit and propagates at its C-J velocity across the porous medium, whereas for $d_c/d_p > 100$ the combustion wave is quenched (Makris et al. 1995). In all cases the velocity at the limits is $V/V_{CJ} \approx 0.3$. The near limit phenomenon appears to be consistent with a convective ignition transfer mechanism in the pores. At the limits, turbulent quenching seems to be responsible for the extinction of combustion within a short distance of penetration in the porous medium.

The critical tube diameter d_c roughly corresponds with the cell width λ for detonation quenching within porous material:

$$d_c \approx 13\lambda , \tag{6}$$

$$\text{with} \quad \frac{d_c}{d_p} > 100 \quad \text{for quenching,} \tag{7}$$

$$=> \lambda > 7.7 d_p \quad \text{for quenching.} \tag{8}$$

Akbar et al. (1997) measured the cell widths for mixtures approximating the retained gas in six tanks. The results are shown in Figs. 1- 6. They obtained a cell width for the retained gas mixture in tank SY-101 of 20-30 mm. Measurements for the other tanks were obtained only for mixtures diluted with air but extrapolations were made with the ZND model to zero dilution. These extrapolations, shown in Figs. 4, 5 and 6, indicate that there is too little N_2O in AW-101, AN-103 or A-101 for a detonation to occur in the undiluted retained gas. In AN-104, the cell width of the retained gas mixture is about 80 mm and in AN-105, 4000 mm. All of these mixtures have a shallow minimum in predicted cell width between 40 and 60% air dilution, with minimum values of the cell width between 10 and 100 mm. The cell width rises sharply with increasing dilution beyond the minimum cell width point. Air dilution results in slightly smaller cell widths than N_2 dilution (Akbar et al. 1997).

With the equations given above (6, 7 and 8) and the cell widths λ for the representative retained gas compositions, one can calculate the effective spherical particle diameter δ for quenching:

$$\lambda \;=\; 10 - 100\; mm \;>\; 7.7 d_p \qquad \text{for quenching,} \tag{9}$$

$$d_p \;\approx\; \frac{\delta}{3}\,, \tag{10}$$

$$=>\; \delta \;<\; 3.9 - 39\; mm \qquad \text{for quenching.} \tag{11}$$

The effective particle diameter of a salt simulant based on the waste in tank SY-101 is approximately 100 μm (see section 3.2), which means, a detonation initiated in the dome of tank SY-101 would not propagate within the porous waste. A statement for other tanks, such as AN-105 and AN-104, can not be made, because no permeability-measurements were carried out and therefore the effective particle diameter of the porous waste in these tanks is not known.

The validity of Eq. 10 has not been demonstrated for waste-like materials and depends on the mechanism of the gas retention. If the gas retention is in the pores of an idealized porous structure, the use of Eq. 10 or similar relations is considered to be reasonable. However, since the ratio of the pore to particle size depends upon the shape of crystals, it may be possible to have propagation with irregular shaped particles under conditions that would not result in propagation with spherical particles.

The flammable gas will be retained in several different ways depending upon the properties of the waste. Gauglitz et al. (1996) indicates that dendritic bubbles can be formed as a result of displacement of solids. The characteristic dimension of the dendritic bubbles may exceed the typical particle sizes ($d_p \;>\; \delta$) and would be on the order of a few millimeters (Gauglitz et al. 1996). In addition, Abbott (1998) observed that waste simulants would crack during drainage of liquid and form channels. The existence of dendritic bubbles or cracks in the waste indicates that scaling arguments based on spherical particles are not completely reliable guides and detonation propagation cannot be ruled out definitively on this basis.

However, as discussed in the safety assessment of mixer pump operations in tank SY-101, the possibility of detonation in the dome space is unlikely. This is due to the

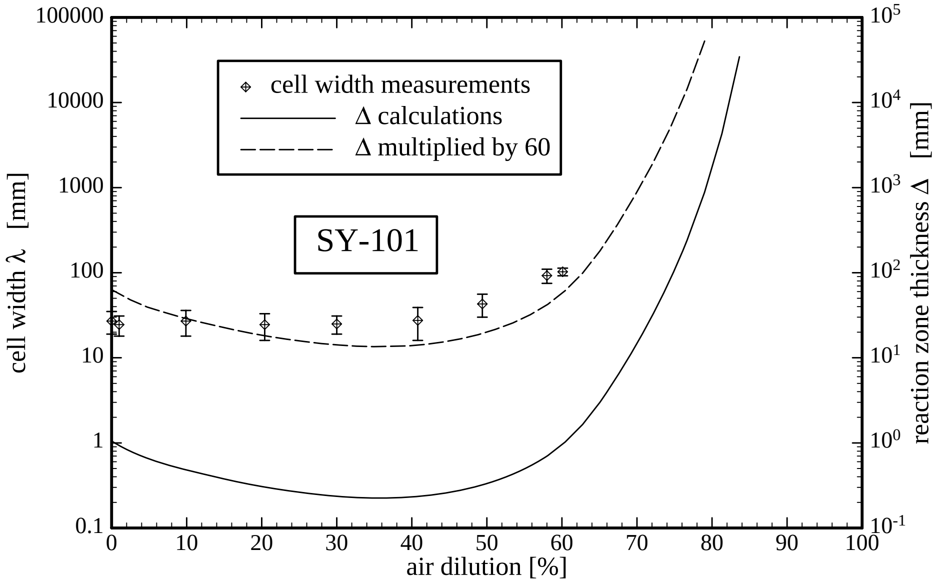

Figure 1: Measured cell width and calculated reaction zone thickness for approximate SY-101 mixtures: 29% H_2 + 24% N_2O + 35% N_2 + 11% NH_3 + 1% CH_4 (Akbar et al. 1997).

large values (greater than 100 mm) of the detonation cell width that results when the gas release is mixed with the atmosphere within the dome. This assessment is based on detonation cell sizes measured by Akbar et al. (1997), which are shown in Figs. 1-6.

The detonation cell width λ measurements were made over a limited range and then extrapolated using the ZND model to compute the reaction zone thickness Δ from a detailed chemical reaction mechanism as discussed in Akbar et al. (1997). The extrapolation is based on a simple proportionality $\lambda = A\Delta$; a value of $A = 60$ was used for all cases. While this gives a reasonable match for all of these cases, it is known that λ can have a complex dependence on the reaction zone thickness and the extrapolated values are only rough estimates. The very large (greater than 10 m!) cell widths estimated for retained gas in tanks A-101, AN-103 and AW-101 make detonation initiation and propagation effectively impossible for these mixtures. On the other hand, addition of about 50% air will make the detonation sensitivity comparable to the SY-101 mixture without dilution. The undiluted retained gas in Tank AN-105 has a cell width of about 4 m, making this mixture effectively impossible to detonate, irrespective of the porous medium. Only in tanks SY-101 and AN-104 is detonation propagation conceivable in the undiluted retained gas.

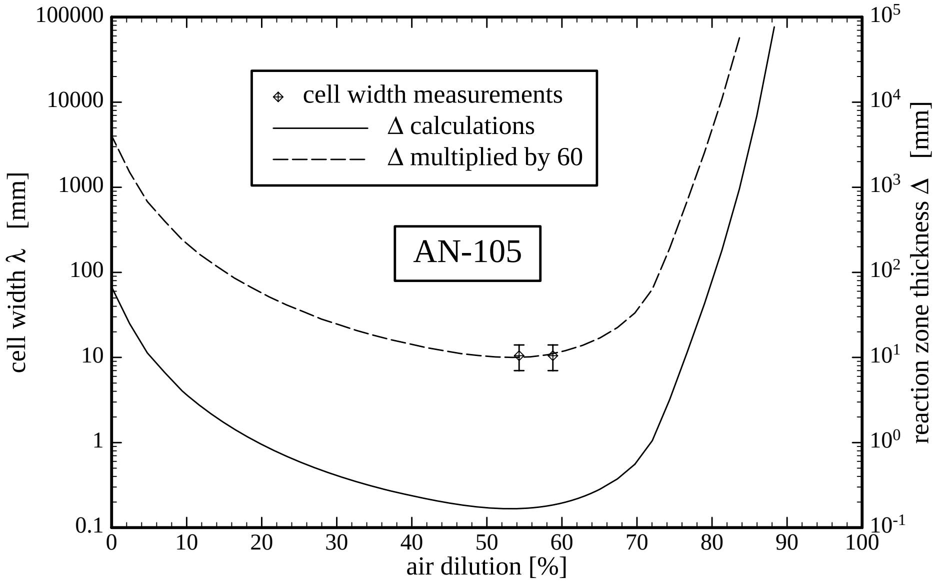

Figure 2: Measured cell width and calculated reaction zone thickness for approximate AN-105 mixtures: 63% H_2 + 11% N_2O + 25.28% N_2 + 0.02% NH_3 + 0.7% CH_4 (Akbar et al. 1997).

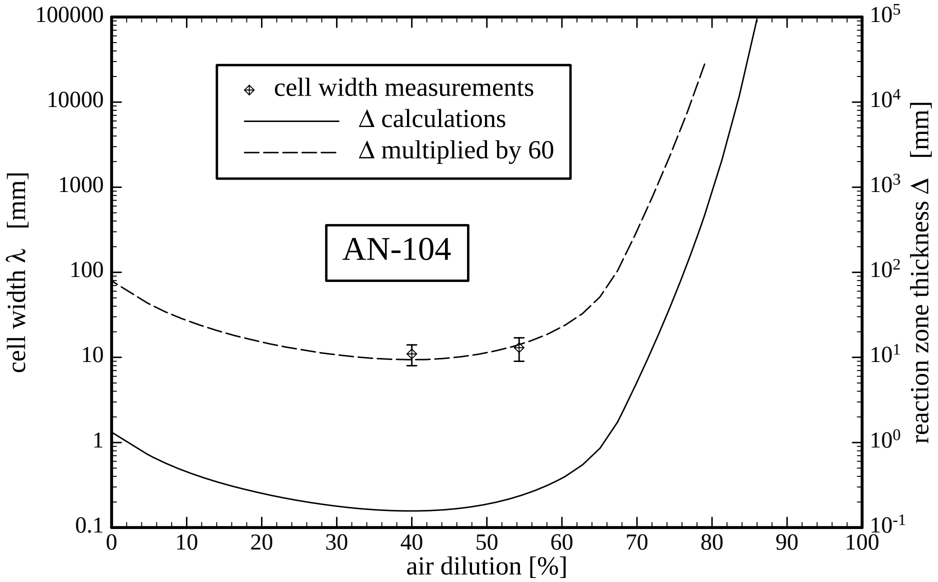

Figure 3: Measured cell width and calculated reaction zone thickness for approximate AN-104 mixtures: 47% H_2 + 19% N_2O + 33.08% N_2 + 0.02% NH_3 + 0.9% CH_4 (Akbar et al. 1997).

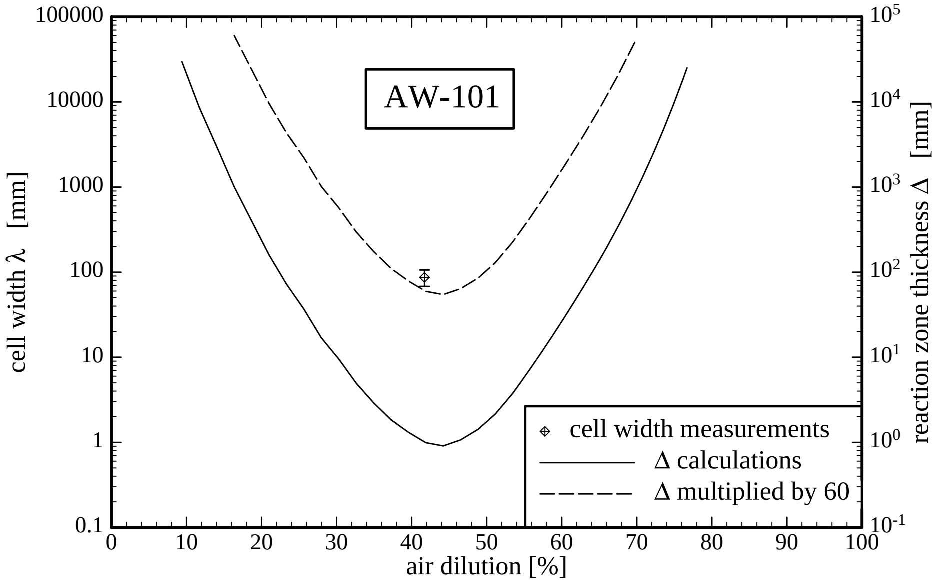

Figure 4: Measured cell width and calculated reaction zone thickness for approximate AW-101 mixtures: 31% H_2 + 4.3% N_2O + 63.08% N_2 + 0.02% NH_3 + 1.6% CH_4 (Akbar et al. 1997).

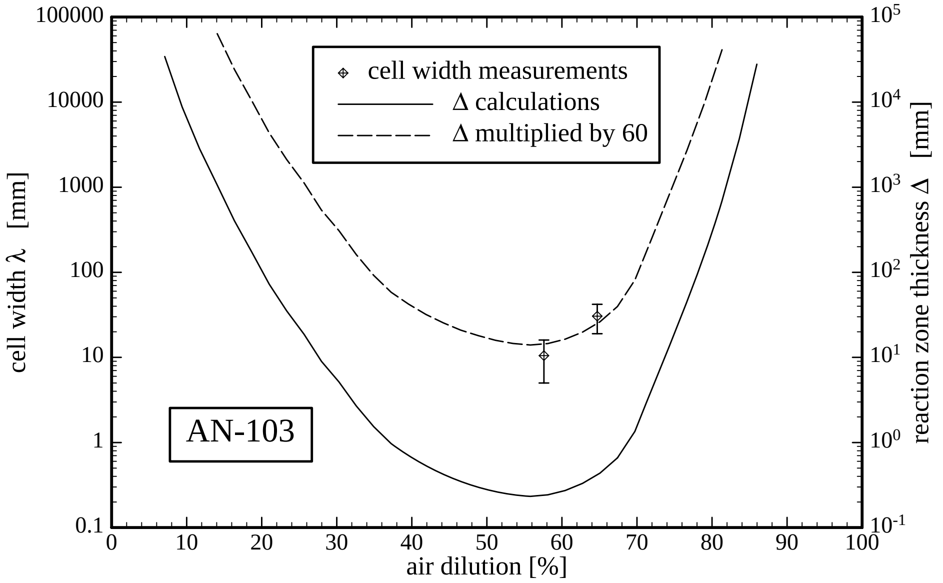

Figure 5: Measured cell width and calculated reaction zone thickness for approximate AN-103 mixtures: 61% H_2 + 3.8% N_2O + 35.14% N_2 + 0.05% NH_3 + 0.01% CH_4 (Akbar et al. 1997).

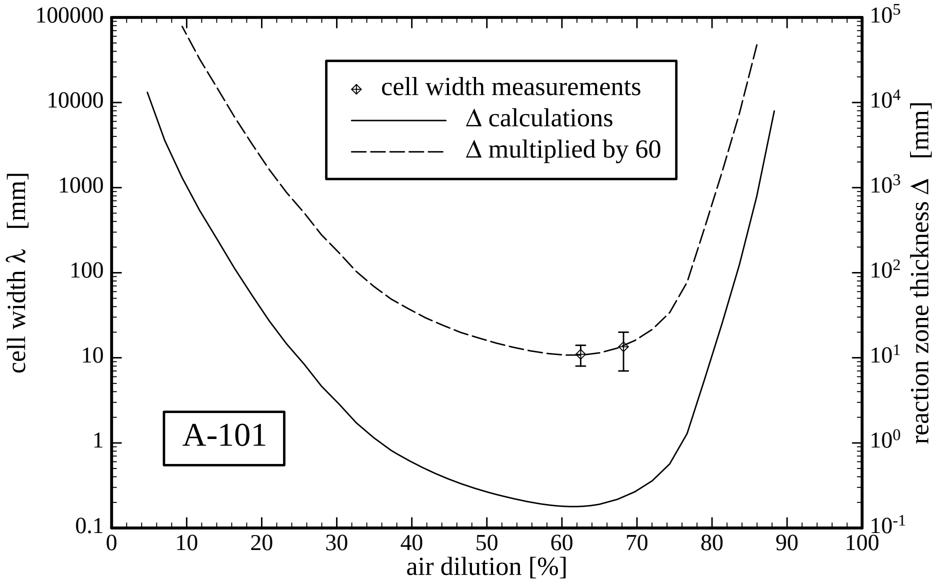

Figure 6: Measured cell width and calculated reaction zone thickness for approximate A-101 mixtures: 75% H_2 + 5.6% N_2O + 16.3% N_2 + 2.4% NH_3 + 0.7% CH_4 (Akbar et al. 1997).

We conclude that detonation is clearly not possible within the undiluted retained gas of tanks AN-105, A-101, AN-103 and AW-101. This is due to both the large cell widths for the pure retained gas mixtures and the apparently large ignition energies (discussed below) needed for flame propagation. In tank SY-101 and AN-104, detonations are possible within the undiluted retained gas but the cell sizes are sufficiently large that propagation within sub-millimeter pores is not possible. In tank SY-101, the cell width of the undiluted retained gas is on the order of 20-30 mm indicating that pore/crack/dendritic bubble sizes will have to exceed 2.5 mm in order for detonation propagation to be possible. However, as discussed below, it is extremely difficult to even ignite flames in these mixtures and it is very unlikely that detonation initiation will occur with ignition sources created by typical intrusive operations.

We conclude that detonation propagation within sub-millimeter pores is unlikely for all of the tanks examined in our study. For other tanks, a bounding estimate can be developed by observing that of all the possible retained gas mixtures containing H_2, N_2O, CH_4, NH_3, and N_2, the most sensitive to detonation will be a stoichiometric H_2–N_2O mixture (Akbar et al. 1997). The cell width for this mixture is 1 to 3 mm, so that a minimum pore size of 130 to 390 μm is required for detonation propagation in undiluted retained gas within the waste. Dilution with air or N_2 will increase the cell width (Akbar et al. 1997) and the corresponding minimum pore size for detonation propagation.

3.2 Deflagration

Previous studies on flame propagation within porous material concentrate on determining the Peclet number Pe, which is a function of the laminar burning velocity S_L, the equivalent porous cavity diameter d_m, the specific heat capacity c_p, the density ρ, and the heat conductivity k of the unburned gas mixture. Flame quenching occurs when the Peclet number is less than some critical Peclet number Pe^*:

$$Pe \leq Pe^* \qquad \text{for quenching,} \tag{12}$$

$$Pe > Pe^* \qquad \text{for propagation.} \tag{13}$$

Lyamin and Pinaev (1985) obtained a critical Peclet number of $Pe^* = 85$ for flame propagation through a tube, whereas Trimis and Durst (1996) measured a critical Peclet number of $Pe^* = 65$ for combustion in a porous medium. With the help of the critical Peclet number critical values for the equivalent porous cavity diameter d_m, for the pebble or sphere diameter δ and for the hydraulic diameter d_h can be calculated:

$$d_{m,crit} = \frac{Pe^* \ k}{S_L \ c_p \ \rho} \ , \tag{14}$$

$$\delta_{crit} \approx 2.77 \ d_{m,crit} \qquad \text{for sphere packings,} \tag{15}$$

$$\varepsilon \approx 0.36 \qquad \text{for sphere packings,} \tag{16}$$

$$d_{h,crit} = \frac{\varepsilon \ \delta_{crit}}{3(1-\varepsilon)} \ . \tag{17}$$

Flame quenching occurs if the actual values are less than the critical values:

$$d_m \leq d_{m,crit} \ , \tag{18}$$

$$\delta \leq \delta_{crit} \ , \tag{19}$$

$$d_h \leq d_{h,crit} \ . \tag{20}$$

Table 3 shows the calculated values for the representative retained gas compositions SY-101, AN-105 and AN-104 in comparison to stoichiometric hydrogen-nitrous oxide and hydrogen-air mixtures. The retained gas compositions in the tanks SY-101, AN-105 and AN-104 are given in Table 2. Furthermore, the retained gas compositions in the tanks AW-101, AN-103 and A-101 are shown in Table 2, but no calculations were carried out for these three compositions. Those compositions are expected to be less flammable because of the very low amount of oxidizer and in the case of tank AW-101 also because of the high nitrogen dilution (60%). All calculated data in Table 3 are based on the thermodynamic conditions and laminar burning velocities which are also given in Table 3. The laminar burning velocities S_L for SY-101, AN-105 and AN-104 of 1.0 m/s are assumptions based on our previous work (Pfahl and Shepherd 1997).

Equations 15-17 are valid for spherical particles. We do not have data to show that these relations are valid for wastes in the Hanford storage tanks. As discussed before, the form of retention would alter the characteristic dimension used in Peclet number criteria. The use of maximum packing fraction is questionable if particles are displaced as a result of gas expansion. The gas void fraction would be an alternative parameter in those cases. The maximum void fraction is reported to be as high as 50% (Gauglitz et al. 1996). Therefore, $d_{h,crit}$ may increase at higher void fractions and Eqs. 14-20 must be used with care.

The flammability of each representative mixture in Table 3 was determined by experimental measurements and the results are given in Table 4. As one can see in Table 4, all three investigated representative retained gas compositions are not flammable without dilution at quiescent conditions, but at present it is not known, how much air dilution is necessary to reach the upper flammability limit. For the gas composition in tank SY-101, 10% air dilution is sufficient to observe flammability at turbulent conditions and the gas composition in tank AN-104 is flammable without dilution at turbulent conditions (see Table 4). Note that the results of Table 4 were obtained with approximate gas compositions rather than the values given in Table 2. The 2% H_2O for SY-101 and the 0.02% NH_3 for AN-105 and AN-104 were not included and the balance was assumed to be N_2 for SY-101, CH_4 for AN-105, CH_4 and N_2O for AN-104. Note that the experiments given in Table 4 were performed with an ignition energy of 8 J. Much more energetic ignition sources (up to 5000 J), simulating ignition sources such as lightning strikes, are capable (Cashdollar et al. 1992) of igniting very lean hydrogen–nitrous oxide mixtures. Further study would be needed to examine this issue for the mixtures in Table 4. It may be possible that some of the mixtures will support flame propagation if such an extreme ignition stimulus is used. Certainly, the SY-101 mixture will detonate with a 80 kJ ignition source, such as used in the cell width measurements of Akbar et al. (1997).

With an assumed initial pressure of 1 atm, the critical hydraulic diameter $d_{h,crit}$ is 2.7 mm for SY-101, 2.0 mm for AN-105, 2.3 mm for AN-104, 0.18 mm for a stoichiometric hydrogen-nitrous oxide mixture and 0.41 mm for a stoichiometric hydrogen-air mixture. Abbott (1998) obtained in his experiments for a salt simulant based on the waste in tank SY-101 permeabilities between 2.8×10^{-8} and 3.7×10^{-8} cm^2. Compared to measured permeabilities of glass bead packed beds (Dullien 1979; Shepherd and Begeal 1988) these permeabilities correspond to particle diameters in the order of 100 μm or characteristic pore sizes of 36 μm, roughly a factor 50 smaller than needed for flame propagation. This indicates that flame propagation is unlikely for representative retained gases in SY-101 if the measured permeabilities of the simulant are truly representative of the actual porous waste within the tank. The evaluation for other tanks cannot be made since the materials have not been characterized.

The permeability measurements of Abbott (1998) were obtained using a gas-free waste simulant (except for dissolved ammonia). The actual permeability of waste or crust may be higher than what is measured by Abbott (1998). Higher permeability would yield higher pore sizes, particle or hydraulic diameters. Considering all of the different forms of gas retention and waste types, we conclude that the propagation of deflagration is unlikely but not impossible.

Table 2: Representative retained gas compositions from the waste tanks at Hanford Site.

mixture	H_2 [%]	N_2O [%]	N_2 [%]	NH_3 [%]	CH_4 [%]	H_2O [%]	O_2 [%]
SY-101	29	24	33	11	1	2	
AN-105	63	11	25	.02	0.7		
AN-104	47	19	32	.02	0.9		
H_2-N_2O	50	50					
H_2-air	29.6		55.6				14.8
AW-101	31	4.3	60	.02	1.6		
AN-103	61	3.8	34	.05	.01		
A-101	75	5.6	16	2.4	0.7		

4 Conclusions

This evaluation indicates, that in the tanks SY-101, AN-105, AN-104, AW-101, AN-103 and A-101 at Hanford Site a flame or detonation would probably not propagate through the porous waste.

Table 3: Calculated critical length-scales for the representative retained gas compositions and for stoichiometric hydrogen-nitrous oxide and hydrogen-air mixtures.

mixture	Pe*	T [^{o}C]	S_L [m/s]	ρ [kg/m^3]	c_p [J/(kg*K)]	k [W/(m*K)]	$d_{m,crit}$ [mm]	δ_{crit} [mm]	$d_{h,crit}$ [mm]
SY-101	65	45	1.0	0.862	1.426E+3	9.648E-2	5.1	14.1	2.65
AN-105	65	45	1.0	0.502	2.276E+3	6.867E-2	3.9	10.8	2.03
AN-104	65	45	1.0	0.704	1.700E+3	7.999E-2	4.3	12.0	2.26
H_2-N_2O (ϕ=1)	65	22	10.0	0.938	1.465E+3	7.433E-2	0.35	0.97	0.18
H_2-air (ϕ=1)	65	22	3.5	0.852	1.389E+3	5.085E-2	0.80	2.21	0.41

Table 4: Flammability of representative retained gas compositions from the waste tanks at Hanford Site; initial conditions: p_0 = 100 kPa, T_0 = 295 K; ignition energy: 8 J.

mixture	H_2 [%]	N_2O [%]	N_2 [%]	NH_3 [%]	CH_4 [%]	condition	flammable	peak pressure [bar]
$\sim$SY-101	29	24	35	11	1	quiescent	no	
$\sim$SY-101	29	24	35	11	1	turbulent	no	
90% $\sim$SY-101	29	24	35	11	1	turbulent	yes	10% air: 8.9
$\sim$AN-105	63	11	25		1	quiescent	no	
$\sim$AN-105	63	11	25		1	turbulent	no	
$\sim$AN-104	47	20	32		1	quiescent	no	
$\sim$AN-104	47	20	32		1	turbulent	yes	8.5

Flammability measurements (for tanks SY-101, AN-105 and AN-104, see Table 4) and gas compositions (for tanks AW-101, AN-103 and A-101) of the retained gas mixtures sampled to date show, that it is extremely difficult to even ignite flames in these mixtures. However, at present it is not known, how much air dilution is necessary to get flammable gas compositions in tanks SY-101, AN-105 , AN-104 and especially in tanks AN-103 and A-101, whereas the representative retained gases in tank AW-101 are expected to be less flammable with and without air dilution because of the high nitrogen dilution.

In addition to the flammability of the representative retained gases, evaluation of the critical length scales indicates that detonation propagation is unlikely within the waste in the investigated tanks. Furthermore, it indicates that flame propagation is unlikely within the waste in tank SY-101, whereas no evaluation for flame propagation can be made for other tanks since the materials have not been characterized.

However, there is enormous uncertainty in evaluating the possibilities of combustion (deflagration and detonation) within the porous waste that would vary considerably in each waste storage tank and within the waste itself. The main uncertainties are:

1. The wastes in the tanks at Hanford site have to be characterized as a porous medium. Measurements of the following properties need to be carried out: geometrical configuration of pores, porosity, permeability , specific heat, thermal conductivity and water content.

   In a complex material such as the porous waste, experimental characterization is the only reliable way to assess such issues. The work of Abbott (1998) represents the first steps needed in that direction.

2. There have been no studies of flame propagation in porous materials using the gas mixtures of interest to Hanford. The mixtures of interest have very different Lewis numbers and reaction products than the hydrocarbon-air mixtures typically studied in previous work. Although propagation criteria have been proposed based on a critical Peclet number, there is scant evidence that it is applicable to mixtures containing large amounts of nitrous oxide or to mixtures that have large deviation of the Lewis number from unity.

3. The laminar burning velocities (see Table 3) have not been measured accurately yet and the values ($S_L = 1$ m/s) used in our evaluation are just assumptions based on our previous experimental work (Pfahl and Shepherd 1997). Further information on the flammability of the retained gas compositions from the waste tanks is given in Table 4. Whereas the retained gas composition in tank SY-101 is not flammable without dilution, the mixture becomes flammable with just 10% air dilution (see Fig. 48 in Pfahl and Shepherd 1997).

4. Measurements of crusts in several tanks indicate the presence of significant amounts of water (20 to 30%) which has been postulated to have a mitigating effect on flame initiation and propagation. At present, there are no data on the effect of water on combustion in porous materials but we expect that this will depend strongly on the specific surface area of the water and the flame speed. Lacking any experimental data on this effect, it is difficult to make predictions.

5. The porous waste can be unstable, which means it can crack and be separated into different layers. This can lead to void areas in the porous material. Even if the flame quenches in the porous waste itself it could possibly propagate within larger cracks or void volumes.

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