Fate of Formaldehyde in MDF Sawdust during MSW Composting

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ABSTRACT

Medium Density Fibreboard (MDF) sawdust is an important source of carbon and is therefore in demand as an amendment in commercial municipal solid waste (MSW) composting operations. However, MDF contains formaldehyde which causes two concerns. The first one is that the bactericidal and fungicidal properties of formaldehyde may adversely affect the composting process. The second one is that formaldehyde residues may cause the production of compost of unacceptable quality for unrestricted use.

A project has been designed to determine the fate of formaldehyde when MDF sawdust is co-composted with MSW at various proportions (2.5% and 5.0% MDF added) and two temperatures (45°C and 55°C) in a lab-scale reactor under controlled and optimal conditions such as moisture content 45-60%, aeration 1.0 l/h, particle size 40 mm, and C/N ratio 20-30 during periods of 10 days. 2.5% or 5.0% MDF sawdust was added to the MSW mixture. Samples of the substrate were collected every two days to determine the degradation rate of formaldehyde. Formaldehyde concentrations in the solid matrix were determined according to EPA Method 8315A and HPLC.

Dry matter loss over 10 day was less than 14% for the mesophilic phase and 13% for the thermophilic phase. Formaldehyde was reduced over 90% in both mesophilic and thermophilic phases and no gaseous formaldehyde emissions were detected during either phase. The formaldehyde degradation during the thermophilic phase composting was not significantly different whether 2.5% or 5.0% MDF sawdust was added.

INTRODUCTION

During the past 40 years medium density fiberboard (MDF) produced from small wood fragments and sawdust bonded with urea-formaldehyde resin has been increasingly used to make furniture and components of building materials due to its good strength and low cost (CanFibre, 2000). This use of MDF leads to large quantities of sawdust which become part of the municipal solid waste (MSW) stream. Instead of landfilling the material, it could be diverted and used as a carbon source in composting process. However, as MDF sawdust contains formaldehyde at concentrations of 330 to 360 μg/g (Maxxam Analytical Inc.), composting facility operations are reluctant to use it unless they can be assured that formaldehyde will decompose during the process.

Formaldehyde, which is an important industrial chemical, has various uses. It is widely used in the chemical industry, textile processing, paper industry and wood processing. It is used as a reactant to make other chemicals, such as phenolic compounds, acetylenic compounds, pentaeryythitol, hexamethyl tetramine, methyline dianiline, pyridine, nitroparaffin derivatives, herbicides, fertilizer coating, pharmaceuticals, and elastomeric sealants (EPA, 1998). It is also found in wastewater from resin manufactures (Goeddertz *et al.*, 1990), textiles, and petrochemical plants (Sharma *et al.*, 1994).

In general formaldehyde gas is potentially harmful, causing eye and nose irritation. It is also considered to be a carcinogen (Mazumber, 1997). The allowable levels of formaldehyde are equal to or less than 0.4 μg/L in household areas following the air quality standards of the U.S. Government (Keener *et al*, 1994). Not only is formaldehyde a toxic substance to humans, animals and plants, but it also inhibits the growth of microorganisms such as bacteria and fungi (Eicker and Apostolides, 1986; Gerrits, 1986, Griess, 1985; Qu and Bhattacharya, 1997; Omil *et al.*, 1999).

Formaldehyde is degradable under aerobic and anaerobic conditions in aqueous and soil media (Adroer *et al.*, 1990; Azachi *et al.*, 1995; Gerike and Gode, 1990; Omil *et al.*, 1999). In industrial wastewater, formaldehyde is able to degrade under aerobic conditions (Szetela, R *et* al., 1987). There is little information regarding formaldehyde degradation during composting. Keener et al. (1994) studied formaldehyde emission during composting of spent press-molded, wood fibre pallets bonded with urea-formaldehyde. They reported on dry matter loss, formaldehyde emission and ammonia emission. No evidence is directly determined for formaldehyde degradation from their experiments.

It is well-known that temperature is one of the most important parameters controlling the biodegradation rates of the composting process. In composting the temperature changes from a room temperature to the temperature of 45°C due to an activity of mesophilic microorganisms in degrading organic materials. This activity leads to temperature increase from 45°C to 55°C or higher. The temperature that is higher than 55°C is an optimum temperature for thermophilic microorganism growth. Generally, the degradation rates of organic materials between these two temperatures are quite different. Therefore, the rate of degradation of formaldehyde in MDF may differ under mesophilic (<45°C) and thermophilic (50-60°C) conditions. Knowledge of the effect of temperature on formaldehyde biodegradation would be an important step in safe disposal and composting efficiency. Furthermore, the role of temperature in affecting the physical removal of formaldehyde via volatilization has yet to be determined. However, no research has studied the biodegradation of formaldehyde contained in a solid matrix, as would be the case in composting of MDF waste. Several researchers studied formaldehyde biodegradation on soil. For example, Mohn (1997) studied the biodegradation of urea formaldehyde polymer which is used as a sorbent for containment and clean up of hydrocarbons. The results showed that degradation of urea formaldehyde polymer was very slow and incomplete. Furthermore, nitrogen is required for degradation of the hydrocarbon sorbed on polymer.

MATERIALS AND METHODS

Simulated MSW and MDF sawdust

The simulated MSW formula, which was used by Arsenault (1996) and Macdonald (1995), was also used in these experiments. It is consists of water, sand, rabbit chow, and newspaper in order to obtain a suitable simulated MSW substrate. The relative amount of these components is shown as table 1.

Table 1. Simulated MSW characteristics (MacDonald, 1995)

Component	Mass (g)	Carbon (%)	Nitrogen (%)	Water (%)
Water	196	-	-	100
Sand	95	-	-	-
Rabbit Chow	65	42.9	2.3	10.0
Newspaper	44	46.4	0.17	8.9
Total (g)	400	56.2	1.57	206.4

The simulated MSW is ground to a particle size of approximately 40 mm. Thorough mixing of the various constituent materials is performed to produce a relatively homogeneous substrate. MDF sawdust is added to the simulated MSW at 2.5 and 5.0 % (by weight) for each experiment. A sample of the simulated MSW is taken for analysis. The formaldehyde content of the MDF sawdust and the synthetic sample mixed with MDF sawdust were determined.

Bioreactor

The experimental apparatus is shown in Figure 1. The 2-litres, glass reactors were placed in a water bath to control the temperature.

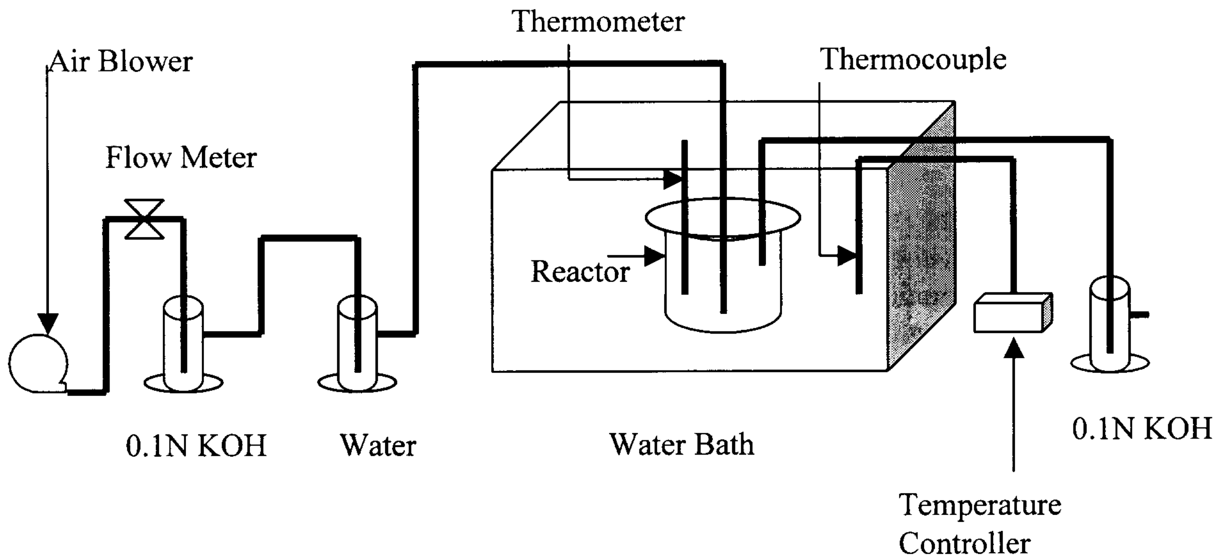

Figure 1. The laboratory scale reactor.

Procedures

1.0 Formaldehyde concentrations

Formaldehyde concentration was analyzed by using high performance liquid chromatography (HPLC) following EPA method 8315A. This proved to be challenging as formaldehyde is part of the solid MDF matrix. Formaldehyde extraction was accomplished using an acidic extraction technique over a period of 18 hours. The extracted solution was repeatedly reacted with 2,4-dinitrophenylhydrazine (DNPH), after which the sample was eluted in the cartridge column and analyzed using HPLC. The results were comparable with the standard formaldehyde solution to calculate the concentrations.

2.0 Fixed temperatures

Two reactors were used to determine the role of temperature on formaldehyde removal during composting. Simulated MSW was used as a positive control in one reactor for a period of 10 days. The other reactors contained MDF as well. Temperature of the water bath was maintained at 40-45^{o}C for the mesophilic phase and 50-55^{o}C for the thermophilic phase, while the air supply set at 1.0 $Lmin^{-1}$ was controlled by using a timer. This on-off sequencing of the timer is often used with the static pile system and some reactor systems (Haug, 1993). Each experiment was duplicated. Experimental samples were comprised of the simulated MSW combined with MDF sawdust to give various C/N ratios and formaldehyde concentrations.

3.0 Biodegradation rates

Experimental substrate will be comprised of the simulated MSW combined with 2.5% and 5.0% MDF sawdust to give a C/N ratio equal to 20-30. Temperature will be maintained at 55 ^{o}C for the thermophilic experiments, while the air supply, set at 1.0 $Lmin^{-1}$, may be adjusted depending on temperature and biodegradation of organic materials. Each experiment will be done at least three times depending on the standard errors per experimental unit (Cochran and Cox, 1950). In all cases the substrate will be kept in the reactor for a period of 10 days.

4.0 Adiabatic system

Experimental substrate will be comprised of the simulated MSW combined with 2.5% MDF sawdust and all samples were composted in an adiabatic system. The temperature of the reactor is controlled by submerging it in a water bath with a temperature controller. The temperature of the water bath is controlled by the temperature of substrate in the reactor, thus minimizing the heat loss from the reactor to the environment.

The total carbon and total nitrogen contents (APHA, 1991) were analyzed to determine carbon removal. Formaldehyde emission was determined daily from exhausted gas using the Dräger tube technique. The CO_2 gas produced was trapped in a 0.1N KOH solution. The solution was titrated daily by using 0.1 N HCl to determine the CO_2 concentration. The moisture content of the substrate was measured as described in Standard Methods for the Examination of Water and Wastewater (APHA, 1991).

RESULTS AND DISCUSSION

Formaldehyde concentration

Formaldehyde concentrations in MDF sawdust were determined following EPA method 8315A. The results demonstrated that in the original MDF samples formaldehyde concentrations varied from 217.75 to 405.35 μg/g of dry MDF. These high concentrations may inhibit the microbial growth in the composting process because formaldehyde can inhibit the growth of aerobic and anaerobic bacteria (Qu and Bhattacharya, 1997). As low as 1-2 mg/L of formaldehyde inhibited the growth of *Pseudomonas fluorescens* and *E. coli* (Verschueren, 1983) while 30 mg/L of formaldehyde inhibited oxygen consumption of activated sludge (Gerike and Gode, 1990).

Fixed temperatures

The experiments were performed under the fixed temperatures of 45°C (mesophilic phases) and 55°C (thermophilic phases) in order to determine the types of microorganisms on formaldehyde degradation. A typical temperature profiles is shown in Figure 2. The results showed that in both cases there was an overshoot of the set point temperature at day1. It took about one day for the system to stabilize.

In fixed temperature reactors, the optimal temperature for decomposition can be judged by the CO_2 evolved. The overall CO_2 accumulation at 45°C and 55°C were significantly similar (Fig. 3) at the same substrate composition. Furthermore, the reduction in formaldehyde were not significantly different at these different temperatures (Table 2.). This is an important finding as commercial composting facilities operate at thermophilic temperatures. Hence, formaldehyde contaminated MDF sawdust appears to satisfy the conditions for co-composting with MSW (Institute of Local Self-Reliance, 1992).

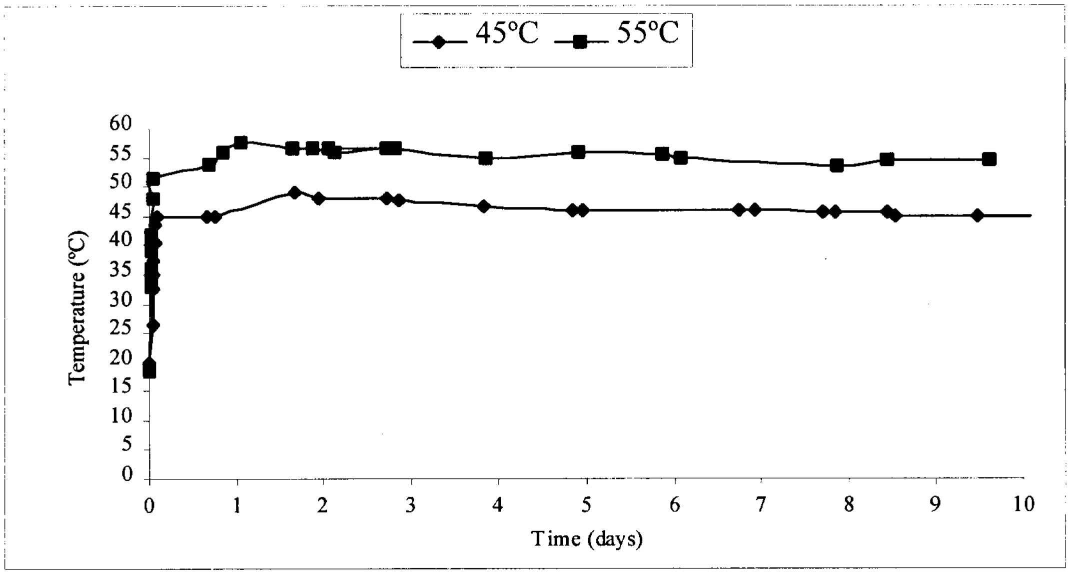

Figure 2 Temperature changes in MSW mixed with MDF sawdust composting reactor under fixed temperature controls

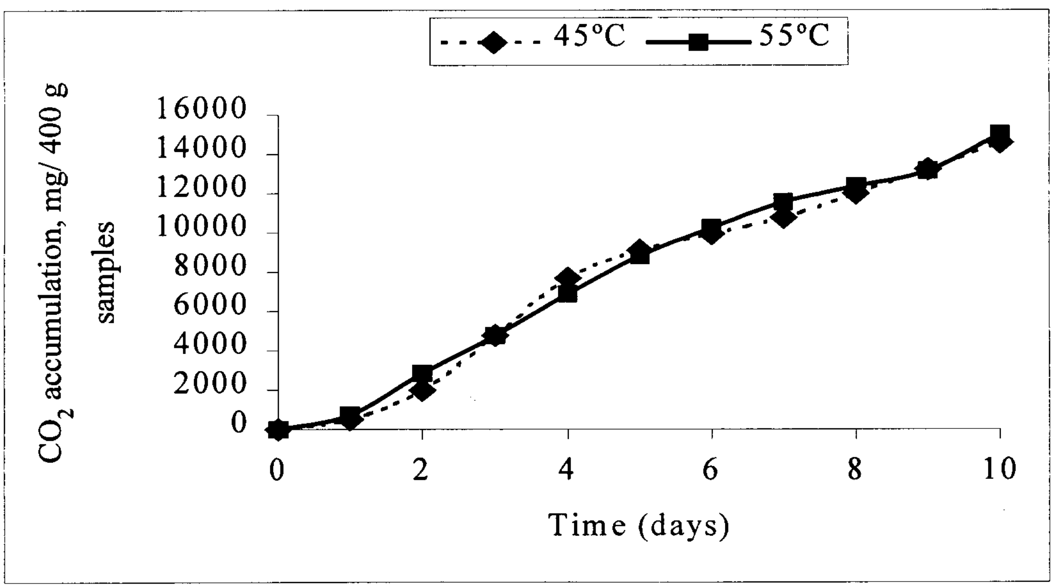

Figure 3 Cumulative CO_2 at different temperatures during MSW mixed with 2.5% MDF sawdust composting

Table 2. The reduction of formaldehyde after 10 day composting

Temperature (°C)	Formaldehyde Concentration (μg/g)		% Reduction
	Initial	Final	
45	14.79	0.97	93.44
	15.90	1.15	92.77
55	18.52	1.36	92.66
	22.58	1.82	91.94

Biodegradation rates

MDF sawdust was added to the synthetic MSW by approximately 2.5 and 5.0% wet weight in order to adjust C/N ratios of the synthetic MSW. This addition caused the C/N ratio of the synthetic MSW to be 26/1 and 37/1 after 2.5% and 5.0% MDF sawdust was added, respectively. The formaldehyde concentrations of the MSW/MDF substrate samples were varied from 37.04 to 45.17 and 5.33 to 19.36 μg/g of dry substrate for the 5.0% and 2.5% mixtures, respectively. Increased C/N ratios due to the addition of MDF sawdust was shown to affect the composting process. Figure 4 shows that there was a difference in the accumulated CO_2 evolution at 2.5% MDF and at 5.0% MDF sawdust. The addition of MDF sawdust accelerates aerobic microbial growth because sawdust increased C/N ratio and porosity (Imbeach, 1998).

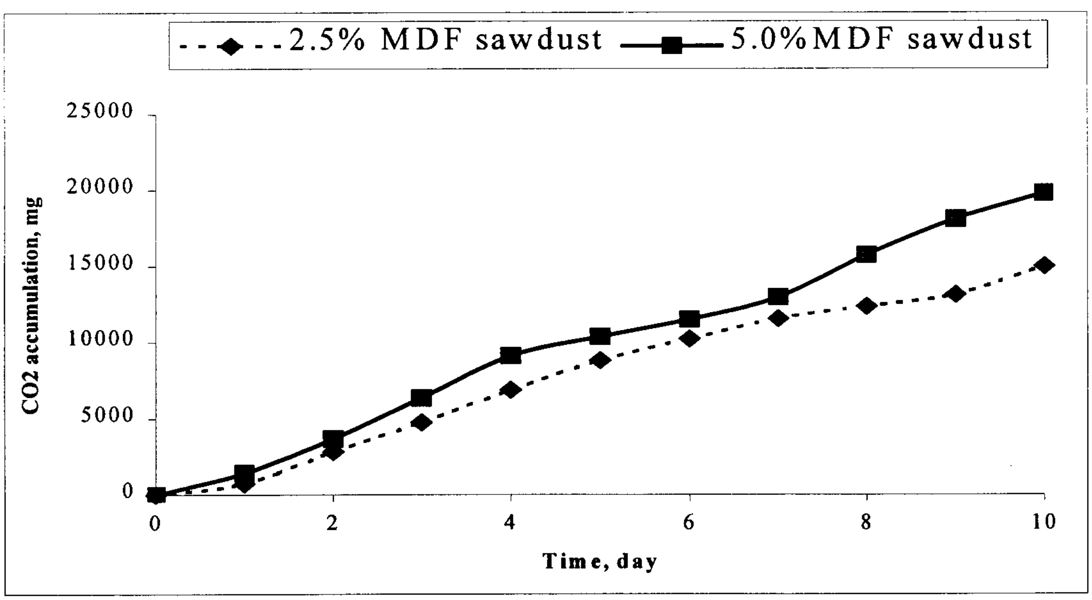

Figure 4 Cumulative CO_2 production at 2.5% and 5.0% MDF sawdust addition at 55°C

The formaldehyde concentrations of the composted substrate samples were substantially lowered (Table 3.). The reduction was about 80%, independent of temperature and the amount of MDF added. Figure 5 shows the measurements of gaseous formaldehyde emission by using the Dräger tube technique. It indicates that formaldehyde emitted in the exhausted air was less than 0.02 ppm so that the reduction in formaldehyde did not involve vaporization. Keener *et al.* (1994) also found that formaldehyde emissions were less than 0.04 ppm during composting when un-amended or urea-amended materials were added.

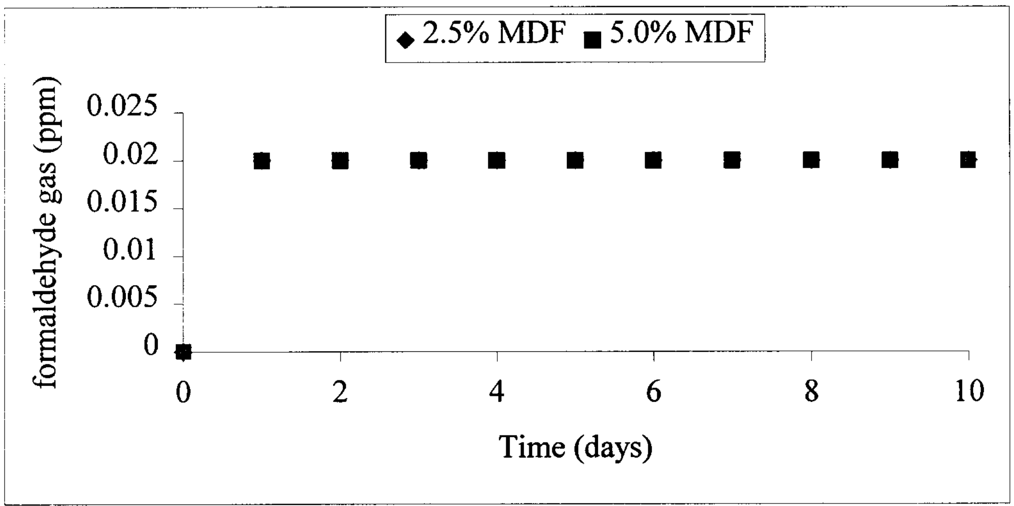

Figure 5 Formaldehyde emission after 2.5% and 5.0% MDF sawdust addition at 55°C

Further experiment was done to determine the formaldehyde biodegradation rate. Even though of the reduction of formaldehyde and the CO_2 accumulation were not significantly difference at 45°C and 55°C, the temperature of 55°C was selected for these experiments. The reasons being 1) it is an optimum temperature for thermophilic organisms, and 2) weed seeds and most microbes of pathogenic significance cannot survive at this temperature.

As seen in the figure 6, the biodegradation of formaldehyde increases rapidly during the second day and slowly after the fourth day. This was due to a reduction in moisture content by more than 50%. To test for this phenomenon, the second experiment will be done by adding some water during the composting process.

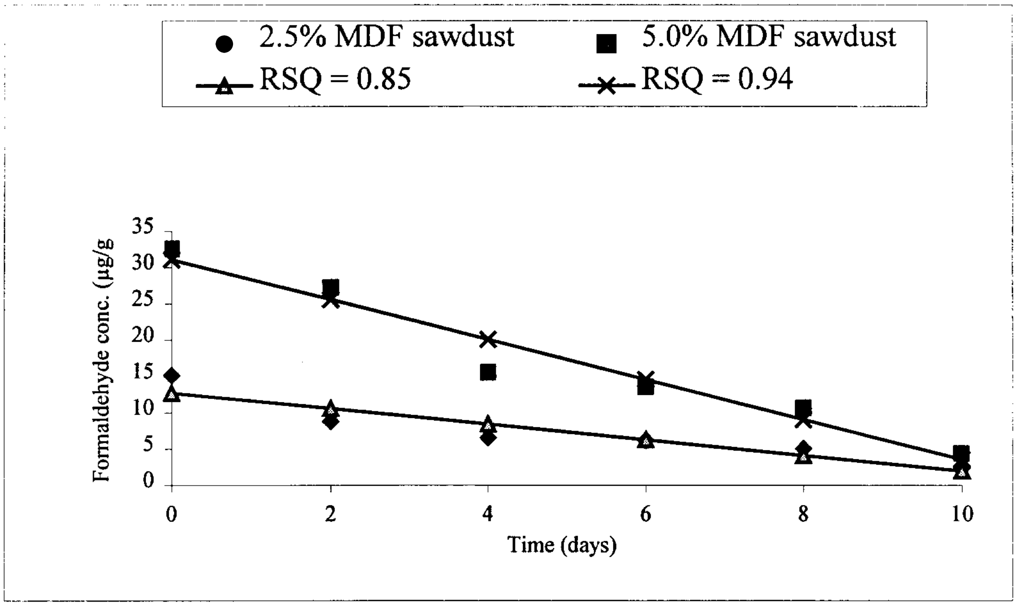

Figure 6 The biodegradation rate of formaldehyde under fixed temperature composting

In addition, the biodegradation rate of formaldehyde at different concentrations was determined. The results showed that the addition of the MDF sawdust between 2.5 and 5.0% by wet weight gave similar results in formaldehyde reduction. However, the biodegradation rate was significantly different. Table 3 shows the results from this experiment.

Table 3 The biodegradation rates of formaldehyde during composting

Time (days)	Formaldehyde concentration (µg/g)		% reduction		Biodegradation rate (µg/g.day)	
	2.5% MDF	5.0% MDF	2.5% MDF	5.0% MDF	2.5% MDF	5.0% MDF
0	15.17	32.63	0.00	0.00	-1.076	-2.756
2	8.80	27.26	41.99	16.46		
4	6.53	15.56	56.93	52.31		
6	6.20	13.56	59.15	58.44		
8	5.02	10.69	66.86	67.24		
10	2.43	4.37	83.98	86.60		

Adiabatic system

The reduction of formaldehyde was determined under adiabatic systems. The phenomenon in the adiabatic systems is almost as same as field composting. The reactor was submerged in a water bath to desire the temperature within the reactor and to reduce heat loss from conduction. The temperature in the reactor was measured and compared to the temperature in the fixed temperature reactor. The results showed that the variation of the temperature of both conditions was similar. Only was the first day of the adiabatic operation higher than the fixed temperature operation. This happened since the temperature of water in the water bath increased more rapidly than in the reactor causing of the heat conduction from the water to the reactor (Incropera and DeWitt, 1993).

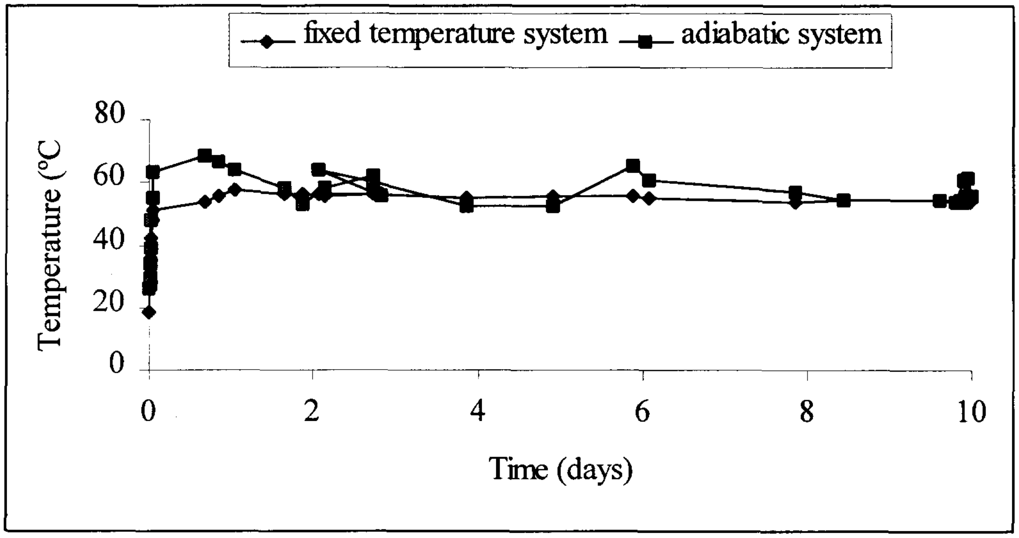

Figure 7 The comparison of temperature under the fixed temperature and the adiabatic system

The CO_2 accumulation under the adiabatic system was similar to the fixed temperature system as shown in figure 8. This event occurred since the groups of microorganisms taking part in both processes are same. The CO_2 was produced at the same level.

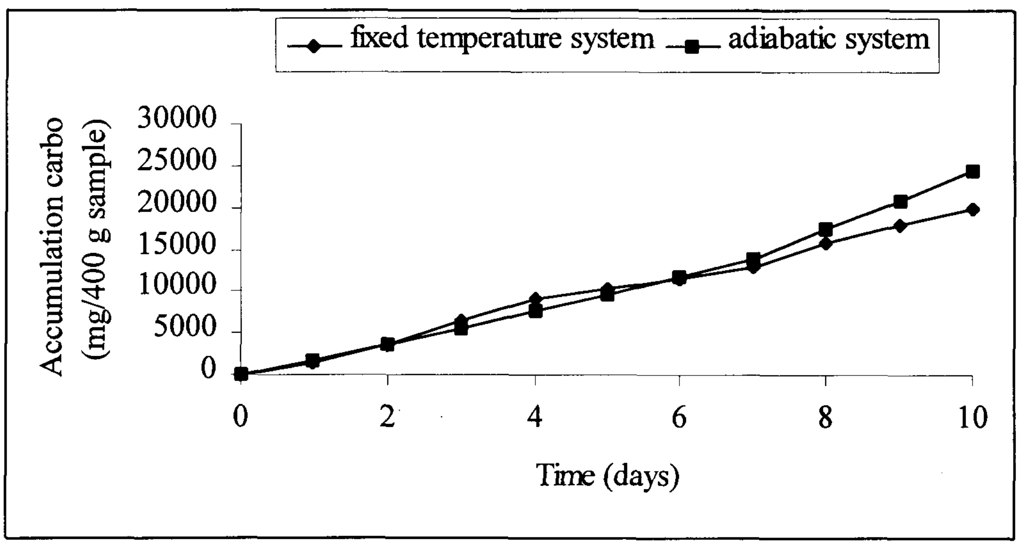

Figure 8 The CO_2 accumulation during composting under the fixed temperature and the adiabatic conditions.

However, operation under the adiabatic system is more complicated than the fixed temperature system in the lab-scale reactor. The temperature within the reactor increased slower than temperature in the water bath due to less solid waste in the reactor. The fluctuation of temperature during adiabatic operation was higher than the fixed temperature operations. Furthermore, the water cannot evaporate well within the reactor causes of an aerobic zone in the reactor (Golueke, 1972). The reduction of formaldehyde under the adiabatic system was shown as Table 4. Formaldehyde can efficiently inhibit an anaerobic microorganism if it has high concentration (Lu and Hegemann, 1998).

Table 4 The reduction of formaldehyde under the adiabatic composting process

Formaldehyde concentration (μg/g)		% reduction
Initial	After	
18.91	9.38	50
14.79	6.65	55
14.01	4.80	66

CONCLUSIONS

Bench-scale experiments demonstrated that formaldehyde contaminated in MDF sawdust depended readily during MSW composting under mesophilic and thermophilic conditions. The reduction in formaldehyde concentration was about 90% at both conditions, independent of the amount of MDF added.

Future study will be determined the effect of the higher formaldehyde concentration on thermophilic microorganisms. In addition, the formaldehyde biodegradation and urea-formaldehyde breakdown will be conducted by using radioactive carbon-14 label. This technique is used widely to determine the transformation and biodegradation of organic carbon and it might be appropriate to determine the formaldehyde biodegradation rate. This technique will also be compared to the determination of formaldehyde biodegradation using the HPLC analysis. Then, the following study will be involved in the mathematical model development of MSW mixed with MDF sawdust composting and be testified the quality of these composted product for plant.

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