MICHIGAN DIVISION
PROCESS MATH MODELING

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Subject: HEAT CONDUCTION IN NUCLEAR WASTE DISPOSAL FORMS

Summary:

A Topical Report was submitted by DOW to the Nuclear Regulatory Commission for approval for disposing low-level nuclear waste (certain ion exchange resins) in vinyl ester-styrene emulsified forms. The aim of this work is to answer some points raised by the NRC with regard to the temperature profile in the cylindrical forms at the end of the ASTM temperature exposure cycles recommended by the NRC.

The forms consist of a solidified emulsion of the wastes in vinyl ester-styrene in a polyethylene cup. The two major temperature exposure cycles as defined by ASTM B-553 are (i) one hour exposure to 60 C and (ii) one hour exposure to -40 C. The thermal cycling tests are performed to ensure the stability of the low-level nuclear wastes during storage, handling and transporation.

The NRC, in its response to the Topical Report, suggested that heat transfer calculations be performed to determine the temperatures in the form at the end of these thermal cycles. In this work, the heat transfer by convection to the cylinder and by conduction within the cylinder are modeled. The temperature profile in the composite cylindrical forms at the end of each exposure cycle is determined. The profiles obtained by numerical method on an IBM PC are compared with that estimated by approximate analytical solution.

The results from this work have been communicated to the Nuclear Regulatory Commission by H. E. Filter and T. D. Boyce.

INTRODUCTION

The nuclear resin wastes are emulsified in vinyl ester-styrene. The emulsion is solidified in a cylindrical polyethylene cup. These cylindrical forms containing low-level radioactive wastes are buried. The Nuclear Regulatory Commission has established guidelines for the performance and technical testing of these forms.

The NRC has issued a Technical Position on Waste Form to list the guidelines on test methods. One of the concerns is that the waste material should be resitant to thermal degradation. The TPWF specifies that the thermal cycle based on ASTM B533-79 ("Standard Test Method for Thermal Cycling of Electroplated Plastics") be followed. One cycle consists of:

1. Waste forms at thermal equilibrium at room temperature.
2. Heating for one hour in a 60 °C oven.
3. Equilibrating to room temperature for at least one hour.
4. Cooling at -40 °C for one hour.
5. Equilibrating to room temperature.

Considering that ample time is given for the forms to equilibriate to room temperatures, the two primary steps are 2 and 4.

OBJECTIVE

The objective is to model the heat transfer process for steps 2 and 4, and determine the temperature profile in the composite cylindrical form at the end of these cycles. The specific temperatures of interest are the temperatures at the surface and at the center of the cylindrical forms.

MODEL

The heat transfer process occurs in two steps:

1. Heat transfer by convection between ambient air and the surface of the cylinder:

   The total heat transfer is given by

$$\boxed{Q = hA(Ta - Ts)} \qquad (1)$$

where
Q = total heat transfer (cal)
h = convective heat transfer coefficient (cal/sec.cm^2.C)
A = surface area (cm^2)
Ta = ambient temperature (C)
Ts = surface temperature (C).

2. Heat transfer by conduction from the cylinder surface to the center:

The problem here is that of unsteady state heat conduction in a composite cylinder. This is defined by the simplified heat equation in cylindrical coordinates

$$\boxed{\frac{\partial T}{\partial t} = \alpha \left[\frac{\partial^2 T}{\partial r^2} + \frac{2}{r} \frac{\partial T}{\partial r} \right]} \tag{2}$$

where
- T = temperature (C)
- r = radial distance (cm)
- α = thermal diffusivity (cm^2/s)

ASSUMPTIONS

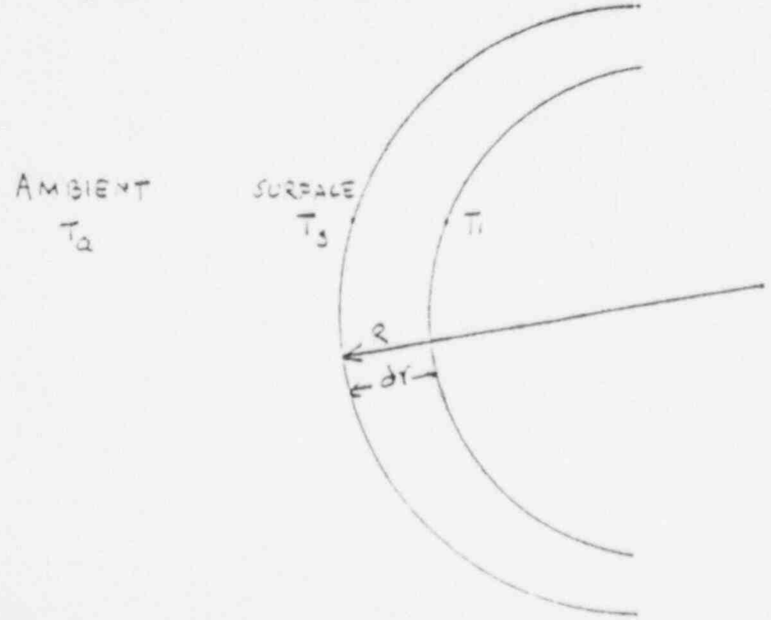
1. Heat transfer is assumed to be in the radial direction only and that in the axial direction is neglected.
2. The contact resistance between the polyethylene cup and the polymer is assumed to be negligible.

DATA

The relevant data is shown in Appendix A.

ESTIMATION OF SURFACE TEMPERATURE

To determine the amount of heat transferred between the ambient air and the surface of the cylinder, it is necessary to have an estimate of the time-dependent surface temperature. Consider an elemental ring of thickness dr at the surface within the polyethylene cup. An energy balance around this ring can be developed as:



Total heat in at time dt = $h(A1)(Ta - Ts)\,dt$

Total heat out at time dt = $k(A2)[(Ts - T1)/dr]\,dt$

Total heat accumulated at time dt = $m(Cp)(T_{s,n+1} - Ts)$

where
- dt = time step (sec)
- dr = thickness of the elemental ring (cm)
- k = thermal conductivity of polyethylene (cal/sec.cm.C)
- A1 = external surface area of the elemental ring (cm^2)
  = $2(\pi)RL$ where R = radius of the cylinder (cm) and L = length of the cylinder (cm)
- A2 = internal surface area of the elemental ring (cm^2)
  = $2(\pi)(R-dr)L$
- m = mass of the elemental ring (gm)
  = $(A2)(dr)(\rho)$, where ρ = density (gm/cm^3)
- Cp = specific heat of polyethylene (cal/gm.C)
- T1 = temperature of the inside surface of the elemental ring (C)

T1 and Ts represent the respective temperatures at any time t.

$T_{s,n+1}$ = surface temperature at time t+dt (C).

Heat in - Heat out = Heat accumulated

and therefore,

$$\boxed{T_{s,n+1} = [(y)Ta + T1 + (M-y-1)Ts]/b} \qquad (3)$$

where $y = h(dr)[1+dr/(R-dr)]/k$

and $M = (dr)(dr)/\alpha(dt)$

where the thermal properties are those of polyethylene.

Hence, starting with the surface temperature as the room temperature initially, the surface temperature at any time can be evaluated from Eq. 1.

METHOD OF SOLUTION FOR HEAT EQUATION

The entire cylinder is divided into small elemental rings radially. The heat equation (Eq. 2) is then solved for each element by a finite difference scheme. Application of finite difference to Eq. 2 results in the following explicit recursive relationship,

$$\boxed{T_{m,n+1} = [(1+m)T_{m+1} + (M-m-2)T_m + T_{m-1}] / M} \qquad (4)$$

where T_{m+1}, T_m and T_{m-1} correspond to the temperatures at the m+1, m and m-1 node points respectively at time step n, $T_{m,n+1}$ is the temperature at node m at time step n+1, and

$$m = dr/r$$

$$M = (dr)(dr)/\alpha(dt)$$

One possible approach to derive Eq. 4 is presented in Appendix B. For the numerical stability of this scheme, M should be greater than 2. This dictates the size of the time step (dt) and the grid size (dr).

Eq. 4 shows that the temperature at the (n+1)th time step at node m can be calculated from the temperatures at the previous time step at nodes m+1, m and m-1.

The procedural steps and the LOTUS 1-2-3* spreadsheet are shown in Appendix C.

RESULTS AND DISCUSSIONS

The temperature profiles at the end of one hour for the heating and cooling cycles are shown in Tables 1 and 2 and in Figures 1 and 2.

For heating, $T_s = 40.2°C$ $T_c = 37.2°C$

For cooling, $T_s = -10.3°C$ $T_c = -5.8°C$

where Tc is the temperature at the center of the cylindrical form.

The following conditions were applied to get the above results:

1. **Area**- The heat transfer by convection between ambient air and the surface was assumed to be only in the radial direction. Any heat transfer from the ends of the cylinder was neglected.

2. **Convective heat transfer coefficient (h)**- An h value of 2 $Btu/hr.ft^2.C$ was assumed. For the case of natural convection only, h is in the range 0.5 to 1. With minimum forced convection, h=2 is recommended. This represents a conservative estimate for h.

3. Theroom temperature(which is also the initial temperature of the form) was assumed to be 20°C.

DISCUSSIONS

1. The temperature at the surface and that at the center are not significantly different (about 3 to 4°C) when compared to the overall temperature change (about 20 or 30°C). This implies that the presence of polyethylene cup does not offer significant resistance to heat transfer, eventhough polyethylene has a thermal diffusivity about three times lower than the polymer. This is due to the very small thickness of the polyethylene cup (0.15 cm) as compared to that of the polymer (2.337 cm).

2. **APPROXIMATE ANALYTICAL SOLUTION:**

For the case of a one-material cylinder of infinite length and with uniform temperature distribution, an analytical solution can be obtained to calculate the temperature profile. This can be modified (as shown in Appendix D) to give an approximate analytical solution for this problem, which is given by,

$$\boxed{(T_a - T_{uni})/(T_a - T_{ini}) = \exp(-0.005ht)} \quad (5)$$

where t = time (min)
T_{uni} = uniform temperature of the cylinder (C)
T_{ini} = initial temperature of the cylinder (C), and
h is in Btu/hr.ft^2.C

The analytical solution shown in Eq. 5 compares very well with the temperatures obtained from numerical solution. The comparison is shown graphically in Figures 3 and 4 for the heating and cooling cycles respectively.

Though highly accurate, the numerical procedure can be very time-consuming. As can be seen from Figures 3 and 4, Eq. 5 provides an elegant yet adequately accurate solution to the model.

SENSITIVITY ANALYSIS

Knowing that Eq. 5 represents a good solution to the problem, it was used to study the sensitivity of h and the surface area on the temperature profile. This analysis is shown in Table 3. In Table 3, the effect of h and surface area on the tempeartures are shown as a function of exposure time. The h values studied are 0.5, 1, 2 and 3. For each h, two values of surface area considered. They are, (i) where convection through the ends of the cylinder is neglected, and (ii) the same is accounted for. The analysis shows that the temperature profile is sensitive to both h and the area considered for heat transfer. The value of h and the surface area to be considered should be determined from the actual furnace and freezer conditions (i.e., extent of forced circulation, positioning of the forms, etc.).

Table 1

TEMPERATURE PROFILE IN THE CYLINDER AT THE END OF 1 HOUR

Heating Cycle

T.ambient=60°C   T.initial=20°C

r (cm)	T (C)	
2.489	40.23	=T.surface
2.459	40.03	
2.428	39.82	
2.398	39.62	
2.367	39.42	
2.337	39.22	
2.220	39.02	
2.103	38.83	
1.986	38.65	
1.870	38.49	
1.753	38.33	
1.636	38.18	
1.519	38.04	
1.402	37.91	
1.285	37.80	
1.169	37.69	
1.052	37.59	
0.935	37.51	
0.818	37.43	
0.701	37.37	
0.584	37.31	
0.467	37.27	
0.351	37.24	
0.234	37.22	
0.117	37.21	
0.000	37.21	= T.center

Table 2

TEMPERATURE PROFILE IN THE CYLINDER AT THE END OF 1 HOUR

Cooling Cycle

T.ambient=-40°C   T.initial=20°C

r (cm)	T (C)	
2.489	-10.35	= T.surface
2.459	-10.04	
2.428	-9.73	
2.398	-9.43	
2.367	-9.12	
2.337	-8.83	
2.220	-8.53	
2.103	-8.25	
1.986	-7.98	
1.870	-7.73	
1.753	-7.49	
1.636	-7.27	
1.519	-7.06	
1.402	-6.87	
1.285	-6.70	
1.169	-6.54	
1.052	-6.39	
0.935	-6.26	
0.818	-6.15	
0.701	-6.05	
0.584	-5.97	
0.467	-5.91	
0.351	-5.86	
0.234	-5.83	
0.117	-5.81	
0.000	-5.81	= T.center

Table 3

SENSITIVITY ANALYSIS

Heating Cycle

$A0 = 116.0\ cm^2$ $Ta = 60^\circ C$
$AI = 154.9\ cm^2$ $Ti = 20^\circ C$

Exposure time (min)	AVERAGE TEMPERATURE (C)							
	h=0.5		h=1		h=2		h=3	
	A0	AI	A0	AI	A0	AI	A0	AI
30	2.9	23.8	25.6	27.3	30.4	33.2	34.5	38.1
60	25.6	27.3	30.4	33.2	38.0	42.0	43.7	48.0
90	28.1	30.4	34.5	38.1	43.7	48.0	49.6	53.4
120	30.4	33.2	38.0	42.0	48.0	51.9	53.4	56.4

Cooling Cycle

$A0 = 116.0\ cm^2$ $Ta = -40^\circ C$
$AI = 154.9\ cm^2$ $Ti = 20^\circ C$

Exposure time (min)	AVERAGE TEMPERATURE (C)							
	h=0.5		h=1		h=2		h=3	
	A0	AI	A0	AI	A0	AI	A0	AI
30	15.7	14.3	11.6	9.1	4.5	0.2	-1.7	-7.1
60	11.6	9.1	4.5	0.2	-7.1	-13.1	-15.6	-22.0
90	7.9	4.4	-1.7	-7.1	-15.6	-22.0	-24.4	-30.1
120	4.5	0.2	-7.1	-13.1	-21.9	-27.9	-30.1	-34.6

Appendix A

DATA

Property	Units	Symbol	Polymer	Polyethylene
Radius	cm	R	2.489 [1]	2.337 [1]
Length	cm	L	7.475 [1]	7.475 [1]
Density	gm/cm^3	ρ	0.95 [1]	0.95 [2]
Specific heat	cal/gm.C	Cp	0.47 [1]	0.55 [2]
Thermal conductivity	cal/cm.min.C	k	0.138 [1]	0.0468 [2]
Thermal diffusivity	cm^2/min	α	0.294 [1]	0.084 [2]

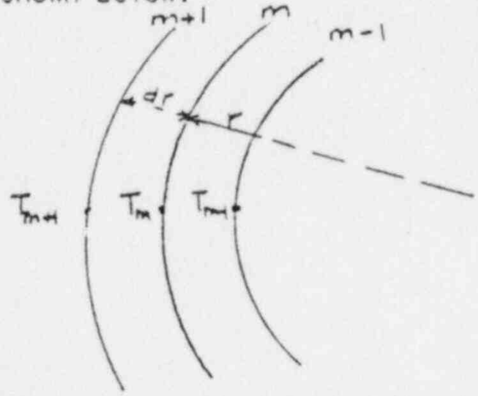
[1]Properties of Radioactive Wastes and Waste Containers, First Topical Report, Colombo, P., and Nielson, R. M., Jr., Aug. 1979.

[2]Chemical Engineers' Handbook, Perry, R. H., and Chilton, C. H., McGraw - Hill Book Company, Fifth Edition.

Appendix B

SOLUTION TO THE HEAT EQUATION

The solution to the heat equation (Eq. 2) by finite difference procedure results in the recursive relationship of Eq. 4 for the temperature. The same solution can be derived by a simple energy conservation approach. Consider the cylinder to be divided into elemental rings of width dr in the radial direction. At any time t, let us look at a typical mth elemental ring with inner radius r as shown below.



The temperature at the mth node depends on the temperatures at the two neighboring nodes, m+1 and m-1. At time t, let the temperatures at the respective nodes be T_{m+1}, T_m and T_{m-1}. The objective is to estimate the temperature profile in the cylinder at time t+dt.

$$\text{Total heat in at time dt} = k\, 2\pi(r+dr)L\, [T_{m+1} - T_m)/dr]\, dt$$

$$\text{Total heat in at time dt} = k\, 2\pi(r)L\, [T_m - T_{m-1})/dr]\, dt$$

$$\text{Total heat accumulated} = 2\pi r(dr)L\rho\, C_p\, [T_{m,n+1} - T_m]$$

where $T_{m,n+1}$ is the temperature at node m at time t+dt.

Since, Heat in - Heat out = Heat accumulated

we get, on simplification,

$$\boxed{T_{m,n+1} = [(1+N)T_{m+1} + (M-N-2)T_m + T_{m-1}] / M}$$

which is the same as Eq. 5 with M and N defined as below Eq. 5.

Appendix C

SOLUTION PROCEDURE

The steps in the procedure for numerical solution and the relevant programs are presented here.

1. Calculation of the time step, dt:

As mentioned earlier, the stability of the numerical method relies heavily on dt. For stability, dt should be chosen such that M [=(dr)(dr)/αdt] should be greater than 2. If the time step is made too large (to speed up the calculations), M will be too small, which in turn will lead to disaster. To avoid this, the following method was adopted:

(a) The polyethylene section was divided into 5 elements (arbitrary) and the polymer section into 20 elements (arbitrary). The thickness of elemental rings in each section was calculated by

$$dr1 = (R1 - R2)/5$$

and

$$dr2 = R2/20$$

where R1 = radius of the cylinder and R2 = radius of the polymer section.

(b) The time steps for each material were computed based on M=2.5,

$$dt1 = (dr1)(dr1)/2.5\alpha_1$$

and

$$dt2 = (dr2)(dr2)/2.5\alpha_2$$

(c) Choosing the lower of dt1 and dt2 as the time step dt ensured that the stability criteria is met for each material. Based on this dt, M was recomputed.

2. Initial temperature profile:

At time=0, the initial temperature throughout the entire cylinder was set at 20°C.

3. Next time step:

   The time at a new step was updated by, $t = t + dt$

4. Calculation of surface temperature:

   Based on Eq. 3, the surface temperature was calculated at the (n+1)th time step based on the relevant temperatures at the nth step.

5. Temperature profile:

   Once the surface temperature was known, the temperatures at the (n+1)th time step were calculated element by element using the recursive formula of Eq. 4.

6. For the next time step, the temperatures calculated in step 5 above serve as the temperatures at nth time step. With these temperatures, steps 3 to 6 are repeated until the desired cycle time is reached.

The LOTUS spreadsheet based on the above steps is shown in the next page.

LOTUS 1-2-3 WORKSHEET

HEAT CONDUCTION IN COMPOSITE CYLINDER

September 30, 1986

k'1	0.0468	r'1	2.489				
k'2	0.1380	r'2	2.337	T.avg	38.26		
Alpha1	0.084 (cm2/min)			T.center	37.21		
Alpha2	0.294						
		DELT	0.0044	Flag	1.000	a'1	0.011
DELT1	0.0044	DELT2	0.0093	Time	60.0	a'2	2.500
DELr1	0.030	DELr2	0.117	Time.max	60	Ntime	13634
n'1	5	n'2	20	T.max	60	hc	2
MM1	2.50	MM2	10.55	T.room	20	AREA	116

Element	r	N	Tn	T_n+1
1	2.489	81.00	40.23	40.23
2	2.459	80.00	40.03	40.03
3	2.428	79.00	39.82	39.82
4	2.398	78.00	39.62	39.62
5	2.367	77.00	39.42	39.42
6	2.337	76.00	39.22	39.22
7	2.220	19.00	39.02	39.02
8	2.103	18.00	38.83	38.83
9	1.986	17.00	38.65	38.66
10	1.870	16.00	38.49	38.49
11	1.753	15.00	38.33	38.33
12	1.636	14.00	38.18	38.18
13	1.519	13.00	38.04	38.04
14	1.402	12.00	37.91	37.92
15	1.285	11.00	37.80	37.80
16	1.169	10.00	37.69	37.69
17	1.052	9.00	37.59	37.60
18	0.935	8.00	37.51	37.51
19	0.818	7.00	37.43	37.43
20	0.701	6.00	37.37	37.37
21	0.584	5.00	37.31	37.32
22	0.467	4.00	37.27	37.27
23	0.351	3.00	37.24	37.24
24	0.234	2.00	37.22	37.22
25	0.117	1.00	37.21	37.21
26	0.000	0.00	37.21	37.21

MACROS

```
INITIALIZATION    \A   {goto}FLAG~0~{calc}  -- Set flag to 0. The
                                               temperatures are set
                                               to T.room.

COMMENCE
CALCULATIONS      \C   {goto}FLAG~1~{calc}  -- Flag is set to 1.

ITERATIVE
CALCULATIONS      \B   /rvTn+1~Tn~{calc}      -- Set Tn = T_n+1.
                       /xitime>=time.max~/xq -- If time=time.max, stop.
                       /xg\B                  -- Or else, continue.
```

Appendix D

APPROXIMATE ANALYTICAL SOLUTION

For the case where the temperature is uniform (i.e., high thermal diffusivity) within a cylinder of only one material , an approximate solution can be derived as follows.

Heat in by convection = hA(Ta - T_uni) (D1)

Heat accumlated = m(Cp) dT/dt (D2)

where m = mass of the cylinder (gm) T_uni is the uniform temperature.

Equating equations D1 and D2 and integrating over time, we get

(Ta - T_uni)/(Ta - T_ini) = exp[-(hA/m(Cp)) t] (D3)

where T_ini is the initial temperature.

If convection from ends is neglected, A = 116 cm^2.

h in cal/min.cm^2.C = (60/7380)x(h in Btu/hr.ft^2.F)

m = 138.2 gm, Cp = 0.47 cal/gm.C

and therefore,

(Ta - T_uni)/(Ta - T_ini) = exp[-0.0149ht] (D4)

where h is in Btu/hr.ft^2 .F.

Eq. D4 represents the situation where the temperature in the cylinder is uniform, i.e., where the thermal diffusivity is large. It was found that, for the problem at hand, using a thermal diffusivity three times lower provided an adequately good approximation. This is given by

$$\boxed{(Ta - T_{uni})/(Ta - T_{ini}) = \exp[-0.005ht]} \quad (D5)$$

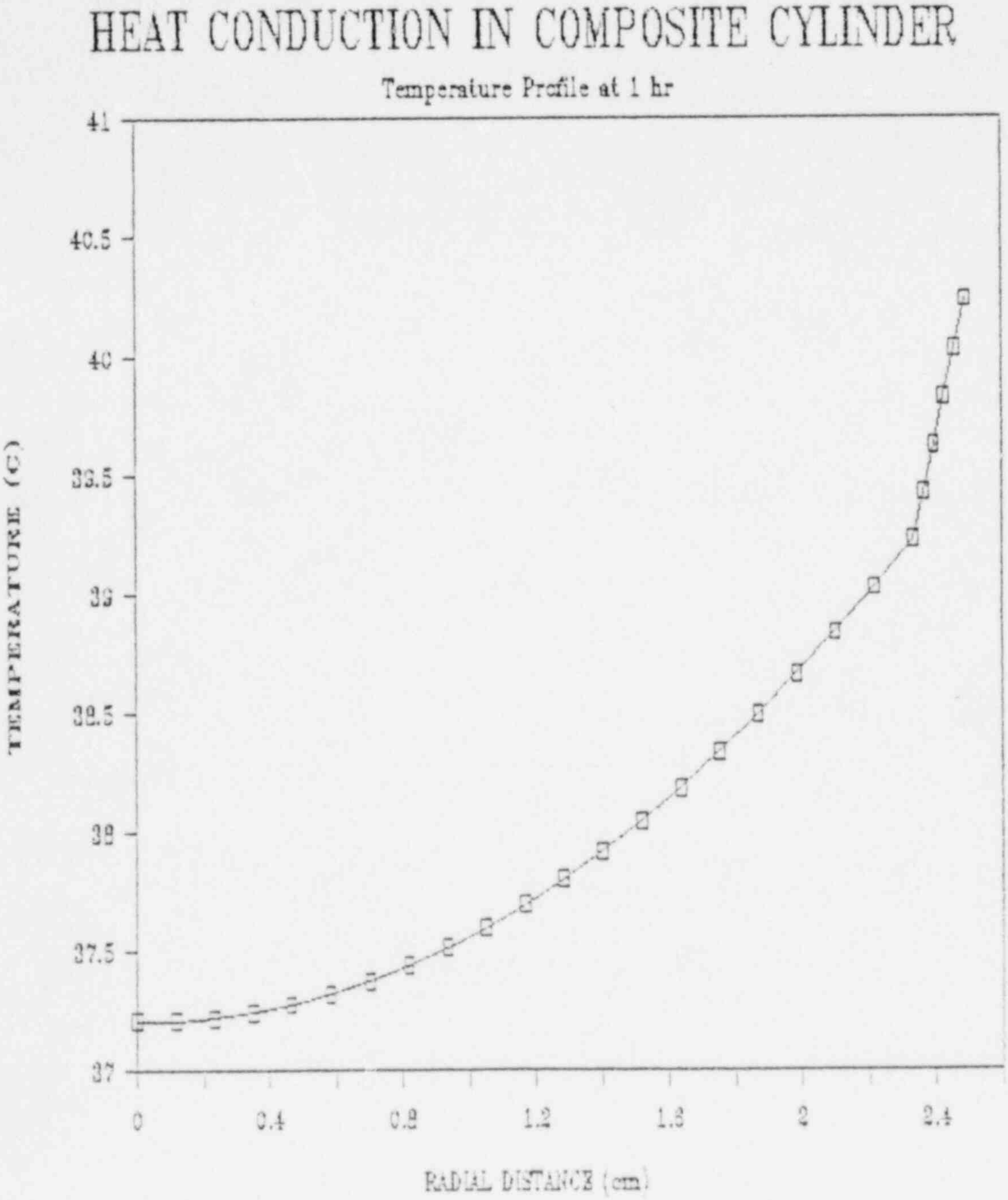


Figure 1: Temperature profile at the end of heating cycle.

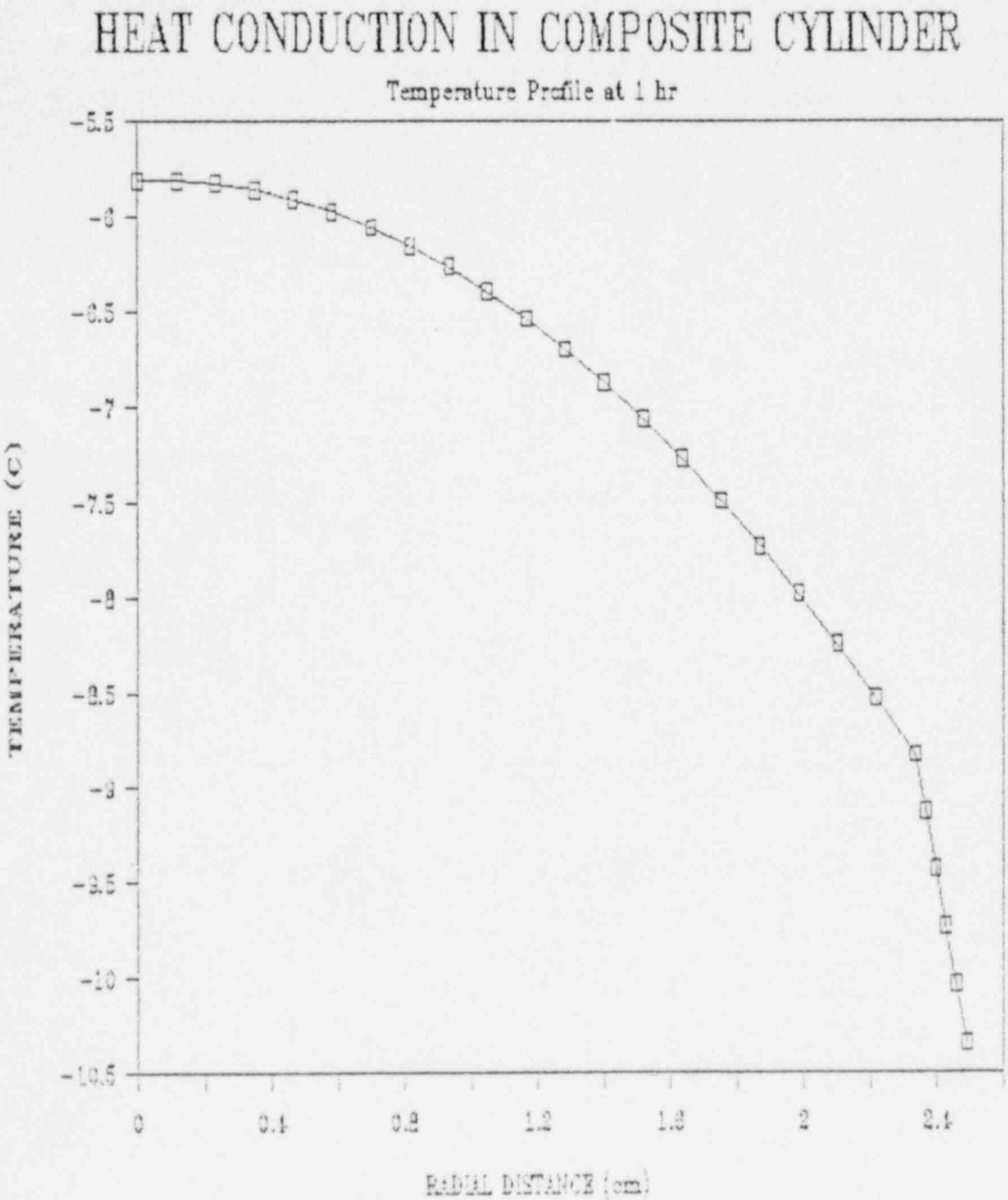


Figure 2: Temperature profile at the end of cooling cycle.

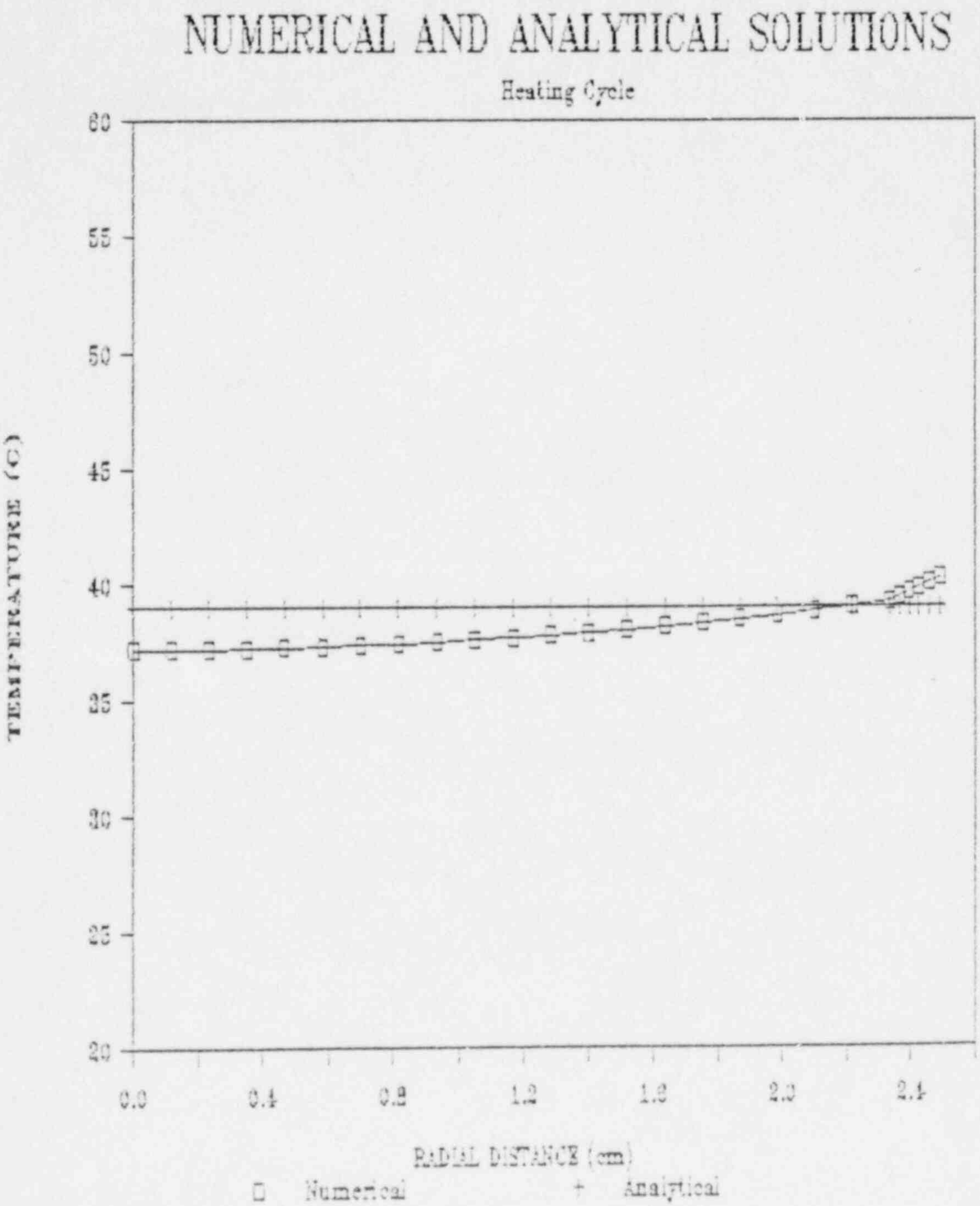


Figure 3: Comparison of numerical and approximate analytical solutions - Heating cycle

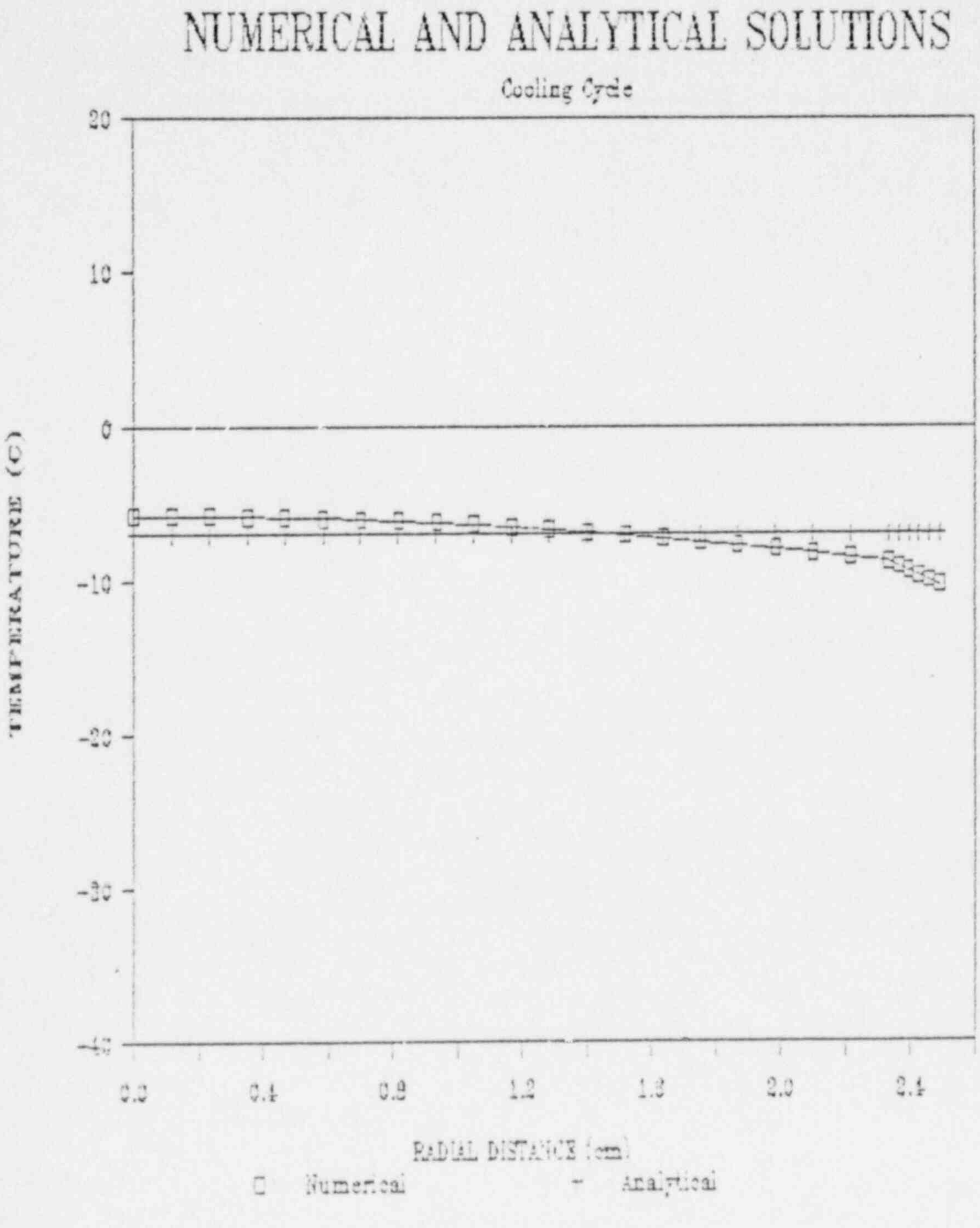


Figure 4: Comparison of numerical and approximate analytical solutions -Cooling cycle