# Modeling the Iodine Removal Efficiency and Temperature Behavior for a FADS Charcoal Filter by FEMLAB

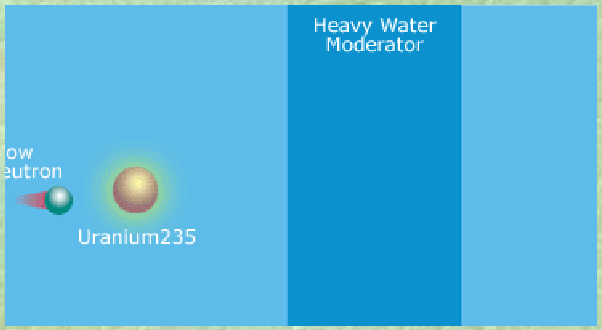
Zack Qin and J. Clara Wren

The University of Western Ontario, London, Ontario, Canada

C. J. Moore

Chalk River Laboratories, Atomic Energy of Canada Limited, Chalk River, Ontario, Canada

# Nuclear Energy




- It is a clean energy source that does not produce global warming gases.
- It is safe. The effects of nuclear power generation on public health are actually less than the alternative sources.
- It is affordable, comparing favourably with the average cost of electricity generated by coal, gas or oil.
- There were 441 nuclear plants in 32 countries supplying about 21.2% of the world's electricity (International Atomic Energy Agency, 2002).
- Nuclear energy produced 16% of the electricity in Canada, 50.2% in Ontario (Ontario Power Generation, 2004).

# CANDU (CANada Deuterium Uranium)

# Filtered Air Discharge System (FADS)

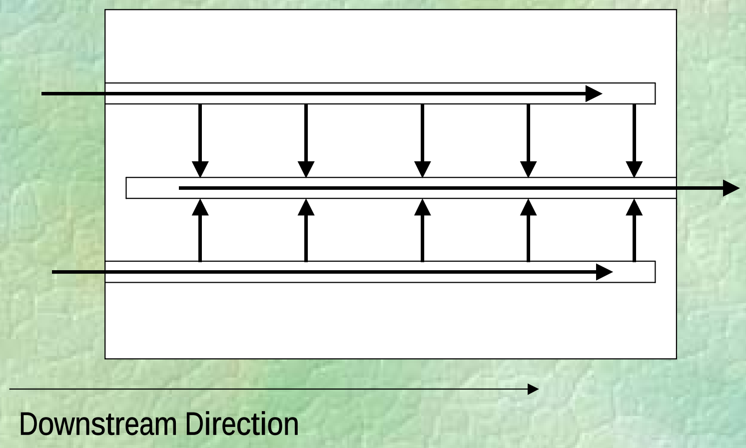
- A significant safety feature of multi-unit CANDU nuclear power stations.
- The functions of the FADS are to maintain the containment pressure sub-atmosphere and to control the release of gaseous fission products from containment to the environment during the long-term management phase of a reactor accident.
- FADS components:
  Roughing filter → Demister → Heater → HEPA filter → Charcoal filter → HEPA filter → Blower
- These filters are isolated from containment during normal operation by double isolation valves, but would be exposed to a very fast airflow in the unlikely event of an accident.

# FADS Charcoal Filters

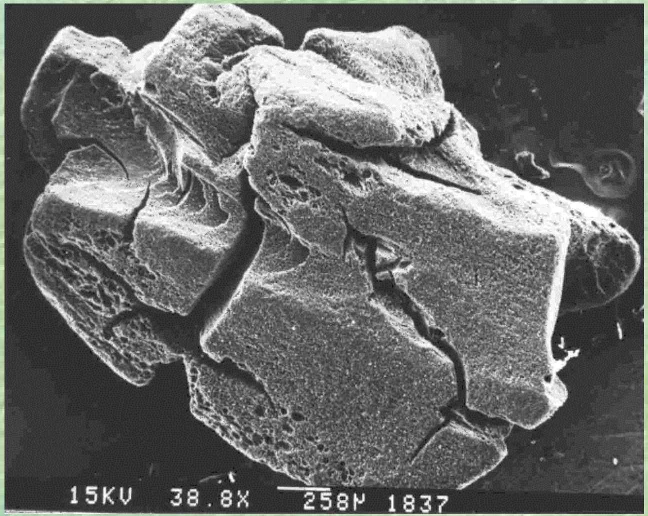
- One of the key components of the FADS
- It is intended to remove gaseous fission products, mainly radioiodine, by adsorption onto charcoal.

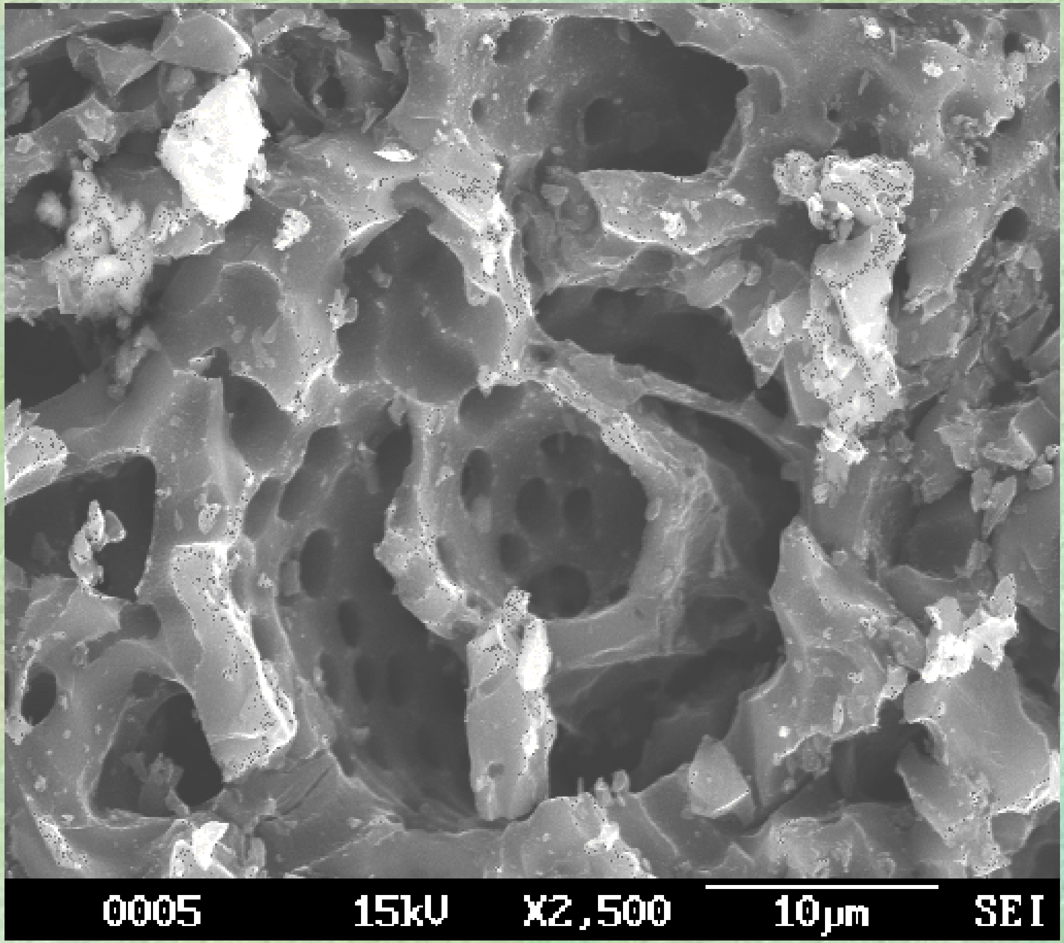
20 cm deep

~10 $m^2$ total cross section



# Highly Activated Charcoal




Grain size: ~ 8 × 10 mesh

Surface area: ~ 1000 $m^2/g$

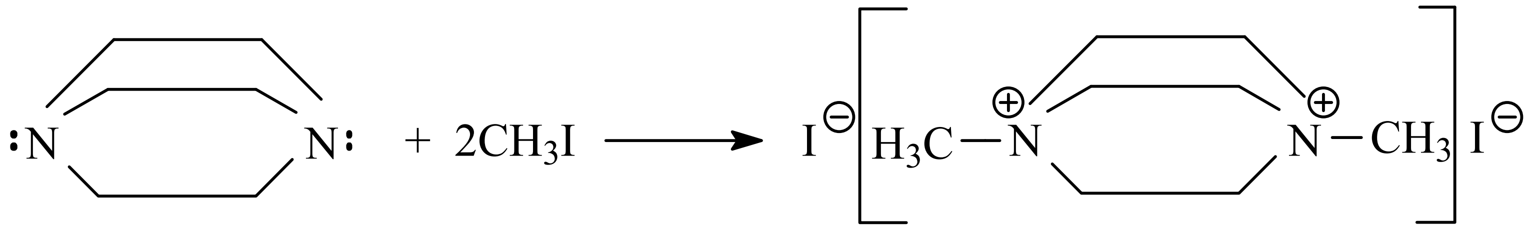
Pore size: ~ 7.5 - 8.5 Å

Pore volume: ~ 0.3 $cm^3/g$

TEDA content: 1~ 5% wt

# TEDA Impregnated Charcoal

- The charcoal is impregnated with triethylenediamine (TEDA) to increase trapping efficiency for organic iodides through the formation of an ammonium iodide complex.




$$:N(CH_2CH_2)_3N: \; + \; 2\,CH_3I \;\rightarrow\; I^{\delta-}\,[CH_3N^{\delta+}(CH_2CH_2)_3N^{\delta+}CH_3]\,I^{\delta-}$$

(TEDA) (Methyl iodide) (Ammonium Salt)

- $CH_3I$, as the most penetrating organic iodide species expected in containment, represents the most challenge to remove.
- The ammonium iodide complex is very stable, and its vapor pressure very low.

# Main Issues of Charcoal Filter

1. **Removal efficiency**

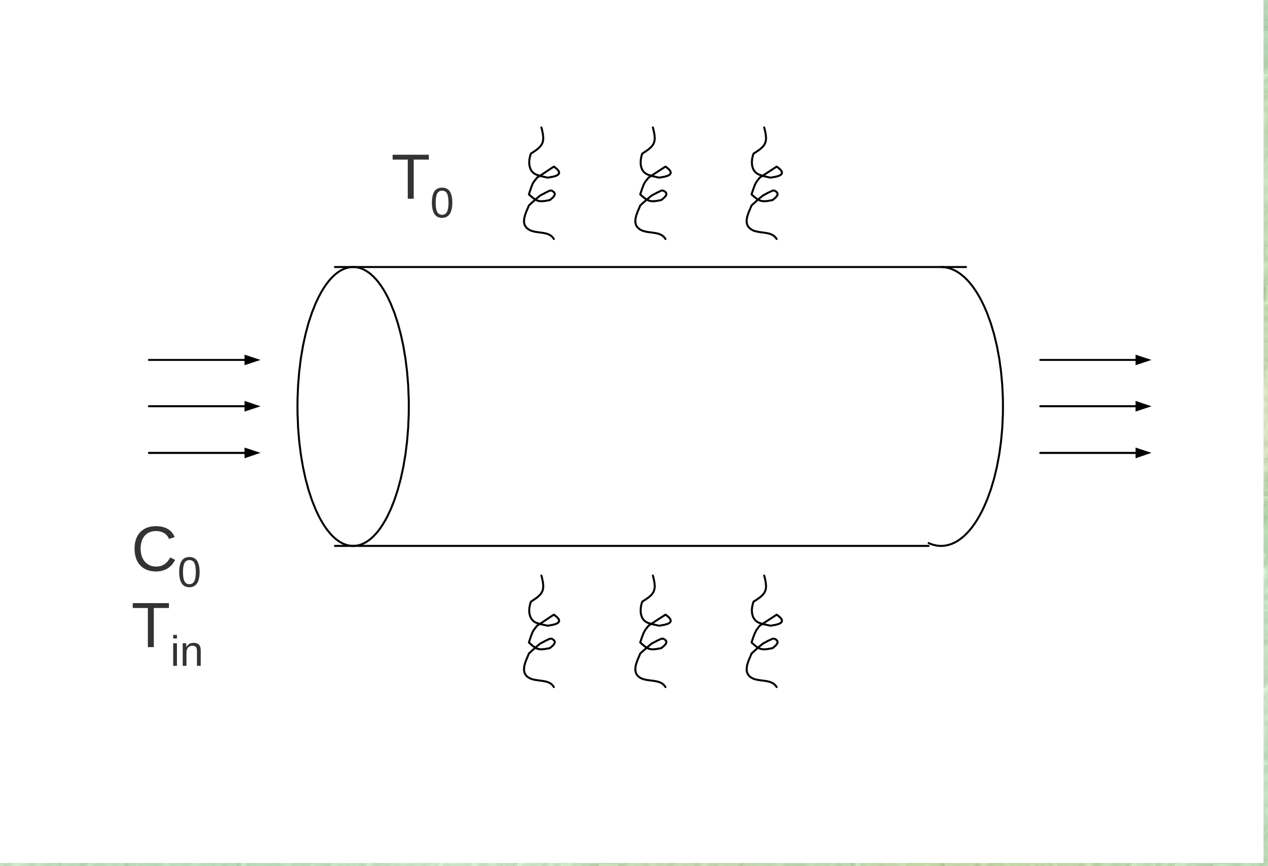
   The effectiveness of the charcoal filter for removing and retaining gaseous radioiodine under postulated accident conditions must far exceed that credited in safety analysis (>99.9%).

2. **Transient temperature behavior**

   The charcoal temperature may rise on the start-up of the FADS due to the exothermic absorption of water when cool charcoal is exposed to a warm, humid airflow, which may result in the degradation of removal efficiency, or cause other safety problem concerns.

# Mathematical Models

1. Iodine Removal model – *Removal*
2. Transient temperature model - *DryAir*



# Iodine Removal Model - *Removal*

- The removal and trapping of $CH_3I$ from flowing airstreams depend on the diffusion and convection of $CH_3I$ in the airflow, and on the adsorption/desorption of $CH_3I$ onto/from the charcoal.

$$\frac{\partial}{\partial t}C(\mathbf{r},t) = D\nabla^2 C(\mathbf{r},t) - \mathbf{v}\cdot\nabla C(\mathbf{r},t) - \frac{\varepsilon}{\sigma_v}\cdot\frac{\partial}{\partial t}W(\mathbf{r},t)$$

- Adsorption of $CH_3I$ on TEDA-impregnated charcoal occurs through reversible physisorption on charcoal surfaces, and through irreversible chemisorption on TEDA.

$$W(\mathbf{r},t) = W_{PHYS}(\mathbf{r},t) + W_{CHEM}(\mathbf{r},t)$$

# Adsorption Rate Equations

**physisorption** (reversible)

$$\frac{\varepsilon}{\sigma_v} \cdot \frac{\partial}{\partial t} W_{PHYS}(\mathbf{r},t) = k_{PHYS}^{A} \cdot C(\mathbf{r},t) - k_{PHYS}^{D} \cdot \frac{\varepsilon}{\sigma_v} \cdot W_{PHYS}(\mathbf{r},t)$$

**Chemisorption** (irreversible)

$$\frac{\varepsilon}{\sigma_v} \cdot \frac{\partial}{\partial t} W_{CHEM}(\mathbf{r},t) = k_{CHEM}^{A}(\mathbf{r},t) \cdot \left( \alpha\, C(\mathbf{r},t) \; + \; (1\text{-}\alpha) \frac{\varepsilon}{\sigma_v} \cdot W_{PHYS}(\mathbf{r},t) \right)$$

$$k_{CHEM}^{A}(\mathbf{r},t) = k_{CHEM}^{o} \cdot \left( 1 - \frac{W_{CHEM}(\mathbf{r},t)}{W_{CHEM}^{o}} \right)$$

# Boundary Conditions (*Removal*)

- Inlet

$$C = \begin{cases} C_0 & (t \le t_{load}) \\ 0 & (t > t_{load}) \end{cases}$$

- Outlet

$$\hat{n} \cdot (-D \nabla C) = 0$$

- Wall

$$\hat{n} \cdot (-D \nabla C + C \cdot \mathbf{v}) = 0$$

- Initial

$$C = 0$$

# Temperature Model - *DryAir*

- This temperature model describes the heat transfer behavior of the filter that is exposed to a dry airflow.
- The exposure to a dry airflow provides a simpler heat transfer system than a humid airflow system because no mass transfer and exothermic reactions are involved.
- The current model will be extended to a charcoal filter exposed to a humid airflow later.

**Assumptions:**

i. The charcoal and air are in thermal equilibrium.
ii. The porous medium is homogeneous.
iii. Airflow is uniform without detailed fluid dynamics at the pore level.

# Heat Transfer Equation

$$\rho_e C_P^e \frac{\partial T}{\partial t} - \kappa_e \nabla^2 T + \rho_e C_P^e \mathbf{v}_e \cdot \nabla T = 0$$

$$\begin{cases} \rho_e \equiv \sigma_v \rho_a + (1-\sigma_v)\rho_c \\ \kappa_e \equiv \sigma_v \kappa_a + (1-\sigma_v)\kappa_c \\ C_P^e \equiv \dfrac{\sigma_v \rho_a C_P^a + (1-\sigma_v)\rho_c C_P^c}{\rho_e} \\ \mathbf{v}_e \equiv \dfrac{\sigma_v \rho_a C_P^a}{\rho_e C_P^e} \mathbf{v}_a \end{cases}$$

# Boundary Conditions (*DryAir*)

- Inlet

$$T = \begin{cases} T_{in} & (t \le t_{off}) \\ T_0 & (t > t_{off}) \end{cases}$$

- Outlet

$$\hat{n} \cdot \kappa_e \nabla T = 0$$

- Wall

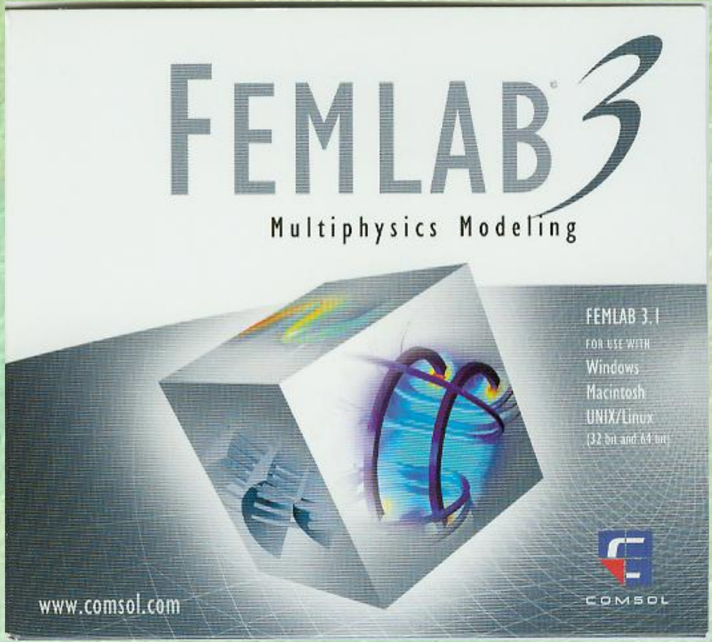
$$\hat{n} \cdot \kappa_e \nabla T = h_s (T_0 - T)$$

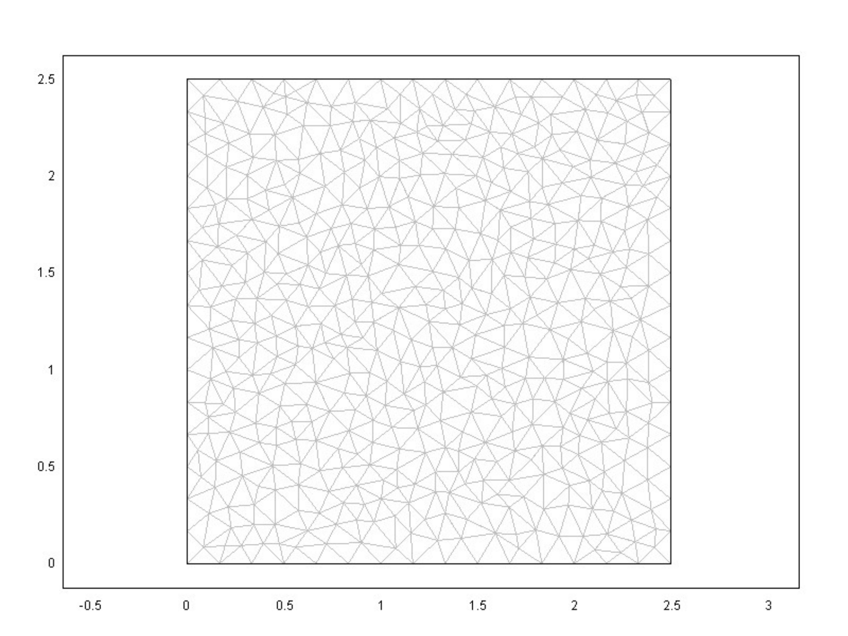
- Initial

$$T = T_0$$

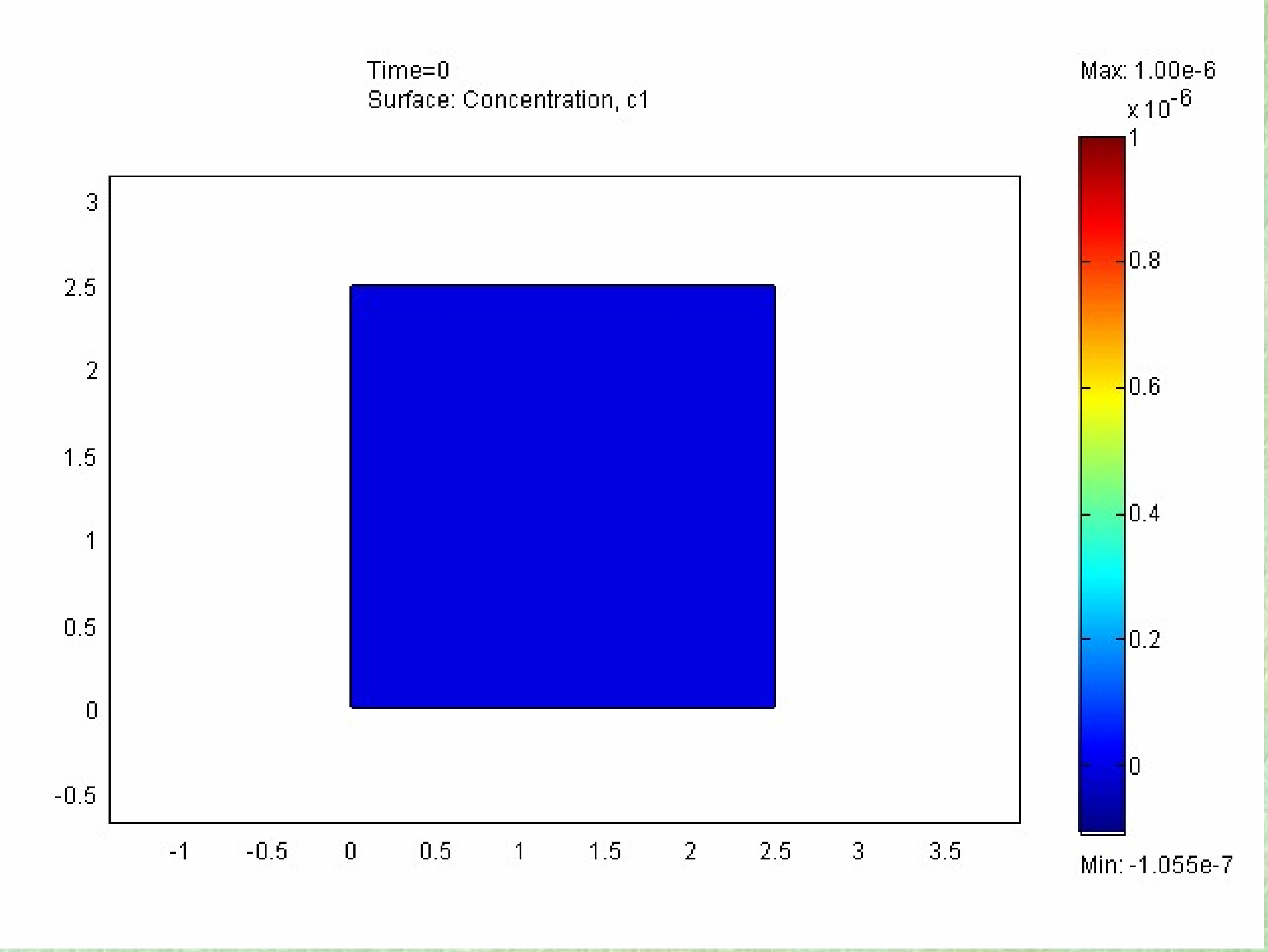
# FEMLAB Simulation

- The mathematical models are difficult to solve analytically.
- Solutions can be found numerically by the finite element method using the software package FEMLAB.
- *Removal* – diffusion and convection application mode; *DryAir* – convection and conduction application mode.



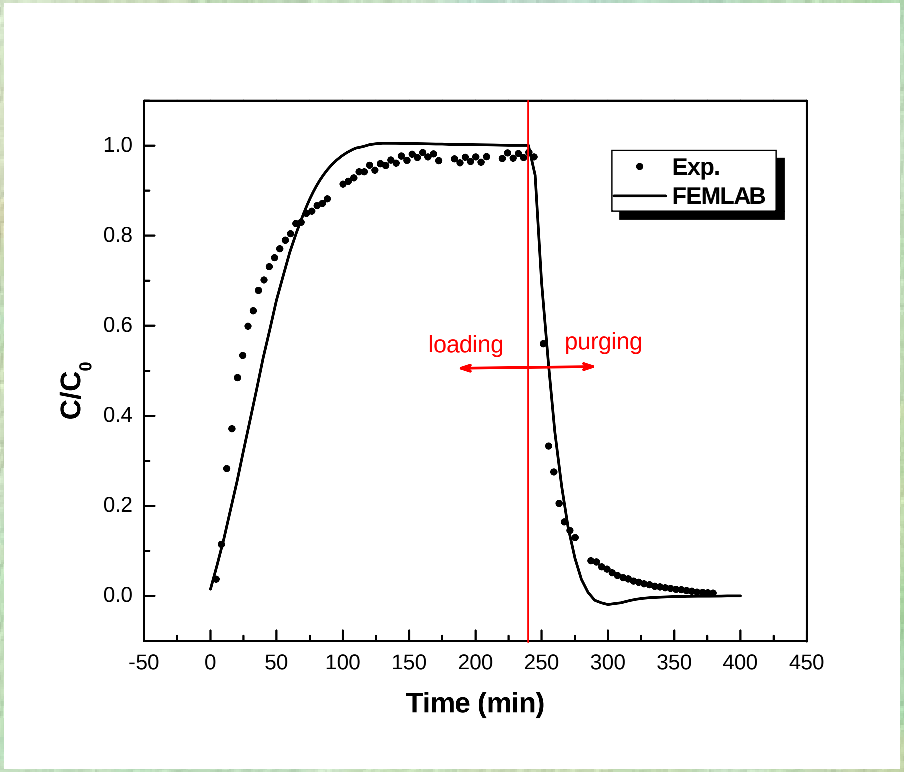



# Simulation by *Removal*

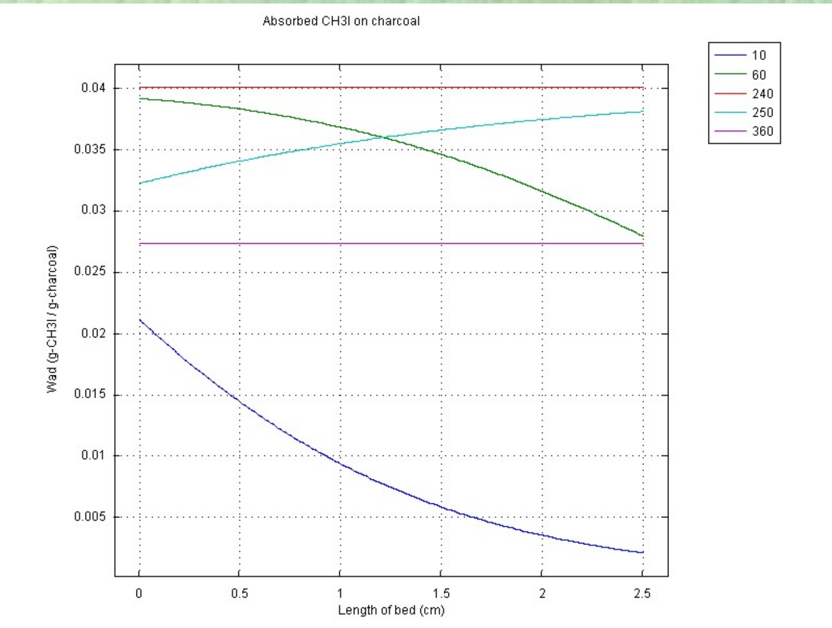


# Comparison with Experimental Data

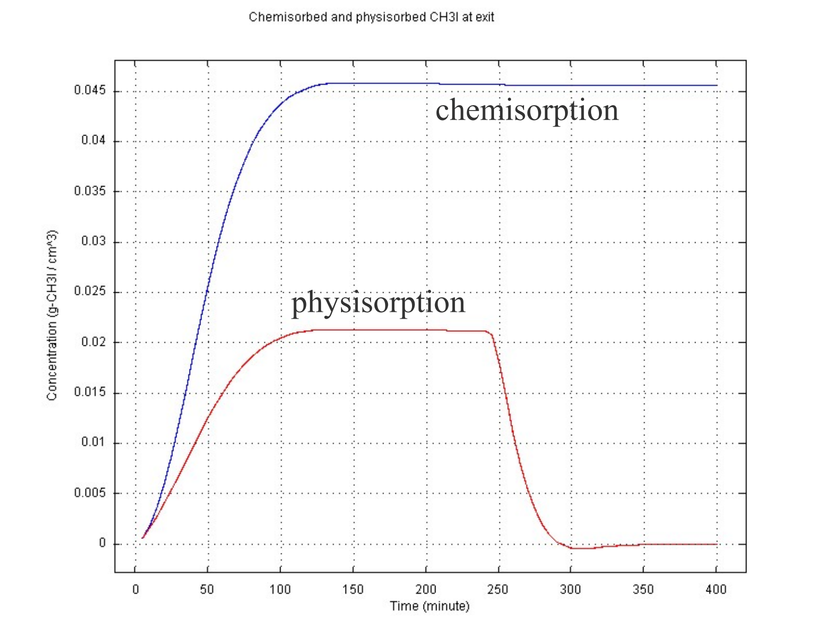
- Experiment (Chalk River Lab, AECL):
  loaded with 1mg/L $CH_3I$ airflow until the bed was saturated, then purged with air, free of $CH_3I$.
- The simulation by *Removal* qualitatively reproduced the experimental breakthrough curve.



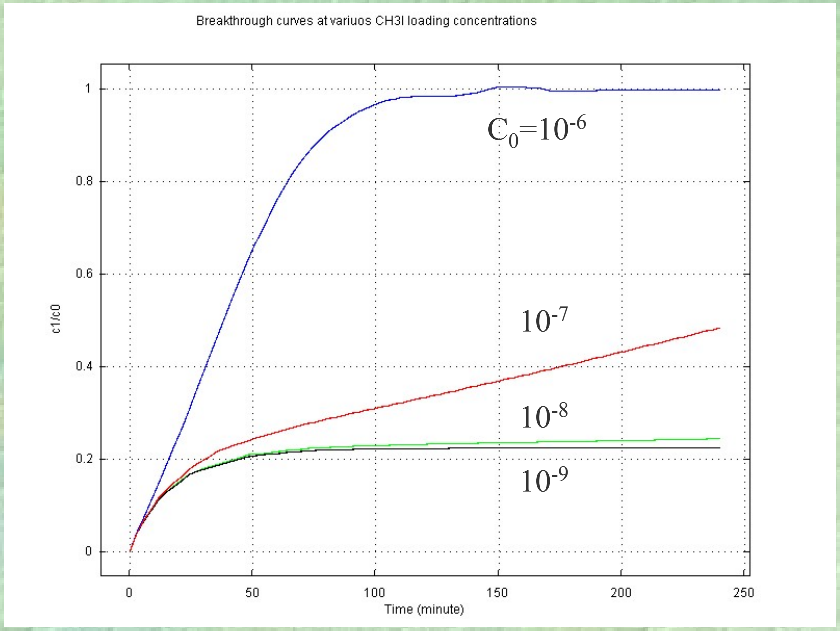
The distributions of absorbed $CH_3I$ on charcoal change with time along the length of the bed



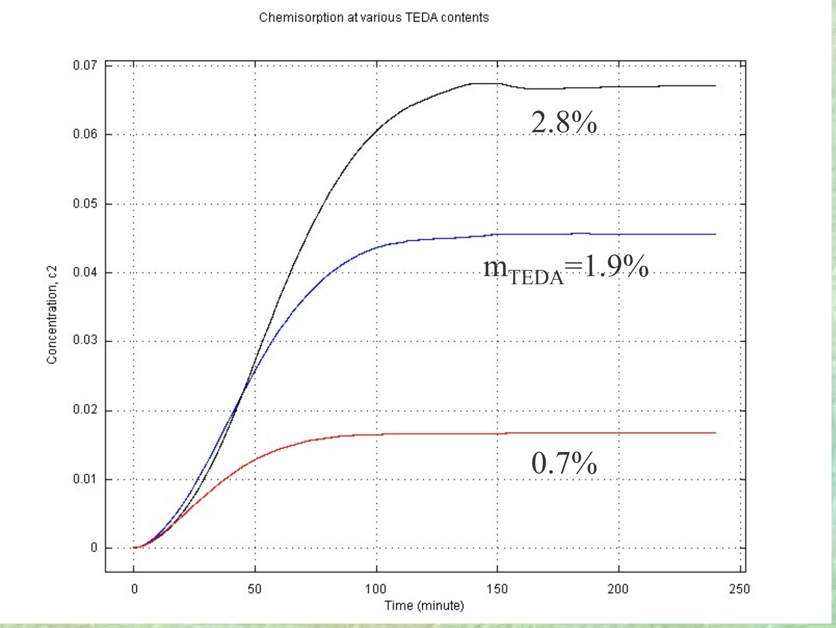

The physically absorbed $CH_3I$ releases quickly after purging while the chemisorbed $CH_3I$ retains on charcoal



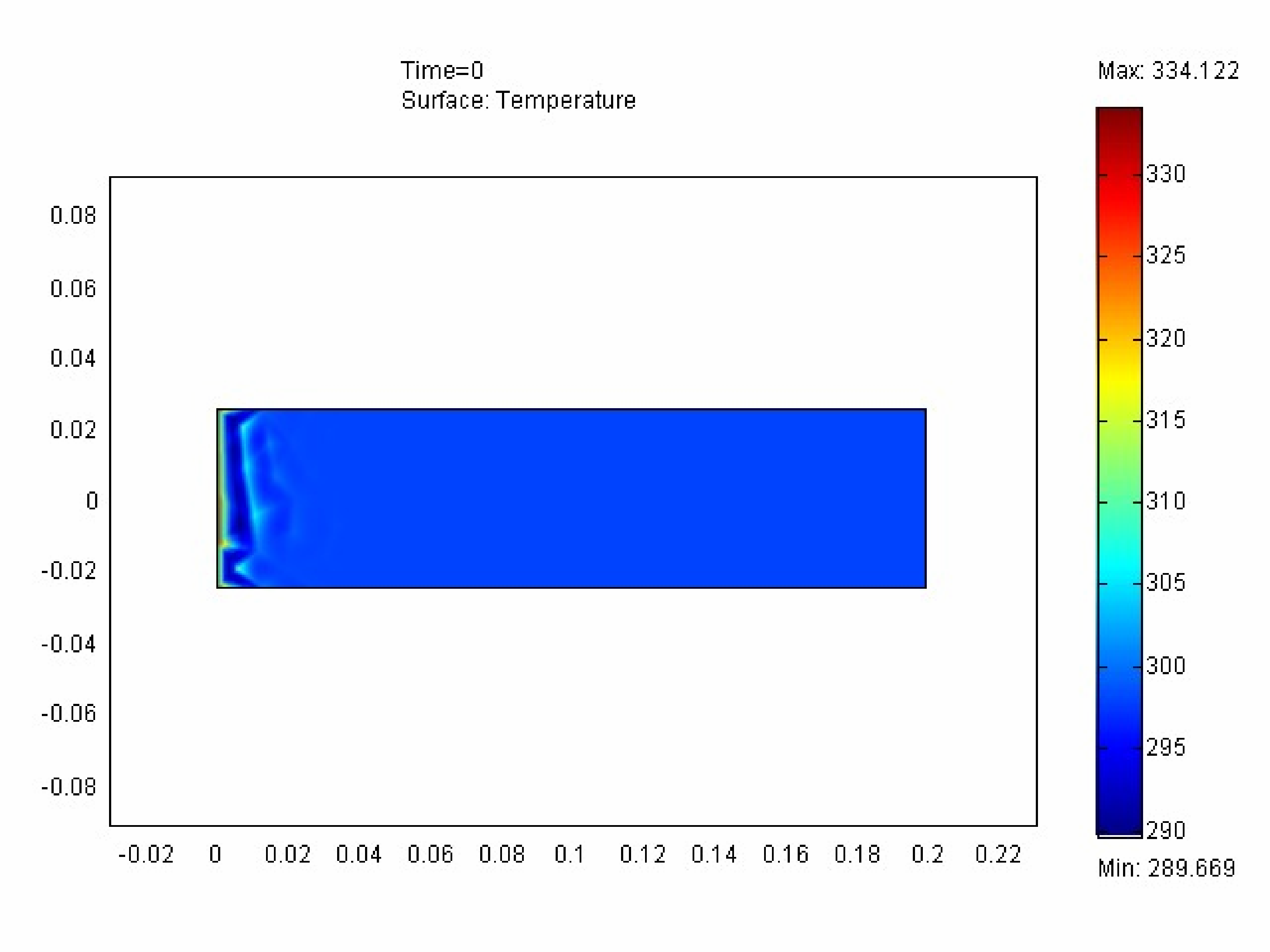
The higher the loading $CH_3I$ concentration, the sooner the bed becomes saturated.




Chemisorption depends on TEDA contents



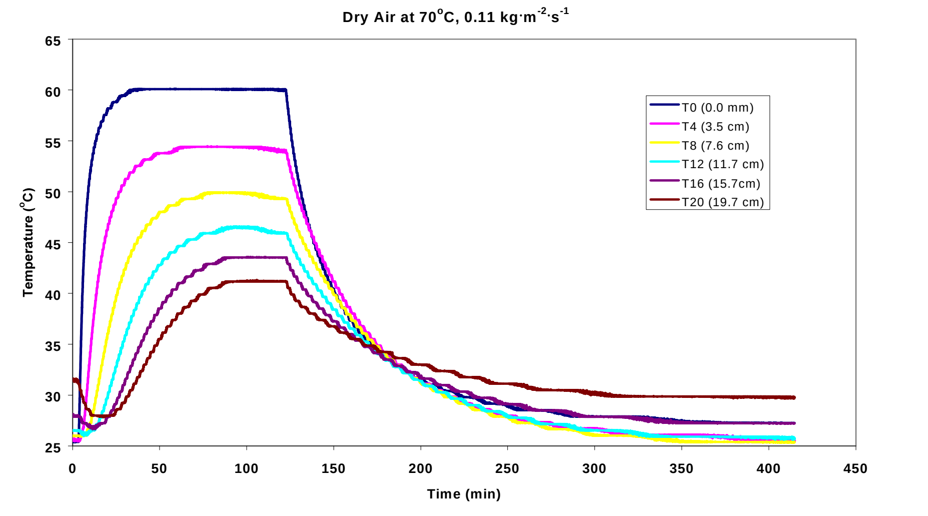
# Simulation by *DryAir*

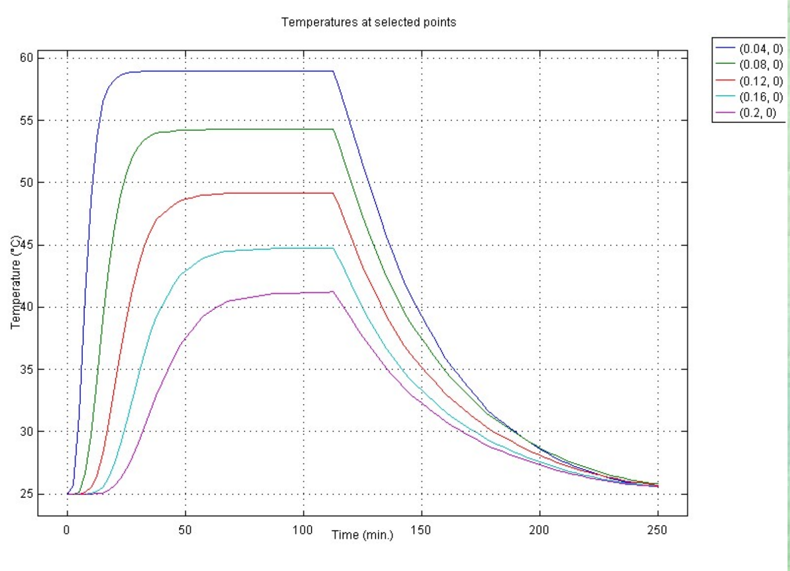


# Comparison with Experimental Data

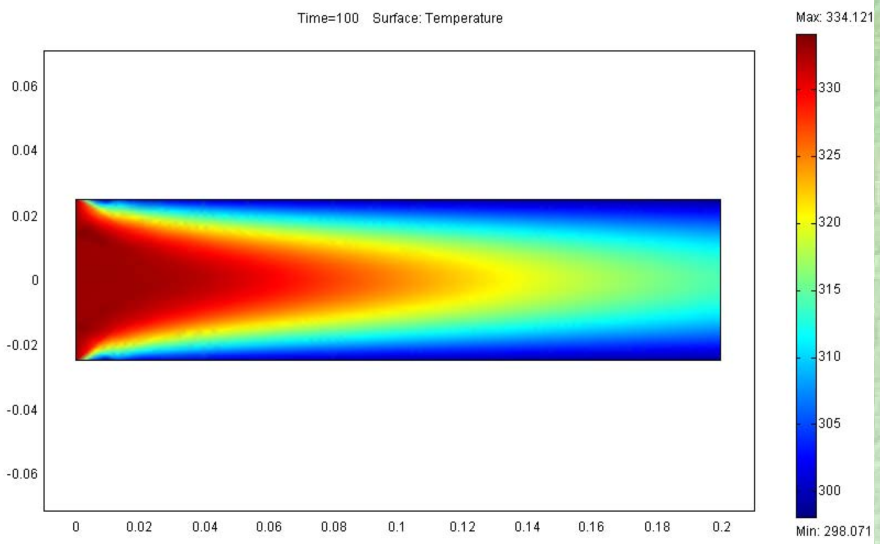
- Experiment (Chalk River Lab, AECL) exposed to a warm dry airflow for 2 hours, then cooled by heat exchange with the surroundings.
- The simulation by DryAir well reproduced the temperature profile measured along the length of the bed.
- The small heating source, such as the humidity probe, existed in the experiment was not modeled.

Experiment (Chalk River Lab, AECL)

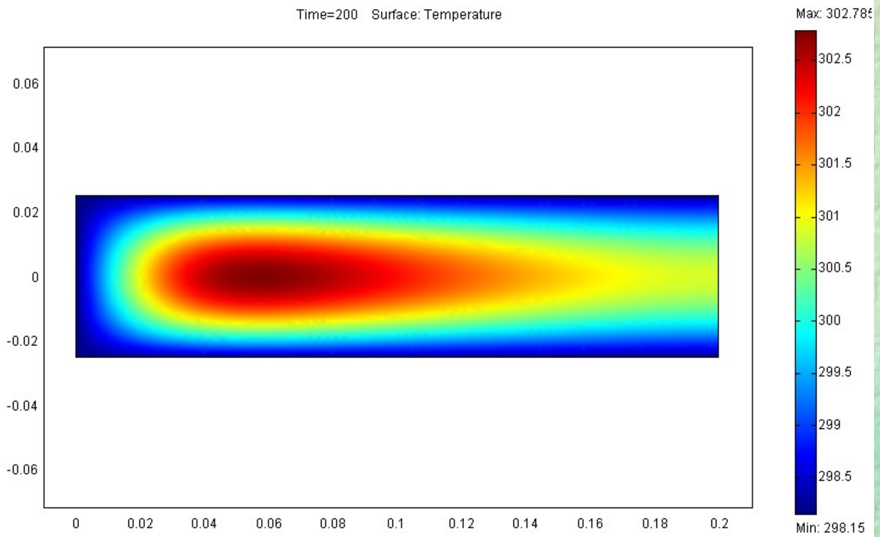


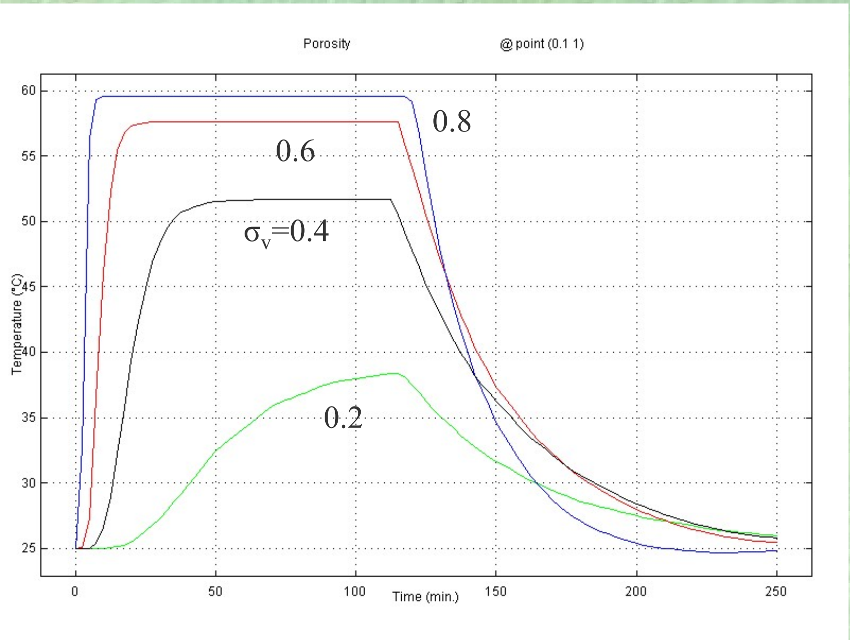
Temperature profile after 100 minutes exposed to a warm and dry airflow



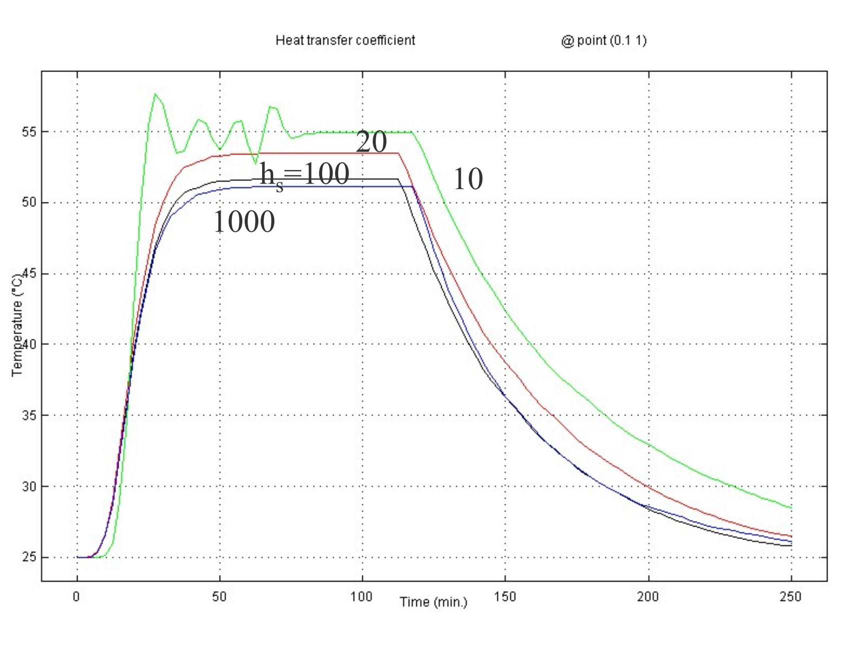

At 200 minutes when airflow is off at 120 minutes



The higher the porosity, the higher the equilibrium temperature and the sooner to achieve the equilibrium temperature




The wall heat transfer coefficient values greater than 100 would not lead to significantly more heat loss



# Conclusions

- The efficiency of TEDA-impregnated charcoal of the FADS filter for removing $CH_3I$ was modeled according to the diffusion and convection of $CH_3I$ in airstreams, and to the adsorption and desorption of $CH_3I$ on/from charcoal.
- The transient temperature model based on fast thermal equilibrium between air and charcoal was developed for a FADS charcoal filter when it is exposed to a dry airflow.
- The models were numerically solved by FEMLAB.
- The results from the FEMLAB simulation reproduced experimental data over a wide range of conditions.
- The influence of some parameters, which are difficult to evaluate experimentally, was also investigated by simulation.
- The current models will be refined based on these findings.

## Acknowledgements

- Partially funded by CANDU Owners Group R&D program, Containment Behavior, under the joint participation of Atomic Energy Canada Limited (AECL), Ontario Power Generation, Bruce Power, Hydro Quebec and New Brunswick Power.
- Helpful discussions with Dr J.M. Ball at AECL and Dr. D.W. Shoesmith at the University of Western Ontario.


Western
Leading. Thinking.

# Thank You