Sound absorption modelling of granular activated carbon

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Outline

- Motivation
- Dynamic bulk modulus of granular activated carbon (GAC)
- Wave equation upscaling including sorption and rarefaction effects
- A model for sound propagation in GAC
- Acoustic characterisation of GAC
- Sound absorption of GAC - measurements vs predictions
- Future work and conclusions
Granular activated carbon (GAC) has been experimentally shown to exhibit unusually high levels of low frequency sound absorption.
Dynamic bulk modulus of granular activated carbon

\[ \frac{K(\omega \to 0)}{P_0} = \frac{1}{\phi} > 1 \]

\[ \frac{K_{AC}(\omega \to 0)}{P_0} < 1 \]

- Rarefaction effects

- Finite heat capacity of the solid
  [Lafarge et al., JASA 102 (4), 1997]

- Mass transfer processes
  [Raspet et al., JASA 105(1), 1999]

- Particle vibration
  [Okudaira et al. Powder Technol. 77(1), 1993]

SORPTION PROCESSES


[Bechwati et al. JASA 132(1), 2012]

[Mellow et al., Proc of Acoustics 08, CD01. 795, 2008]
Sorption processes = adsorption + desorption

- Adsorption is a physical or chemical process in which the fluid molecules are adhered on to a surface.
  - The adherence in physical adsorption is caused by weak van der Waals forces while by chemical bonding in chemical adsorption.
- Desorption is the opposite phenomenon whereby the fluid molecules are released from the surface.
  - The release of the molecules is caused by either an increase of temperature or a decrease in pressure.

- Based on equating the rates of adsorption and desorption.

- Assumptions:
  - The surface of the solid is homogenous - adsorption energy is constant over all sites of the surface.
  - Adsorption is localised - molecules are adsorbed at definite, localised sites (no mobile adsorption).
  - Each site can accommodate only one molecule - monolayer coverage.

\[
\frac{d\tilde{\rho}_a}{dt} = k_a \tilde{p}(\tilde{\rho}_N - \tilde{\rho}_a) - k_d \tilde{\rho}_a
\]

\[
\rho_a = \frac{k_a k_d \rho_N}{(j\omega + \omega_a)\omega_a} p
\]

\[
\omega_a = k_a P_0 + k_d
\]

\(\rho_N\): maximum density increment
\(k_a\): adsorption coefficient
\(k_d\): desorption coefficient
\(\omega_a\): sorption characteristic frequency
Rarefaction effects

- Continuum description starts to break down when the characteristic size of the material is comparable to the mean free molecular path.

- No-slip condition is replaced by slip boundary condition - velocity is proportional to the tangential shear stress.

- Zero temperature boundary condition is replaced by the temperature-jump boundary condition - temperature is proportional to the normal temperature gradient.

\[ l_{\text{mean}} = \frac{\eta}{p} \sqrt{\frac{\pi R_g \tau}{2}} \]

\[ u = -c_v l_{\text{mean}} \left( t_1 \cdot (\nabla u) \cdot n \right) t_1 \]

\[ \tau = c_t \frac{2\gamma}{(\gamma + 1) N_{pr}} l_{\text{mean}} \nabla \tau \cdot n \]
Linearised equations

Conservation of momentum

\[ \eta \nabla^2 \mathbf{u} - \nabla p = j \omega \rho_0 \mathbf{u} \]

Conservation of mass

\[ j \omega (\rho + \rho_a) + \rho_0 \nabla \cdot \mathbf{u} = 0 \]

Conservation of energy

\[ j \omega \rho_0 C_p \tau = \kappa \nabla^2 \tau + j \omega p \]

Equation of state

\[ \frac{p}{P_0} = \frac{\tau}{\tau_0} + \frac{\rho}{\rho_0} \]

Langmuir equation

\[ \rho_a = \frac{k_a k_d \rho_N}{(j \omega + \omega_a) \omega_a} p \]

Slip and temperature-jump boundary conditions

\[ \mathbf{u} = -c_v l_{mean} (t_1 \cdot (\nabla \mathbf{u}) \cdot \mathbf{n}) t_1 \]

\[ \tau = c_t \frac{2 \gamma}{(\gamma + 1) N_{pr}} l_{mean} (\nabla \tau \cdot \mathbf{n}) \]
Dimensionless equations

Conservation of momentum
\[ \varepsilon^2 \eta \nabla^2 u \quad \nabla p = j \omega \rho_0 u \]

Conservation of mass
\[ j \omega \left( \rho + \frac{k_a k_d \rho_N}{(j \omega + \omega_a) \omega_a} p \right) + \rho_0 \nabla \cdot u = 0 \]

Conservation of energy
\[ \varepsilon^2 \kappa \nabla^2 \tau = j \omega \rho_0 C_p \tau - j \omega p \]

Equation of state
\[ \frac{p}{P_0} = \frac{\tau}{\tau_0} + \frac{\rho}{\rho_0} \]

Slip and temperature jump

boundary conditions
\[ u = -\varepsilon Kn (t_1 \cdot (\nabla u) \cdot n) t_1 \]
\[ \tau = \varepsilon \frac{2 \gamma Kn}{(\gamma + 1) N_{pr}} (\nabla \tau \cdot n) \]

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Wave equation including rarefaction and sorption effects

\[ \frac{j \omega}{K(\omega, Kn, S)} p^{(0)} = \frac{k(\omega, Kn)}{\eta} \Delta_x p^{(0)} \]

\[ K(\omega, Kn, S) = \left( \frac{1}{K_R(\omega, Kn)} + \frac{1}{K_S(\omega, S)} \right)^{-1} \]

\[ K_S(\omega) = \frac{\gamma P_0}{\phi} \left( \frac{\gamma P_0 k_a k_d}{(j \omega + \omega_a) \omega_a} \frac{\rho_N}{\rho_0} \right)^{-1} \]

\[ \omega_a = k_a P_0 + k_d \]

- The overall flow is not affected by sorption processes.
- The dynamic bulk modulus is modified by sorption processes.

\[ \frac{K(\omega \to 0)}{P_0} = \frac{1}{\phi} \cdot \frac{1}{1 + \frac{P_0 k_a k_d}{\omega_a^2} \frac{\rho_N}{\rho_0}} \]

\( \rho_N \): maximum density increment
\( k_a \): adsorption coefficient
\( k_d \): desorption coefficient
\( \omega_a \): sorption characteristic frequency

R. Venegas and O. Umnova, JASA, to be submitted
Model for sound propagation in granular activated carbon

Model published in [Venegas & Umnova, JASA 130 (5), 2011] is extended to account for an additional scale, and sorption and rarefaction effects.

High permeability contrast between the mesoscopic and microscopic and nanoscopic scale.

\[ k_{tb}(\omega) \approx k_p(\omega) \]

Low permeability contrast between the microscopic and nanoscopic scale.

\[ K_u(\omega, K_n, S) = \left( \frac{1}{K_m} + \frac{(1-\phi_m)}{K_n} \right)^{-1} \]

\[ K_{tb}(\omega \rightarrow 0) = \frac{1}{\phi_{tb}} \left( \frac{1}{\omega_n^2} \right) \]
Model for sound propagation in GAC

- Granular activated carbon is modelled as a packing of spherical porous grains
- Two inner-particle scales (e.g. microscopic and nanoscopic scales) are both modelled as arrays of cylindrical pores

\[ k_{tb}(\omega) \approx k_p(\omega) \]  
[Boutin & Geindreau, JASA, 124 (6), 2008]

\[ \frac{1}{K_{tb}}(\omega, Kn, S) = \left( \frac{1}{K_p} + \frac{(1-\phi_p)}{K_u} F_d \left( \frac{P_0}{\phi_u K_u} \right) \right)^{-1} \]  
[Boutin & Geindreau, Phys. Rev. E 82, 2010]

\[ K_u(\omega, Kn, S) = \left( \frac{1}{K_m} + \frac{(1-\phi_m)}{K_n} \right)^{-1} \]  
[Kozlov et al. JASA, 117, 2005]

\[ [\text{Venegas & Umnova, JASA 130 (5), 2011}] \]

\[ [\text{Boutin & Geindreau, JASA, to be submitted}] \]
Characterisation of GAC – particle radius - optical granulometry

\[ r_p = 0.7535 \pm 0.1609 \text{ mm} \]

\[ r_p = 0.7363 \pm 0.2056 \text{ mm} \]

R. Venegas and O. Umnova, JASA, to be submitted

- Flow resistivity measured using BS EN 29053:1993

- Mesoporosity calculated by inverting:

\[ k_0 = \left( \frac{r^2}{3\beta^2} \right) \left( \frac{2 + 3\beta^5}{\beta(3 + 2\beta^5)} \right) - 1 \]

[Boutin & Geindreau JASA, 124 (6), 2008]  \hspace{1cm} \beta = (1 - \phi_p)^{1/3}

\[ \phi_p = 0.3083 \pm 0.005 \]

\[ \phi_p = 0.2997 \pm 0.005 \]

\[ \sigma = 21.068 \pm 1.444 \text{ kPa.s.m}^{-2} \]

\[ \sigma = 24.592 \pm 1.510 \text{ kPa.s.m}^{-2} \]
- Overall porosity calculated from measured bulk density

- Nanoporosity measured from isotherms (Nitrogen @ 77K) using the Dubinin-Radushkevich method

- Microporosity calculated from the other porosity values

- Nanopore radius, micropore radius, and adsorption and desorption coefficients are adjusted through a best fitting routine

- Values of adsorption and desorption coefficients match the measured static bulk modulus
- Micropore and nanopore radii in the order of those commonly found in activated carbon

- Sorption characteristic time is proportional to the square of the particle radius when the particle is “large” [Do, Imperial College Press, 1998]

<table>
<thead>
<tr>
<th>Sample</th>
<th>$\phi_p$</th>
<th>$r_p [\text{mm}]$</th>
<th>$\phi_m$</th>
<th>$r_m [\text{um}]$</th>
<th>$\phi_n$</th>
<th>$r_n [\text{nm}]$</th>
<th>$k_a [1/\text{Pa/s}]$</th>
<th>$k_d \times 10^{-3} [1/\text{s}]$</th>
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</thead>
<tbody>
<tr>
<td>A</td>
<td>0.3083</td>
<td>0.7536</td>
<td>0.5571</td>
<td>0.2048</td>
<td>0.1602</td>
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<tr>
<td>B</td>
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<td>0.7064</td>
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<td>0.2593</td>
<td>1.2868</td>
<td>13.8479</td>
<td>8.3216</td>
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</tbody>
</table>
Future work

• Several physical processes are likely to accompany sound propagation in granular activated carbon:
  
  • **Diffusion** [Nori & Brandani, JASA 135(5), 2014] [Crank, Clarendon press, Oxford, 1975]
  • **Multi-layer sorption** [Do, Imperial College Press, 1998]
  • **Surface diffusion** [Do, Imperial College Press, 1998]
  • **Film and capillary condensate flow** [Do&Do, Chem. Eng. J. 84, 2001]
  • **Phase change (evaporation-condensation)** [Raspet et al., JASA 105(1), 1999]
  • **Competitive sorption of multiple species**
  • **Multi-phase flow**
  • **Poroelasticity** [Dazel&Tournat, JASA 127(2), 2010]

• Upscale the wave equation for multiscale materials including the above physical phenomena.
• Use sound as a probe.
• Too many open questions...
Conclusions

• A model for sound absorption of activated carbon has been introduced in a phenomenological manner.

• The model:
  • accounts for the multi-scale nature of the material, physical processes specific to small pores, and the interaction among several scales.
  • depends on several parameters that can be measured independently.
  • provides satisfactory predictions of sound absorption coefficient of activated carbon

• Some of the parameters have been adjusted. However, their values are representative to those normally found in activated carbon.

• Several research topics have been mentioned.
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MANY THANKS FOR YOUR ATTENTION. YOUR QUESTIONS ARE MORE THAN WELCOME!

“THERE’S PLENTY OF ROOM AT THE BOTTOM”, R. FEYNMAN, Caltech, December 29th, 1959