MODELLING OF FULLERENE PRODUCTION
BY
THE ELECTRIC ARC-DISCHARGE METHOD
Seminar

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   - Nanotubes

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Fullerenes

What are fullerenes?

Fullerenes are carbon forms that consist of a:
- spherical (buckyballs),
- ellipsoid, or
- cylindrical (buckytubes = nanotubes)

arrangement of carbon atoms.

Unique properties:
- chemical stability
- extreme strength
- can absorb light
- act as superconductors

Possible applications:
- medicine and electronics
- engineering and construction
- composites, paints, coatings
- aluminium, steel
Buckyballs

- Formed of pentagon and hexagon units of carbon
- Form hollow geodesic domes; bonding strains are equally distributed
- Most common: $C_{60}$

Figure: Buckyballs $C_{60}$, $C_{70}$ and $C_{80}$ [1].
Single-wall carbon nanotubes (SWNT).

- type: closed, open
- graphene sheet rolled into a tube, caped with half a buckyball
- type: armchair ($m = 1$), zigzag ($n = m$), chiral

Figure: Single-wall nanotubes [3].

Figure: SEM (left) and TEM (right) images of SWNT bundles [4].
Multi-wall carbon nanotubes (MWNT)

- multiple concentric or rolled graphene sheets
- type: Russian Doll model (concentric cylinders), Parchment model (a single sheet of graphene is rolled around itself)

Figure: Multi-wall nanotube [5].

Figure: SEM (left) and TEM (right) images of MWNT bundles [4].
Fullerene production

Production techniques

- **Arc-discharge:**
  - Arc vaporisation of 2 carbon rods placed a few mm apart.
  - SWNT, MWNT, buckyballs. Short tubes of random sizes in need of purification. The most efficient.

- **Laser ablation (vaporisation):**
  - Blast graphite with intense laser pulse.
  - Mostly MWNT, SWNT. Long tubes, MWNTs riddled with defects, diameter of SWNTs is controllable.

- **Chemical vapour deposition (CVD):**
  - A gas of carbon source is injected into an oven where heated substrate is present.
  - SWNT, Long bundles of tubes, good diameter control, few defects, costly.

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**Figure:** 1. Arc-discharge apparatus [9]. 2. Laser ablation apparatus [7]. 3. CVD apparatus [8].
Arc-discharge cell

Figure: A scheme of a typical arc discharge reactor chamber and the locations where products are formed (Grlj, 2010).
MODELLING OF FULLERENE PRODUCTION BY THE ELECTRIC ARC-DISCHARGE METHOD

Models of fullerene growth

Fan (plasma) jet is formed as a consequence of heat and mass transfer.

Review of fullerene modelling:

- Kerstinin, Moravsky (1998): mathematical model combined with kinetics for fullerene formation
- Farhat et al. (2005): mathematical model based on carbon deposition on rotating cathode.

Figure: Fullerene formation scheme in arc discharge (Grlj, 2010).
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Physical model

Overview of the general assumptions:

- Steady state.
- Axisymmetric, laminar flow with Re < 10.
- Local thermodynamic equilibrium.

Bilodeau (1998):

- Uniform anode erosion rate over the electrode surface.
- Surface deposition on the cathode is governed by diffusion.
- Energy input in the arc is due to ohmic heating and to the enthalpy flux of electrons.
- 1D electric field.

Farhat (2006):

- Radiation losses are accounted for by the net emission coefficient.
- Temperature dependent fluid properties.

Figure: The sketch of jet fan in arc-discharge reactor.
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Physical model
Governing equations including chemical reactions

Continuity equations

Continuity equation - general form:
\[ \frac{\partial \rho}{\partial t} + \vec{\nabla} \cdot (\rho \vec{u}) = S_m \]

Cylindrical coordinates:
\[ \frac{\partial \rho}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r}(\rho ru_r) + \frac{\partial}{\partial z}(\rho u_z) = S_m \]
\[ \frac{1}{r} \frac{\partial}{\partial r}(\rho ru_r) + \frac{\partial}{\partial z}(\rho u_z) = 0 \]

2D steady state model (Bilodeau, 1998)
\[ \vec{\nabla} \cdot (\rho \vec{u}) = S_m \]

1D steady state model (Farhat, 2006)
\[ \frac{\partial \rho}{\partial t} = -\frac{u_z}{\rho} \frac{\partial \rho}{\partial z} - 2V - \frac{\partial u_z}{\partial z} = 0 \]
MODELLING OF FULLERENE PRODUCTION BY THE ELECTRIC ARC-DISCHARGE METHOD

Physical model

Governing equations including chemical reactions

Momentum equations

Momentum equation - plasma - general form:

\[ \frac{D(\rho \vec{u})}{Dt} = -\vec{\nabla} P + \vec{\nabla}^2 (2\mu \vec{u}) + \vec{\nabla} \cdot (\mu \vec{\nabla} \times \vec{u}) + \rho \vec{g} + \vec{j} \times \vec{B} + \rho \vec{f} \]

Local thermodynamic equilibrium (L.T.E.)

Momentum equation - gas - general form:

\[ \frac{\partial(\rho \vec{u})}{\partial t} + \vec{\nabla} \cdot (\rho \vec{u} \vec{u}) = -\vec{\nabla} P + \mu \vec{\nabla}^2 \vec{u} + \rho \vec{g} + \vec{j} \times \vec{B} + \rho \vec{f} \]

Equation of state - general form:

\[ P = \frac{\rho RT}{M} \]
MOMENTUM EQUATIONS 2

2D model (Bilodeau, 1998)

\[ \nabla \cdot (\rho \vec{u} \vec{u}) = -\nabla P + \mu \nabla^2 \vec{u} + \rho \vec{g} + \vec{j} \times \vec{B} \]

Radial momentum:

\[
\rho \left( \frac{\partial u_r}{\partial t} + u_r \frac{\partial u_r}{\partial r} + u_z \frac{\partial u_r}{\partial z} \right) = -\frac{\partial P}{\partial r} + \mu \left( \frac{1}{r} \frac{\partial}{\partial r} \left( ru_r \right) + \frac{\partial^2 u_r}{\partial z^2} \right) + \rho g_r - j_z B_\theta + \rho f_r
\]

Axial momentum:

\[
\rho \left( \frac{\partial u_z}{\partial t} + u_r \frac{\partial u_z}{\partial r} + u_z \frac{\partial u_z}{\partial z} \right) = -\frac{\partial P}{\partial z} + \mu \left( \frac{1}{r} \frac{\partial}{\partial r} \left( ru_z \right) + \frac{\partial^2 u_z}{\partial z^2} \right) + \rho g_z + j_r B_\theta + \rho f_z
\]

1D model (Farhat, 2006)

\[
\rho \frac{\partial V}{\partial t} = -\frac{\partial}{\partial z} \left( \mu \frac{\partial V}{\partial z} \right) - \rho u_z \frac{\partial V}{\partial z} - \rho V^2 - \frac{1}{r} \frac{\partial P}{\partial r} = 0
\]
Conservation of species

Species conservation equation - general form:

\[ \frac{\partial (\rho Y_i)}{\partial t} + \nabla \cdot (\rho Y_i \vec{u}) = \nabla \cdot (\rho D_i \nabla Y_i) + S_{in} \]

2D steady state model (Bilodeau, 1998):

\[ \nabla \cdot (\rho \vec{u} Y_C) = \nabla \cdot (\rho D_C \nabla Y_C) + S_{in} \]

1D steady state model (Farhat, 2006):

\[ \rho \frac{\partial Y_i}{\partial t} + \frac{\partial (\rho Y_i V_i)}{\partial z} + \rho u \frac{\partial Y_i}{\partial z} = M_i \omega_i \]

Simplification:

\[ \rho \frac{\partial Y_i}{\partial t} = M_i \omega_i \]
Conservation of energy

The conservation of energy equation - general form:

\[
\frac{D(\rho h)}{Dt} = \nabla \cdot (k \nabla T) + \frac{j^2}{\sigma} + \frac{5}{2} \frac{k_B}{e} \nabla \cdot (Tj) - (k - \rho D_C c_p) \nabla (T_C - T_g) \cdot \nabla Y_C - Q_{rad} - S_h
\]

2D steady state model (Bilodeau, 1998):

\[
\nabla \cdot (\rho \nabla h) = \nabla \cdot (k \nabla T) + \frac{j_z^2}{\sigma} + \frac{5}{2} \frac{k_B}{e} c_p j \cdot \nabla T - (k - \rho D_C c_p) \nabla (T_C - T_g) \cdot \nabla Y_C - Q_{rad} + S_h
\]

1D steady state model (Farhat, 2006):

\[
\rho c_p \frac{\partial T}{\partial t} = \frac{\partial}{\partial z} \left( k \frac{\partial T}{\partial z} \right) - \rho c_p u_r \frac{\partial T}{\partial z} - \sum_{i=1}^{n_g} \left( c_{pi} \rho Y_i V_i \frac{\partial T}{\partial r} + \omega_i h_i \right) + S_h - Q_{rad} = 0
\]
Boundary and initial conditions

Figure: Boundary and initial conditions (Bilodeau, 1998).
Chemical kinetics model

Kinetic models for fullerene growth:

- intermediate cluster formation
- the pentagon road
- the fullerene road
- the ring road

**Figure:** Comparison of different fullerene growth models [6].
MODELLING OF FULLERENE PRODUCTION BY THE ELECTRIC ARC-DISCHARGE METHOD

Physical model
Boundary and initial conditions

Chemical kinetics model

The fullerene model:

\[ k_{1,2r} = A_{1,2r} T^{\beta_{1,2r}} \exp\left( -\frac{E_{1,2r}}{RT} \right) \]

\[ q_r = k_{1r} \prod_{i=1}^{n_g} C_i^{\nu_{ir}} - k_{2r} \prod_{i=1}^{n_g} C_i^{\nu'_{ir}} \]

\[ \omega_i = \sum_{r=1}^{R} \nu_{ir} q_r = \frac{dC_i}{dt} \]

Chemistry of small clusters
- \( C + C \leftrightarrow C_2 \)
- \( C + C_2 \leftrightarrow C_3 \)
- \( C_2 + C_2 \leftrightarrow C_3 + C \)

Formation of carbon clusters CC
- \( C_3 + C \leftrightarrow 0.100 CC \)
- \( C_3 + C_2 \leftrightarrow 0.125 CC \)
- \( C_3 + C_3 \leftrightarrow 0.150 CC \)

Growth of carbon clusters CC
- \( CC + C \leftrightarrow 1.025 CC \)
- \( CC + C_2 \rightarrow 1.050 CC \)
- \( CC \rightarrow 0.95 CC + C \)

Formation of fullerene molecules \( C_{60}^F \) and \( C_{70}^F \)
- \( CC + C_3 \rightarrow 0.70 C_{60}^F \)
- \( CC + C_2 \rightarrow 0.70 C_{60}^F \)
- \( CC + C \rightarrow 0.6833333 C_{60}^F \)

Decay of fullerene molecules \( C_{60}^F \) and \( C_{70}^F \)
- \( C_{60}^F \rightarrow 1.45 CC + C_2 \)
- \( C_{70}^F \rightarrow 1.70 CC + C_2 \)

Formation of soot nuclei Z and growth of soot
- \( CC + CC \rightarrow Z \)
- \( Z + C_3 \rightarrow 1.0375 Z \)
- \( Z + C_2 \rightarrow 1.025 Z \)
- \( Z + C \rightarrow 1.0125 Z \)
The initial model - domain scheme

Figure: 1D domain scheme.
The initial model is based on 1D Farhat model (Farhat, 2006).

- **Continuity eq.**:
  \[
  \frac{\partial \rho}{\partial t} = - \frac{\partial}{\partial z}(\rho u_z) - 2V - \frac{\partial u_z}{\partial z} = 0
  \]

- **Momentum eq.**:
  \[
  \rho \frac{\partial u_r}{\partial t} = - \frac{\partial}{\partial z} \left( \mu \frac{\partial u_r}{\partial z} \right) - \rho u_z \frac{\partial u_r}{\partial z} - \frac{\partial p}{\partial r} - j_z B_\theta
  \]

- **Energy eq.**:
  \[
  \rho c_p \frac{\partial T}{\partial t} = \frac{\partial}{\partial z} \left( k \frac{\partial T}{\partial z} \right) - \rho c_p u_r \frac{\partial T}{\partial z} - \sum_{i=1}^{n_g} \left( c_{pi} \rho Y_i V_i \frac{\partial T}{\partial r} + \omega_i h_i \right) + \frac{j_z^2}{\sigma} = 0
  \]

- **Species conservation eq.**:
  \[
  \rho \frac{\partial Y_i}{\partial t} = M_i \omega_i
  \]
The initial model - boundary and initial conditions

Figure: Boundary and initial conditions for 1D model (Farhat, 2006).
The improved model - domain scheme

Figure: 2D domain scheme.
Improved model

Initial model is going to be expanded to more dimensions (2D).

Improved model is based on Bilodeau model (Bilodeau, 1998).

- **Continuity eq.:**
  \[
  \frac{\partial (\rho)}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r} (\rho u_r) + \frac{\partial}{\partial z} (\rho u_z) = S_m
  \]

- **Momentum eq.:**
  \[
  \rho \left( \frac{\partial u_r}{\partial t} + u_r \frac{\partial u_r}{\partial r} + u_z \frac{\partial u_r}{\partial z} \right) = - \frac{\partial p}{\partial r} + \mu \left( \frac{\partial}{\partial r} \left( \frac{1}{r} \frac{\partial}{\partial r} (ru_r) \right) + \frac{\partial^2 u_r}{\partial z^2} \right) + j_z B_\theta
  \]
  \[
  \rho \left( \frac{\partial u_z}{\partial t} + u_r \frac{\partial u_z}{\partial r} + u_z \frac{\partial u_z}{\partial z} \right) = - \frac{\partial p}{\partial z} + \mu \left( \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial u_z}{\partial r} \right) + \frac{\partial^2 u_z}{\partial z^2} \right) + \rho g_z + j_r B_\theta
  \]

- **Energy eq.:**
  \[
  \frac{\partial (\rho h)}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r} (r \rho u_r h) + \frac{\partial}{\partial z} (\rho u_z h) = \frac{1}{r} \frac{\partial}{\partial r} \left( rk \frac{\partial T}{\partial r} \right) + \frac{\partial}{\partial z} \left( k \frac{\partial T}{\partial z} \right) + \frac{j_z^2}{\sigma} + \frac{5}{2} \frac{k_b}{e} \left( j_r \frac{\partial T}{\partial r} + j_z \frac{\partial T}{\partial z} \right) - \\
  - \frac{1}{r} \frac{\partial}{\partial r} \left( rk - r \rho D_{C} c_p \right) (T_C - T_g) \frac{\partial Y_C}{\partial r} - \frac{\partial}{\partial z} \left( k - \rho D_{C} c_p \right) (T_C - T_g) \frac{\partial Y_C}{\partial z}
  \]

- **Species conservation eq.:**
  \[
  \rho \frac{\partial Y_C}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r} (r \rho u_r Y_C) + \frac{\partial}{\partial z} (\rho u_z Y_C) = \frac{1}{r} \frac{\partial}{\partial r} \left( r \rho u_r \frac{\partial Y_C}{\partial r} \right) + \frac{\partial}{\partial z} \left( \rho u_z \frac{\partial Y_C}{\partial z} \right) + S_{in}
  \]
MODELLING OF FULLERENE PRODUCTION BY THE ELECTRIC ARC-DISCHARGE METHOD

The model

Planned improvements

The improved model - boundary and initial conditions

Figure: 2D boundary and initial conditions (Bilodeau, 1998).
## Values of reduced input parameters

### Input parameters:

<table>
<thead>
<tr>
<th>constant</th>
<th>label</th>
<th>value</th>
</tr>
</thead>
<tbody>
<tr>
<td>anode diameter</td>
<td>$2r_A$</td>
<td>6 - 7</td>
</tr>
<tr>
<td>cathode diameter</td>
<td>$2r_C$</td>
<td>6 - 16 mm</td>
</tr>
<tr>
<td>anode length</td>
<td>$l_A$</td>
<td>15-70 cm</td>
</tr>
<tr>
<td>cathode length</td>
<td>$l_C$</td>
<td>15 -30 cm</td>
</tr>
<tr>
<td>reactor length</td>
<td>$l_C + l_A + l_g$</td>
<td>39-100 cm</td>
</tr>
<tr>
<td>reactor diameter</td>
<td>$2r$</td>
<td>13.6-30 cm</td>
</tr>
<tr>
<td>current</td>
<td>$I$</td>
<td>60-100 A</td>
</tr>
<tr>
<td>pressure</td>
<td>$P$</td>
<td>100-800 mbar</td>
</tr>
<tr>
<td>anode cathode distance</td>
<td>$l_g$</td>
<td>1-12 mm</td>
</tr>
<tr>
<td>anode, cathode tip $T$</td>
<td>$T_A$</td>
<td>3300-3800 K</td>
</tr>
<tr>
<td>wall temperature</td>
<td>$T_w$</td>
<td>350 K</td>
</tr>
<tr>
<td>mass density of the gas</td>
<td>$\rho$</td>
<td>$9.24 \cdot 10^{-6} \frac{g}{cm^3}$</td>
</tr>
<tr>
<td>carbon mass fraction</td>
<td>$n_C$</td>
<td>$10^{-4} - 10^{-6}$</td>
</tr>
<tr>
<td>initial C mole fraction</td>
<td>$N_C$</td>
<td>$2.57 \cdot 10^{-4}$</td>
</tr>
<tr>
<td>initial $C_2$ mole fraction</td>
<td>$N_{C_2}$</td>
<td>0.583</td>
</tr>
<tr>
<td>current intensity</td>
<td>$j$</td>
<td>$3 \cdot 10^6 - 10^7 \frac{A}{m^2}$</td>
</tr>
</tbody>
</table>
## Values of reduced output parameters

Output parameters:

<table>
<thead>
<tr>
<th>constant</th>
<th>label</th>
<th>value</th>
</tr>
</thead>
<tbody>
<tr>
<td>anode gas velocity</td>
<td>$u_A$</td>
<td>$7818 \text{ cm} / \text{s}$</td>
</tr>
<tr>
<td>deposition rate</td>
<td></td>
<td>$0.57 - 4.71 \text{ mg} / \text{s}$</td>
</tr>
<tr>
<td>electric power dissipation</td>
<td>$q$</td>
<td>$1.24 \cdot 10^7 \text{ W} / \text{m}^2$</td>
</tr>
<tr>
<td>dilution factor at the anode</td>
<td>$\tau$</td>
<td>20</td>
</tr>
<tr>
<td>erosion rate</td>
<td>$\Phi$</td>
<td>$1.3 - 25 \cdot 10^{-3} \text{ g} / \text{s}$</td>
</tr>
<tr>
<td>estimated electron density</td>
<td>$N_e$</td>
<td>$3.5 \cdot 10^{15} \text{ 1/cm}^3$</td>
</tr>
<tr>
<td>temperature</td>
<td>$T$</td>
<td>350-17000 K</td>
</tr>
<tr>
<td>He number density</td>
<td>$n_{He}$</td>
<td>$1.4 \cdot 10^{18} \text{ 1/cm}^3$</td>
</tr>
<tr>
<td>Ni number density</td>
<td>$n_{Ni}$</td>
<td>$2.0 \cdot 10^{14} \text{ 1/cm}^3$</td>
</tr>
<tr>
<td>Y number density</td>
<td>$n_Y$</td>
<td>$3.2 \cdot 10^{14} \text{ 1/cm}^3$</td>
</tr>
<tr>
<td>growth rate</td>
<td>$G$</td>
<td>$1 - 1000 \text{ 1/\mu m/min}$</td>
</tr>
</tbody>
</table>
Qualitative estimation of involved variables

Figure: Estimation of temperature field (Bilodeau, 1998).
MODELLING OF FULLERENE PRODUCTION BY THE ELECTRIC ARC-DISCHARGE METHOD

The model

Estimated parameter fields

Qualitative estimation of involved variables

Figure: Estimation of carbon mass fraction field (Bilodeau, 1998).
MODELLING OF FULLERENE PRODUCTION BY THE ELECTRIC ARC-DISCHARGE METHOD

The model
Estimated parameter fields

Qualitative estimation of involved variables

Figure: Estimation of axial current intensity field (Bilodeau, 1998).
Estimated number of equations calculated for a particular point for each time step (without boundary conditions):

- The initial model: 59 (Chemical reactions: $18 \cdot 3$, 5 governing equations)
- The improved model: 61 (Chemical reactions: $18 \cdot 3$, 7 governing equations)
Meshless method and radial basis functions

Meshless method = mesh reduction technique

A numerical simulation algorithm that uses a set of arbitrary nodes to represent the solution of a physical problem.

Radial basis functions (RBF)

General approximation functions of univariate polynomial splines to a multivariate domain.

$$
\psi_i(r) = \psi(\vec{p} - \vec{p}_i)
$$

Commonly used RBFs:
- Gaussian (GA) $$\psi(r) = e^{-(cr)^2}$$
- Multiquadric (MQ) $$\psi(r) = \sqrt{r^2 + c^2}$$

General form of an approximation function:

$$
\Theta(\vec{p}) = \sum_{i=1}^{N} \alpha_i \psi_i(\vec{p})
$$
Figure: Irregular domain discretized using (a) 3-noded triangular finite elements, b) boundary element, and (c) arbitrary interior and boundary points using a meshless method [10].
Local radial basis function collocation method

Approximation function:

\[ \Theta(\bar{p}) \approx \sum_{i=1}^{N} \alpha_i \psi_i(\bar{p}) \]

Collocation condition:

\[ \Theta(\bar{p}_i) = \theta_i \]

Linear system of N equations:

\[ \Psi \tilde{\alpha} = \tilde{\theta} \]

PDE equations:

\[ \frac{\partial^i}{\partial \bar{p}_i^j} \Theta(\bar{p}) = \sum_{n=1}^{N} \alpha_n \frac{\partial^j}{\partial \bar{p}_n^j} \Psi_n(\bar{p}) \]
Introduction of boundary conditions

- **Dirichlet**
  \[ \Theta(\vec{p}) = \Theta_{BC} \quad \rightarrow \quad \Theta(\vec{p}) = \Theta_{BC} \]

- **Neuman**
  \[ \frac{\partial}{\partial \vec{n}} \Theta(\vec{p}) = \Theta_{BC} \quad \rightarrow \quad \Theta_{BC} = \sum_{i=1}^{N} \alpha_i \frac{\partial}{\partial \vec{n}} \Psi_i(\vec{p}) \]

- **Robin**
  \[ \frac{\partial}{\partial \vec{n}} \Theta(\vec{p}) + b \Theta(\vec{p}) = \Theta_{BC} \quad \rightarrow \quad \Theta_{BC} = \sum_{i=1}^{N} \alpha_n \left( a \frac{\partial}{\partial \vec{n}} \Psi_i(\vec{p}) + b \Psi_i(\vec{p}) \right) \]
Conclusions

- Introduction to fullerene production and modelling.
- Presentation of arc - discharge method.
- Formulation of a mathematical model.
- Description of a numerical method.

Future steps:
- Establishment of a minimal model (implement model, solution procedure)
- Comparison with measurements from actual cell
More than 150 (154) articles were gathered on the subject of fullerene production and modelling. A comprehensive review of literature was done and is available at: https://4pm.cobik.si/projects/tabs/projectPortalTabDefFiles.jsf?list=1&prjld=1118
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