Welding of incompatible thermoplastic polymers
1. Motivation
2. Introduction
3. Experimental
4. Results
5. Conclusion
Need for research in the field of plastics joining

- more and more conventional materials are substituted by polymer materials
- material bonded connections are approaching their limits

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<td>x</td>
<td>x</td>
<td>!</td>
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</table>

- ✓ - compatible  × - incompatible  ! - partly compatible

- joining incompatible thermoplastic polymers by using their blends
Motivation

Example from real life

Fuel tank (PE-HD)

Tank filler neck (PA12)

Aim

Fuel tank components (e.g. filler neck)

Polymer blend

Fuel tank (PE-HD)

source: RF Plast, Norma Rasmussen
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Polymer blends

- two basic types: compatible (miscible) and incompatible (immiscible)
  - most are incompatible

- different mixing ratio → different properties

- to improve properties of incompatible polymer blend → coupling agent
  - commonly used: maleic anhydride groups

Demixing of PE/PA12-Blends
Coupling agent – maleic anhydride

PA chain

Coupling agent

PE chain

Maleic anhydride – grafted on PE

Incompatible
Reaction between amine and maleic anhydride

PE chain

MAH

\[\text{H}_2\text{C} = \text{C} = \text{C} = \text{O}\] + \[\text{NH}_2\]

PA

Ring opening

\[\text{H} = \text{O} = \text{H}\] - \[\text{H}_2\text{O}\]

Ring formation

\[\text{H} = \text{C} = \text{C} = \text{N} - \text{H}_2\text{C} = \text{C} = \text{O}\]
## Reaction between amine and maleic anhydride

<table>
<thead>
<tr>
<th>Group 1</th>
<th>Group 2</th>
<th>Conversion at 2 min at 180°C</th>
<th>Reaction rate (kg/mol.min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maleic Anhydride</td>
<td>NH\textsubscript{2} end-group of PA</td>
<td>99%</td>
<td>(~10^3)</td>
</tr>
<tr>
<td>Carboxylic acid</td>
<td>NH\textsubscript{2} end-group of PA</td>
<td>0%</td>
<td>-</td>
</tr>
<tr>
<td>GMA epoxy</td>
<td>NH\textsubscript{2} end-group of PA</td>
<td>1.8%</td>
<td>0.34</td>
</tr>
<tr>
<td>GMA epoxy</td>
<td>Carboxylic end-group of PA</td>
<td>9%</td>
<td>2.1</td>
</tr>
</tbody>
</table>

Source:
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Blending process

Materials

- Polyethylene: Lupolen GX 5038, LyondellBasell
- Polyamide 12: Grilamid L20, EMS-GRIVORY
- Coupling agent: Scona TSPE 1112 GALL, BYK

<table>
<thead>
<tr>
<th>Blend</th>
<th>Polyamide [% w/w]</th>
<th>Polyethylene (contains 25% coupling agent) [% w/w]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Blend 1 (70 % PA)</td>
<td>70</td>
<td>30</td>
</tr>
<tr>
<td>Blend 2 (50 % PA)</td>
<td>50</td>
<td>50</td>
</tr>
<tr>
<td>Blend 3 (30 % PA)</td>
<td>30</td>
<td>70</td>
</tr>
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</table>
Hot plate butt-welding (welding process with melt flow)

1) Contact

2) Heating

3) Changeover

4) Joining

1. stretching of the molecular chains cause by the melt flow

2. relaxation of the molecular chains and the diffusion through the joining plane

Molecular chains

Joining plane
Diffusion welding (welding process without melt flow)

a) First contact of the melt

b) Beginning of diffusion processes

c) Dissolve of the interface

d) Ending of diffusion processes
Welding experiments

Parameter variation

<table>
<thead>
<tr>
<th>Welding Parameters</th>
<th>Hot plate butt-welding</th>
<th>Diffusion welding</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature of the heating tool [°C]</td>
<td>220 – 300</td>
<td>200- 250</td>
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<tr>
<td>Heating time</td>
<td>30 – 40 seconds</td>
<td>60 - 120 minutes</td>
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<tr>
<td>Welding pressure [MPa]</td>
<td>0.1 – 2.0</td>
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</tbody>
</table>

Optimized parameters

<table>
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<tr>
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</thead>
<tbody>
<tr>
<td>PE</td>
<td>Blend</td>
<td>220 - 230</td>
<td>0.1</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>PA</td>
<td>Blend</td>
<td>270</td>
<td>0.1</td>
<td>200</td>
<td>60</td>
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</tbody>
</table>
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Mechanical properties of the polymer blends

Blends

<table>
<thead>
<tr>
<th>Blend 1 (70% PA)</th>
<th>Blend 2 (50% PA)</th>
<th>Blend 3 (30% PA)</th>
<th>Grilamid L 20 (PA12)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Yield stress</td>
<td>35</td>
<td>25</td>
<td>20</td>
</tr>
<tr>
<td>Yield strain</td>
<td>10</td>
<td>8</td>
<td>6</td>
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</table>

Base materials

<table>
<thead>
<tr>
<th>Lupolen 5038 (PE)</th>
<th>Scona TSPE 1112 (PE_g_MA)</th>
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</thead>
<tbody>
<tr>
<td>Yield stress</td>
<td>12</td>
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<tr>
<td>Yield strain</td>
<td>10</td>
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</tbody>
</table>

In the graph, the x-axis represents different blends and base materials, and the y-axis represents yield stress and yield strain. The bars indicate the median values, with error bars representing the spread. The legend at the bottom left of the graph shows the interpretation of the colors: grey for yield stress and red for yield strain.
The welding factor is calculated as the quotient of the weld strength and the yield stress of the “weakest” material.

Polyamide $\sigma = 41$ MPa

Blend 1 (70% PA)
Blend 2 (50% PA)
Blend 3 (30% PA)

Polyethylene $\sigma = 18$ MPa

Tensile strength [MPa]

<table>
<thead>
<tr>
<th>Blend 1 (70% PA)</th>
<th>Blend 2 (50% PA)</th>
<th>Blend 3 (30% PA)</th>
<th>Blend 1 (70% PA)</th>
<th>Blend 2 (50% PA)</th>
<th>Blend 3 (30% PA)</th>
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<tbody>
<tr>
<td>0,65</td>
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<td>0,54</td>
<td>0,55</td>
<td>0,93</td>
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</table>

The welding factor is calculated as the quotient of the weld strength and the yield stress of the “weakest” material.
Optical microscopy imaging of the weld seam

- in vicinity of the PA-Blend interface → several small micro-cracks
- solely on the part of the blend
- mainly in blends with high ratio of PA
Comparison of diffusion and hot plate welding

Polyamide
\[ \sigma = 41 \, \text{MPa} \]

Blend 1 (70% PA)
Blend 2 (50% PA)
Blend 3 (30% PA)

<table>
<thead>
<tr>
<th>Blend</th>
<th>Hot plate butt-welding</th>
<th>Diffusion welding</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.58</td>
<td>0.43</td>
<td>0.54</td>
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<tr>
<td>0.99</td>
<td>0.95</td>
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Tensile strength [MPa]

Polyamide
\[ \sigma = 41 \, \text{MPa} \]
Optical microscopy imaging of the weld seam

No micro-cracks were found
Conclusion

- suitable choice of polymer blends → welding of incompatible thermoplastic polymers possible
- welding strength at the level of polyethylene
- **hot plate welding**: the potential strength of joints of PA and blend cannot be completely utilized
- **diffusion welding**: increase of the welding strength

Outlook

- full investigation of the bonding mechanism
- clarification of the influence of the coupling agent
- investigation of the micro-cracks
- researches of multi-component injection molding
  - injecting polymer blend to base material
  - analysis of the bonding mechanism

**Aim**: polymer blend as interlayer between incompatible thermoplastic polymers
Thank you for your attention!

TU Chemnitz – Professorship of Plastics Engineering
Prof. Dr.-Ing. Michael Gehde

**Contact information:**
Mirko Albrecht

E-mail: mirko.albrecht@mb.tu-chemnitz.de
Tel.: +49 (0) 371-531 34826
Homepage: www.kunststofftechnik-chemnitz.de