Title: Innovative, sustainable materials for roofing – biobased roofing membranes

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Curriculum: As an environmental economist, Wouter Jan van den Berg has over 10 years of experience in bio-based research and business development. At Icopal (Benelux), Wouter Jan is responsible for the application for subsidies and other types of co-funding for the several innovative and sustainable projects. He has been so from 2004. He also functions as the main contact with the co-funding organisation, and has been project manager for several projects. One of the most challenging projects has been the construction and demonstration of a recycling facility for old roofing felts, co-funded by a European LIFE environment subsidy as well as the European Eco Innovation program. Since 2011, Wouter Jan is also responsible for the management of the so-called 'CO2 performance ladder', a Dutch initiative to stimulate companies to reduce CO2 emissions in a structured approach, and since 2016 for the set-up of an ISO 14001 environmental management system. As from 2015, Wouter Jan is in charge of the project ‘DISCOVER’ with the ambition to develop a 100% bio-based roofing membrane.

Abstract: Conventional roofing materials are high quality products with a long life, but are made mainly from fossil raw materials and its derivatives; like bituminous roofing based on the oil derivative bitumen, or synthetic membranes that are based on polymers stemming from the petroleum chemistry. The uncertainty of petroleum exploitation, the by consequence high fluctuations in price, as well as the CO2 footprint motivate the project partners to address this issue by looking for alternative renewable raw materials, while maintaining the required technical and functional characteristics. Therefore, together with Wageningen UR Food & bio-based Research, Icopal has started the development of a new generation of roof waterproofing material made exclusively from renewable raw materials. Icopal is the leading supplier of products for the protection of buildings and structures in Europe. Icopal provides roofing membranes for flat roofs. In 2016, Icopal, founded in 1876, was taken over by Standard Industries and became a sistercompany of GAF, leader in North America in roofing materials. Recently Icopal merged with Braas Monier, manufacturer of tiles for sloped roofs, into BMI. As a leader in the area of recycling of roofing material and supplier of the most sustainable roofing membrane (Icopal Universal), Icopal also wants to be the number one roofing company with respect to the use of natural resources. The ambition is the development of a 100% bio-based roofing membrane that is not only scientifically sound, but can also be fitted into the existing production-process of Icopal and application techniques of roofing, without any compromise on product quality. In short, a true "drop-in" alternative to existing roofing materials. Such a roofing has yet not been developed. The project name
is 'DISCOVER', meaning ‘Development of Innovative Sustainable COVEring materials for Roofs’. From the inventory of biobased resources several raw materials have been selected as promising in view of the desired criteria for a roofing membrane formulation. Suppliers of these raw materials have been contacted and test samples were ordered. These raw materials have been tested by Icopal and FBR in biobased compounds for their effect on compatibility, stability, viscosity profile and other relevant product properties. Many combinations of raw materials and process conditions have been tested. Some biobased raw materials showed low, others a high compatibility with each other. Substitution of the polymer part in the roofing membrane formulation is challenging and several biobased polymers are studied. Additionally, the novel formulations will be ranked according to desired properties but also compared to known (literature and patents) ones.
BIO-BASED Roofing Membranes

‘Innovative, sustainable materials for roofing’
BPM symposium June 14th 2018
Wouter Jan van den Berg, BMI
Agenda

1 Historical background

2 Current roofing materials

3 Project headlines (Reason why, ambition, partners)

4 Project progress

5 Lessons learnt
1 Historical background

The name was inspired by 'copal', the Aztec word for resin

1876 Icopal (DK)
1927 Esha/ Smid & Hollander (Groningen)
2005 Icopal, owned by InvestCorp
2016 Icopal + GAF (Standard Industries)
2018 BMI (merger with Braas Monier)

Together, we are the largest roofing and water-proofing business in the world. http://www.bmigroup.com/
1 Historical background

Biobased Icopal roofings in the past

- Carrier ((<1970):
  - Wool felt → is now glass fleece / PET fleece
  - Jute/ sackcloth → is now glass fabric

- No biobased bitumen
2 Current flat roofings

- high quality products
- with a long life-time,
- but mainly from fossil raw materials like
  - bituminous roofing based on oil derivative bitumen, or
  - synthetic membranes based on polymers from the petroleum chemistry.

Bitumen as the reference:
there are 4 groups of components: asphaltenes to saturates, with decreasing Mw and polarity. Asphaltenes and saturates are too far apart to be miscible at the molecular level: colloidal structure.

![Diagram of bitumen model](https://example.com/bitumen-model.png)
2 What on earth is bitumen?

- Bitumen is the non-volatile part of crude-oil, a black, sticky, very viscous liquid.

- It consists of
  - Elements: Carbon (~85%), hydrogen (~10%) as well as sulfur, oxygen, nitrogen, and trace metals.
  - Structures: large hydrocarbon molecules, from maltenes (relatively small, apolar) to asphaltaltenes (large, more polar)
2 Sustainability of bitumen

- Samples of ancient use show hardly any deterioration. Below soil application can stay unchanged for many centuries. Durability means sustainability.
- Unlike tar, bitumen has very low amounts of hazardous components PAH (typically <20 ppm total PAH).
- Bitumen has no leaching of any components into water since it is absolutely watertight (extensive research done for Dutch leaching guideline, use in potable water storage).
- Bitumen is easy recyclable; just re-melt and able to rejuvenate (asphalt). BiELSo, Pentack.
2 Current modifications for roofs

Modification with polymers:

- **SBS** thermoplastic rubber, gives high elasticity and wide temperature range.
- **APP** amorphous polyolefine, gives UV resistance and wide temperature range.
  - **POCB** as a high range polyolefine-modified ("FPO plasticised with bitumen").
2 current use of bitumen

<table>
<thead>
<tr>
<th>Estimates</th>
<th>Existing</th>
<th>Annual increase &amp; replacement</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flat</td>
<td>400</td>
<td>24</td>
</tr>
<tr>
<td>Sloped (tiles)</td>
<td>250</td>
<td>9</td>
</tr>
<tr>
<td>Total</td>
<td>650</td>
<td>33</td>
</tr>
</tbody>
</table>

~100 mln. tons of bitumen produced each year worldwide, of which
• About 85% is used as a binder in asphalt for road construction.
• 10% is used in roofing applications and
• 5% is used for sealing and insulating purposes
3 Reason why + ambition

a) The uncertainty of petroleum exploitation,
b) the by consequence high fluctuations in price,
c) as well as the CO$_2$ footprint

motivate us to look for alternative renewable raw materials.

The target is to develop 100% biobased ‘drop in’ roofing that can be produced without changing the production process and/ or existing methods for roof application.
3 Project partners

- Icopal
  Icopal is a leading manufacturer of a wide range of roofing materials. Icopal provides roofing membranes for flat roofs. Recently Icopal merged with Braas Monier, manufacturer of tiles for sloped roofs, into BMI.

- WFBR (Wageningen Food & Biobased Research)
  WFBR is a contract research organisation who has extensive know-how on biobased raw materials, green conversion processes and develops biobased applications together with industrial partners.
4 Project progress: WP’s’s

WP1: project management
WP2: inventory
WP3: lab scale testing
WP4: pilot scale testing
WP5: industrial run
WP6: demonstration
WP7: environmental screening
WP8: dissemination
WP 2 Inventory of resources

+ a lot of interesting biobased materials, but..
- no suitable biobased polymer yet
+ shortlist of materials with a positive effect on these necessary conditions:
  - Processing @ different temperatures (viscosity)
  - Flow resistance (R&B) and/ or
  - Walkability (Penetration)
WP 2 interesting materials

Traditional & new biobased resources

- Forestry & paper industry: e.g. lignin, tall oil pitch, copal, cellulose esters
- Agriculture: f.i. rapeseed oil, linseed standoil, factices

➔ approx. 50 materials/subtypes in total
## WP 2 selecting raw materials

<table>
<thead>
<tr>
<th>Method</th>
<th>Results in ..</th>
<th>(Functional) Informative value</th>
</tr>
</thead>
<tbody>
<tr>
<td>-DSC + melting bench</td>
<td>Values for glass transition temperatures (Tg) and melting temperatures (Tm)</td>
<td>Tg is indication of low temperature flexibility and Tm is for flow resistance. (if for instance, a material has a low Tm, you need a material with a higher Tm to mix it with in order to have a compound that can be used on a roof)</td>
</tr>
<tr>
<td>-FTIR</td>
<td>method to characterize functional groups, bonding types, nature of compounds based on infrared absorption spectra</td>
<td>chemical composition/ chemical stability (like indication of hydrophobicity of a material or on whether a (chemical) modification has been successful (stable))</td>
</tr>
<tr>
<td>-GPC</td>
<td>molecular weight distribution, compatibility (Gibbs theory, the enthalpy component)</td>
<td>is very relevant for the compatibility, gives indication of viscosity (heavier molecules tend to have higher viscosity)</td>
</tr>
</tbody>
</table>
## WP 2 selecting compounds

<table>
<thead>
<tr>
<th>Method</th>
<th>Results in ..</th>
<th>(Functional) Informative value</th>
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<tbody>
<tr>
<td>- R&amp;B</td>
<td>temperature at which a ball 'penetrates' a material in a ring</td>
<td>flow resistance</td>
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<tr>
<td>- viscosity</td>
<td>flow behaviour of a liquid substance @ different temperatures</td>
<td>processing @ different temperatures</td>
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<tr>
<td>- penetration</td>
<td>hardness indication</td>
<td>walkability</td>
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<tr>
<td>- ARES</td>
<td>Temperature at maximum Loss modulus and at maximum phase angle</td>
<td>how a material reacts on external forces, elastic &amp; plastic behaviour</td>
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<tr>
<td>- DSC</td>
<td>Values for glass transition temperatures (Tg) and melting temperatures (Tm)</td>
<td>for blends: Indication of compatibility at molecular level, temperature window for use phase</td>
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</tbody>
</table>
WP 2 (further) selection

DoE: ‘the design of any task that aims to describe or explain the variation of information under conditions that are hypothesized to reflect the variation’ (Wikipedia)

Important calculation indicators

- G-efficiency (→ 100%)
- Condition no (→ 1)

Based on these indicators we made several designs

![Table]

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<th>No.</th>
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WP 2 DSC + melting bench

Different absolute results, same trending

Plant oil

Peak = -18.78 °C
Area = 69.484 J/g
Delta H = 49.4936 J/g

Peak = -15.47 °C
Area = 53.615 J/g
Delta H = 53.6150 J/g
WP 2 TGA example

```
TGA 30-900 °C under air

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<td>800</td>
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<tr>
<td>900</td>
<td>10</td>
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</table>

Legend:
- RM1
- RM2
- RM3
- RM4
- RM5
- RM6
- RM7
- Bitumen
```
WP 2 GPC example

![Biobased raw materials versus bitumen graph](image)
WP 3 Lab scale testing

+ We made a lot of compounds (approx. 100!)
+ Testprotocols
+ Options for improvement (less hydrophobic)? modification
 +/- A lot of (useful info from) new competing patents
 +/- DoE preparation/ DoE implementation (conditions have to be the same for all compounds even if not required by the compound itself)
- we didn’t make a successful blend yet
WP 3 Lab scale testing

The most suitable bio-based raw materials were tested in formulations by Icopal and WFBR on its basic characteristics (e.g. miscibility, compatibility, rheological (processing at different temperatures (viscosity), flow resistance (R&B) and/or walkability (hardness) and thermal behaviour), intermolecular level (e.g. dispersion, visco-elastic behaviour, reactivity).
Flexible, N1
- $E''$ -7°C
- Tan (δ) 54°C

Rigid, N8
- $E''$ 83°C
- Tan (δ) 120°C

Tan (δ) based on storage / loss modulus
Tbd/ No insights yet

WP4 Pilot scale testing

WP5 Industrial run

WP6 Demonstration

WP8 Dissemination
WP7 Environmental screening

Shadowprice (environmental impact measured in money) of biobased often worse than bitumen

- Bitumen € 0,03
- Lignin € 0,09
- Linseed/ rape-oil € 0,64

How to deal with that?
5 Lessons learnt

- Bitumen vs biomass: still 1-0 (hydrophobic vs hydrophylic/ more oxygen)
- No wonder: Oil sector is there for 1,5 century. Distillation of bitumen even more than 800 years. So, no surprise we need more time (than 3 years)
- Even so, we already learned a lot (like methods to analyze, DoE, non-fit raw materials/ compounds, alkyl/ester ratio (should be in favor of alkyl), importance of frequent face to face contact between project team-members)