LOW TEMPERATURE RHEOLOGICAL AND FRACTURE PROPERTIES OF POLYMER-MODIFIED BITUMENS

L.Lapalu[1], J.P. Planche[1], D. Martin[1], D. Anderson[2], J.F. Gérard[3]

[1] ELF Research Center (Solaize)

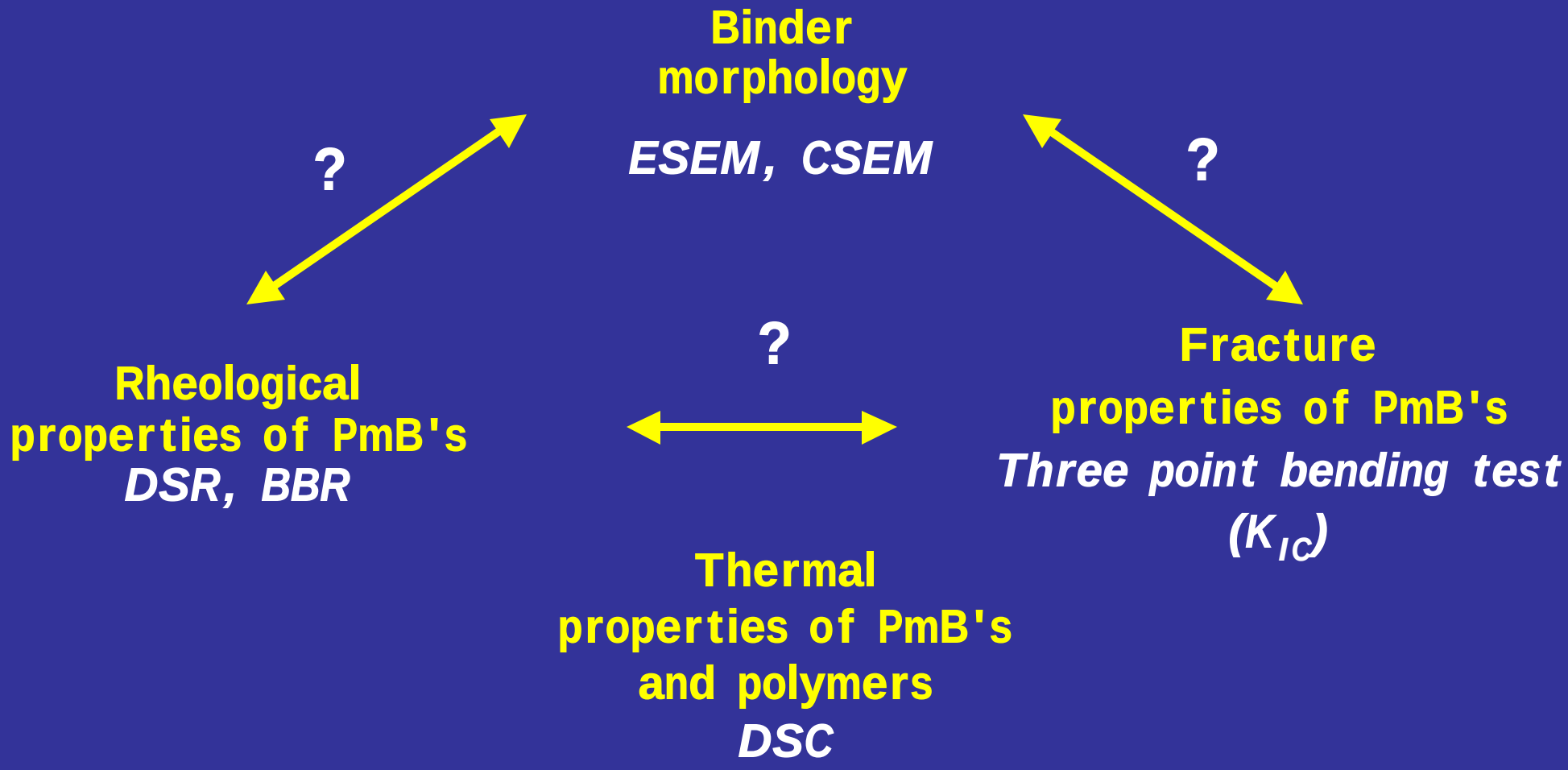
[2] The Pennsylvania State University

[3] National Institute of Applied Science (Lyon)

Eurasphalt & Eurobitume Congress

Barcelona, September 2000

THE AIM OF THE STUDY



Analyze of the effects of the polymer chemical nature, polymer

MATERIALS

Neat bitumen

Grade :	70/100
Penetration at 25°C (1/10 mm) :	85
Ring & Ball softening point (°C) :	46
Tg (°C) :	-27
% Crystallized fractions (DSC) :	4.5

MATERIALS

Semi-crystalline Copolymers : Plastomers

Copolymer	Fraction of the comonomer (% wt.)	T_g (°C)	Crystallinity %
EVA-18	18.6	-22	25.7
EVA-28	28.4	-20	15.3
EMA-28	28.6	-26	9.4
EBA-35	33.9	-46	10.6

PmB's produced by mixing the base bitumen with 4 to 6 weight % semi-crystalline copolymers

All physical blends

MATERIALS

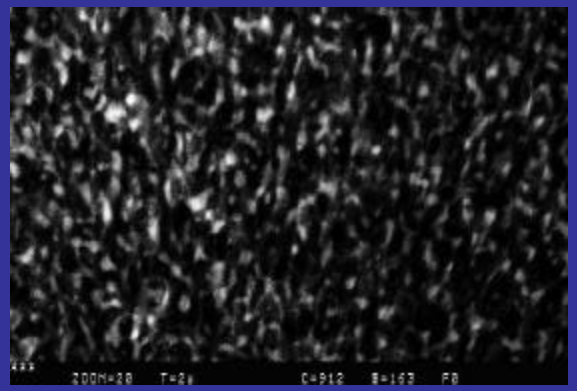
Amorphous copolymers : Elastomers

Copolymer	Styrene content (% wt.)	T_g (°C) (PB / PS blocks)	M_w (dalton)
SBS_1	30	-90 / 51	240 000
SBS_2	30	-90 / 62	135 000
SBS_3	18	-100 / 66	250 000
SB	18	-88 / 66	250 000

PmB's produced by mixing the base bitumen with 4 weight % amorphous copolymers

Four physical blend - One chemically in-situ crosslinked blend

OBSERVATION OF THE BLEND MORPHOLOGY USING CLSM

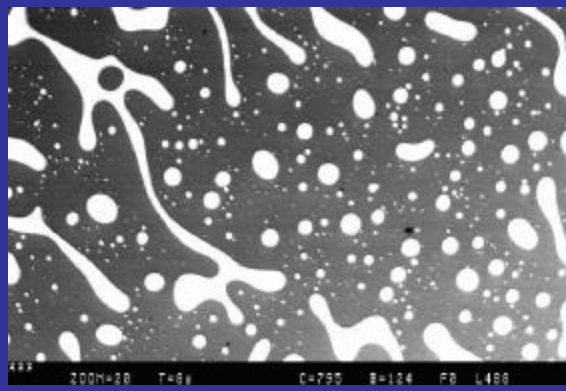


Blend based on 4% wt. EBA-35

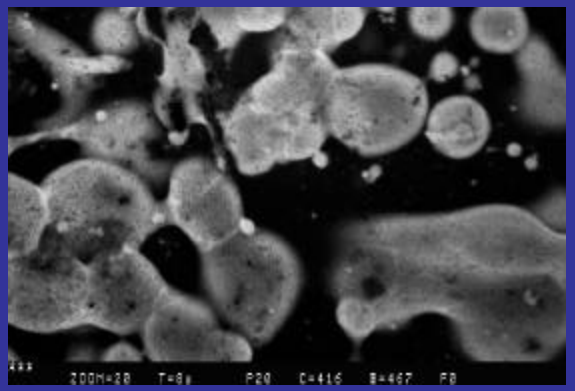
co-continuous morphology

Blend based on 4% wt. EMA-28

polymer-rich particles dispersed in the bitumen-rich matrix



50 µm



Blend based on 4% wt. SBS*[3]

polymer-rich particles dispersed in the bitumen-rich matrix

MEASUREMENT OF RHEOLOGICAL PROPERTIES

DSR :

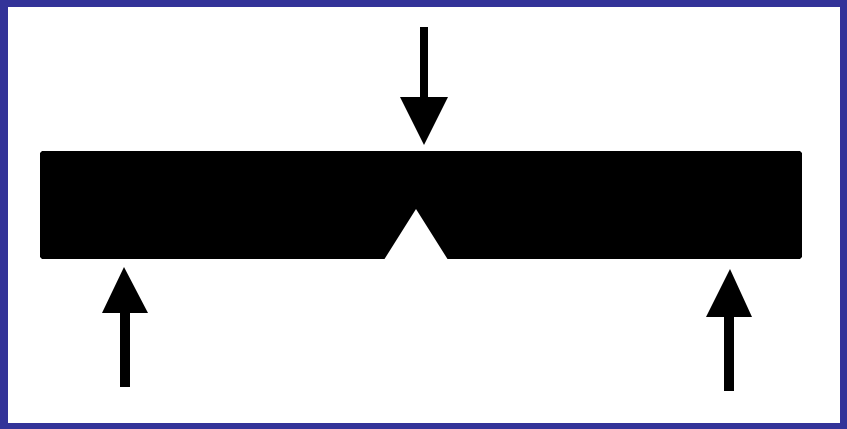
Rheometrics RDAII
Measurement at 1Hz from -120°C to 100°C
G', G'' = f(T)

BBR :

BBR Cannon Instrument Company
Tests performed at -18°C, -21°C and -24°C
Measurement of $T_{S=300MPa}$ and $T_{m=0.3}$

Tests run on unaged binders

THE MODE-I FRACTURE TEST

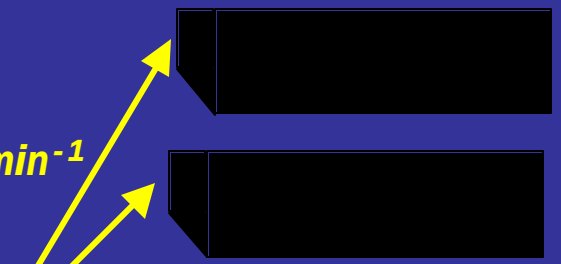


Environmental chamber at -20°C

Test

Speed = 0.6 mm.min^{-1}

T = -20°C

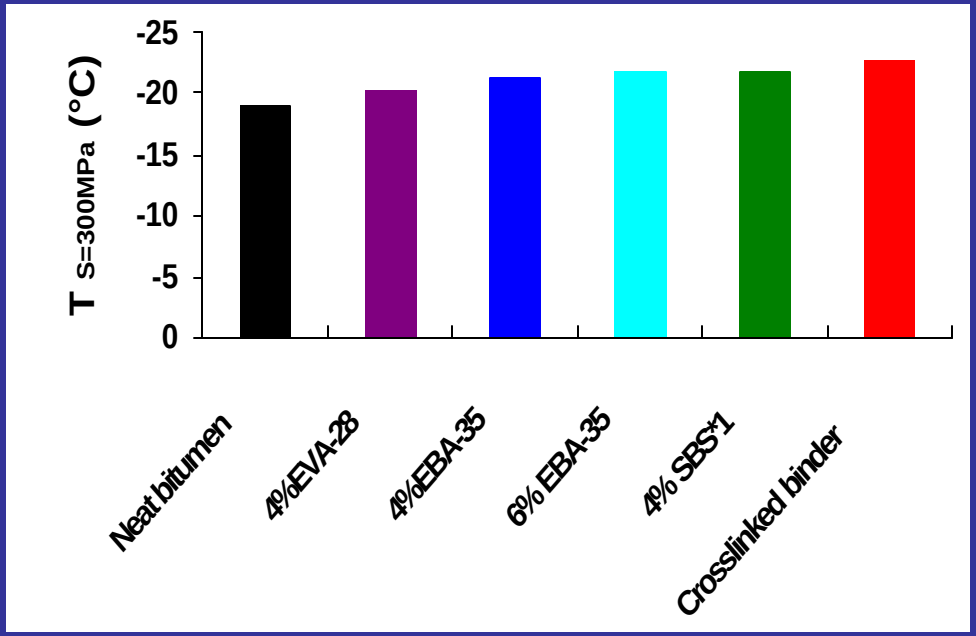


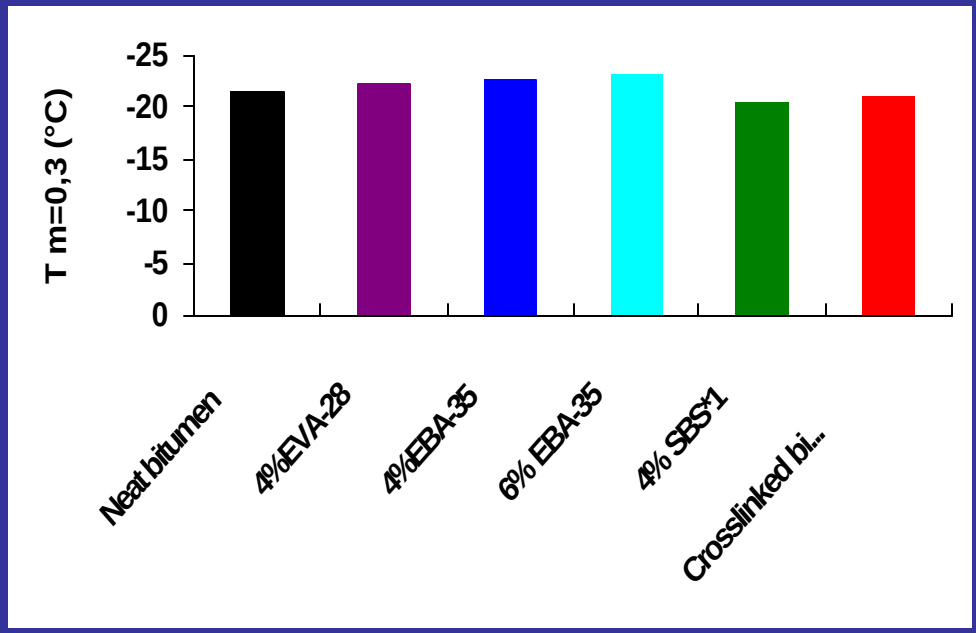
Cutting of the rupture faces for CSEM or ESEM observations

K_{IC} = failure load ´ f (span, crack length, sample dimensions)

8 replicates necessary for each sample

RHEOLOGICAL PROPERTIES : BBR RESULTS





BBR results : Polymer addition imparts low improvement to binder creep properties

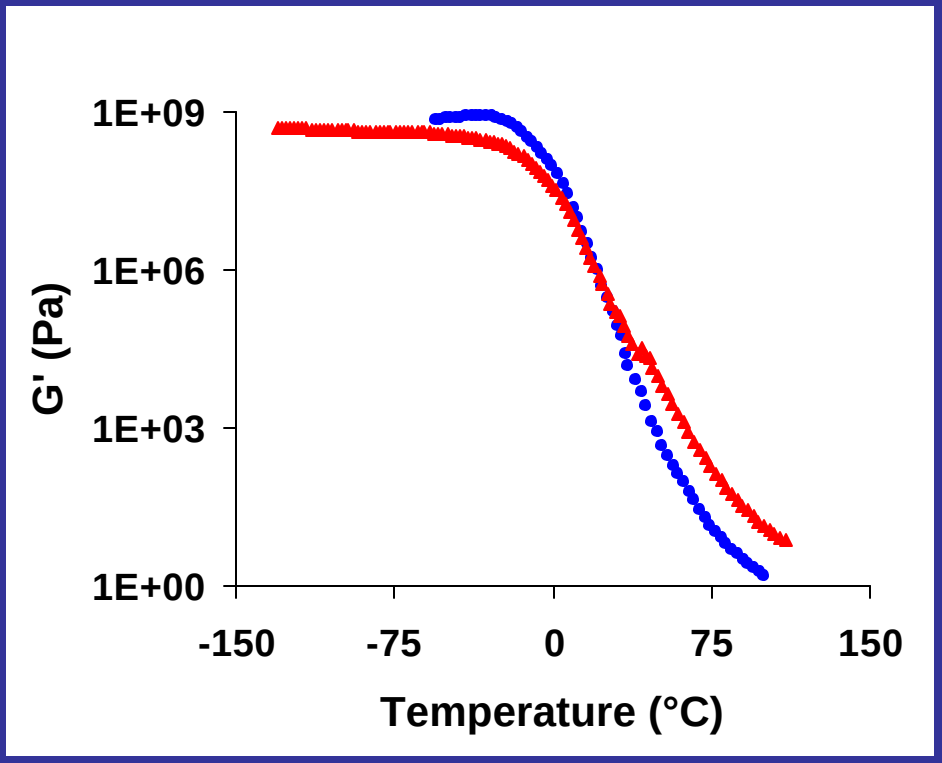
BBR does not discriminate binders with respect to their rheological properties at low temperature

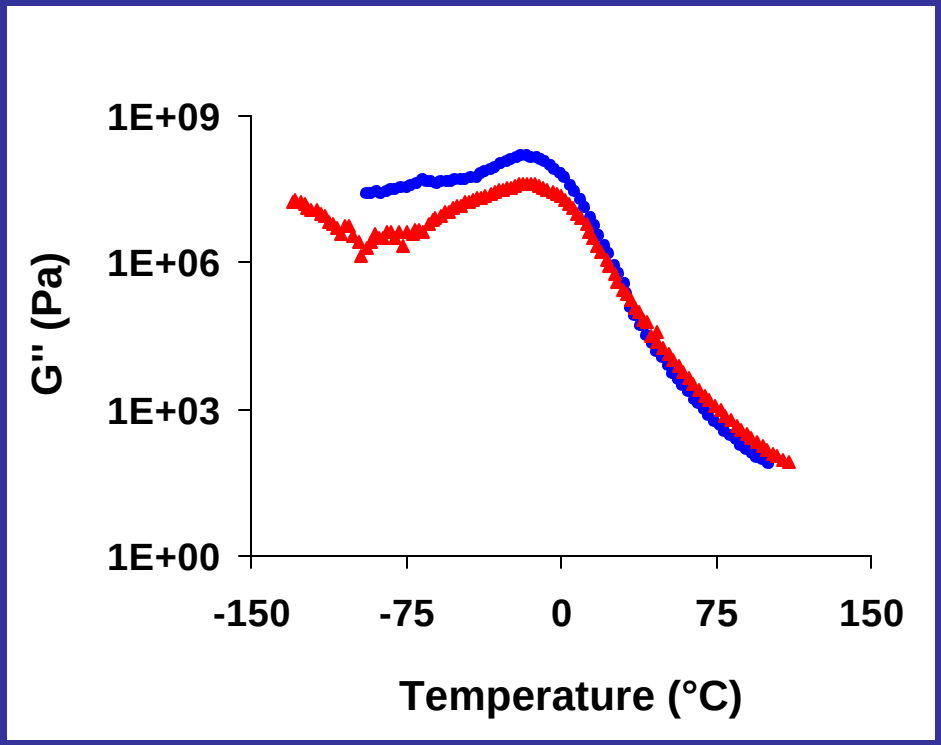
RHEOLOGICAL PROPERTIES : DSR RESULTS

Improving the low temperature rheological properties by polymer modification should result in decreasing the elastic modulus G' and/or increasing the viscous modulus G''

4% SBS*$_3$ Blend

Neat bitumen

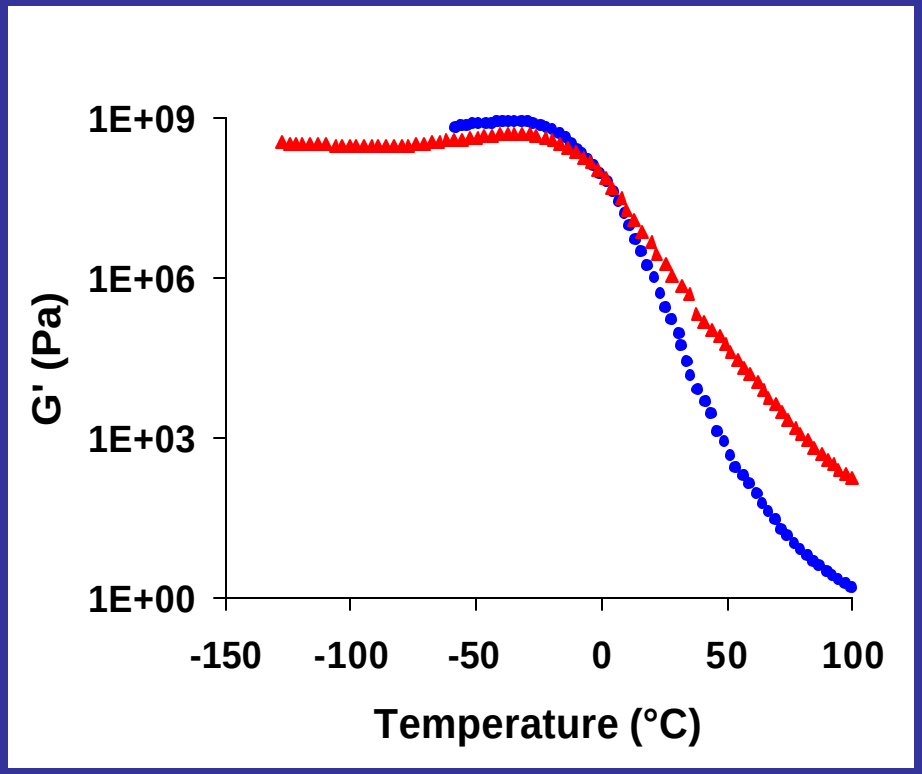


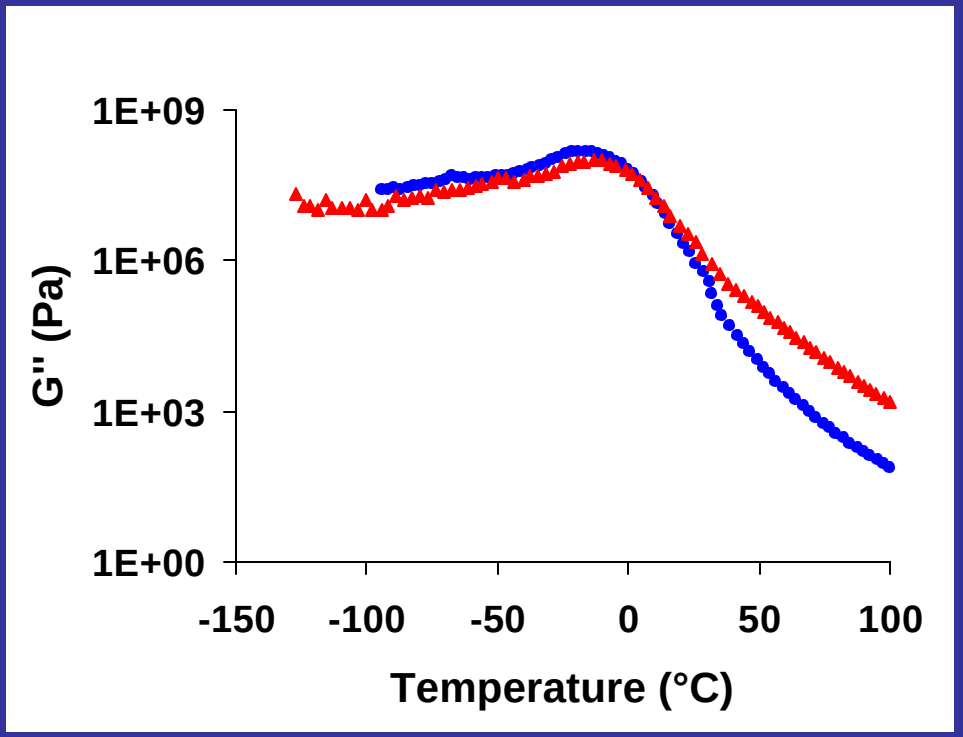


RHEOLOGICAL PROPERTIES : DSR RESULTS

6% EVA-18 Blend

Neat bitumen





In both cases, polymer addition does not significantly improve the rheological properties of the binder at low temperature regardless of the polymer nature

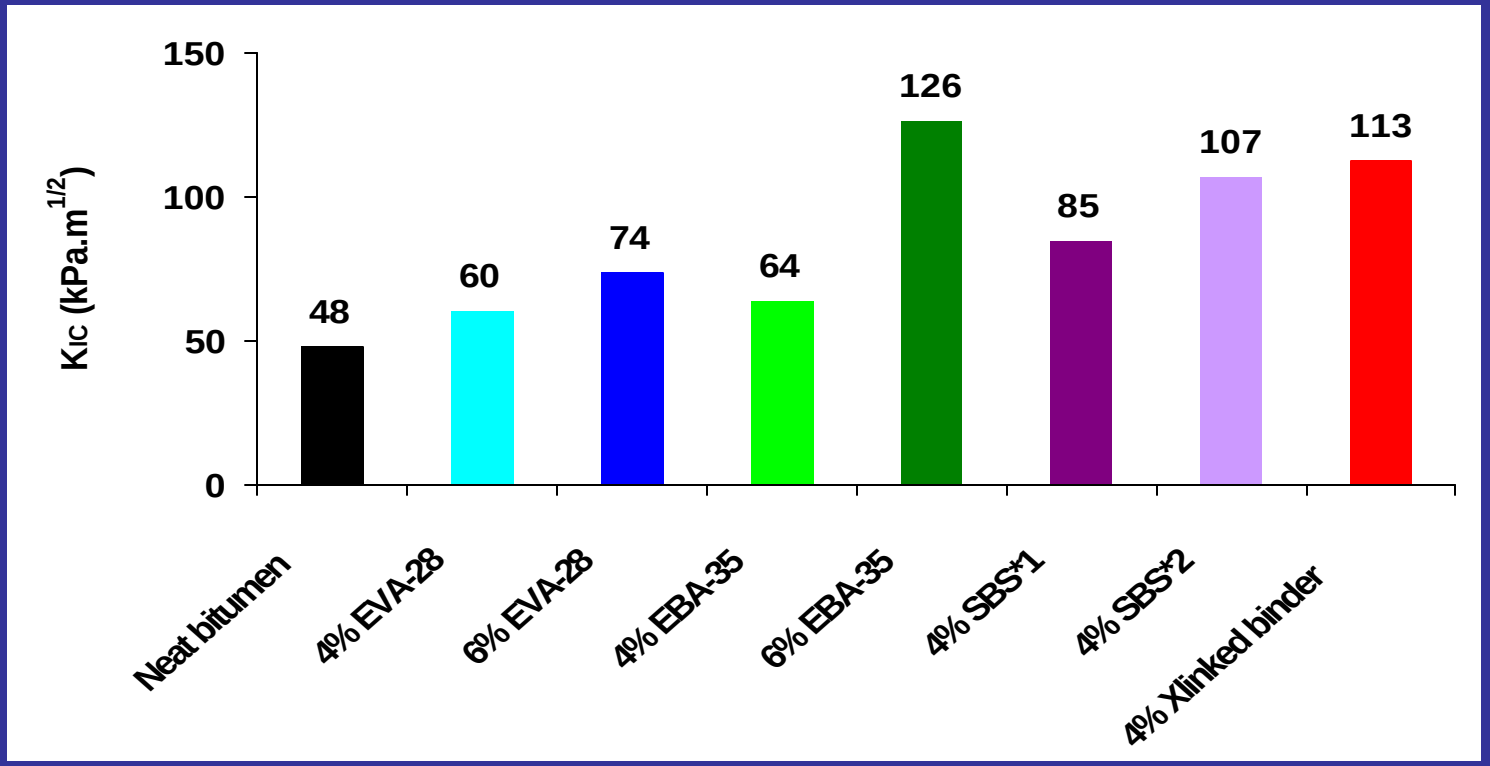
RHEOLOGICAL PROPERTIES : DSR AND BBR CONCLUSIONS

- **Low temperature rheological properties of polymer modified bitumens seem to be controlled by the properties of the higher modulus phase :**
  - ⇨ ***the bitumen matrix***

- ***These conclusions could be different at higher polymer concentrations***

- ***These results are not necessary valid for aged binders***

FRACTURE RESULTS AT -20°C



- Plastomer blends K_{IC} is slightly higher than the base bitumen
- 4% SBS* and SB mB's display higher K_{IC} than 4 % ethylene copolymer mB's
- 6% EBA needed to match 4% SBS* or SB mB's K_{IC}
- The 4% crosslinked binder displays a high K_{IC}
- ***Fracture properties significantly differentiate various polymer modification systems***

FRACTURE MECHANISMS

4 parameters have to be considered to explain the fracture mechanism

- Binder morphology
- Volume fraction of the dispersed phase
- Adhesion between the phases
- Temperature difference between the testing temperature (-20°C) and the T_g's of each binder component

FRACTURE MECHANISM FOR A NEAT BITUMEN

$K_{IC} = 48\ kPa.m^{1/2}$

No topographic contrast

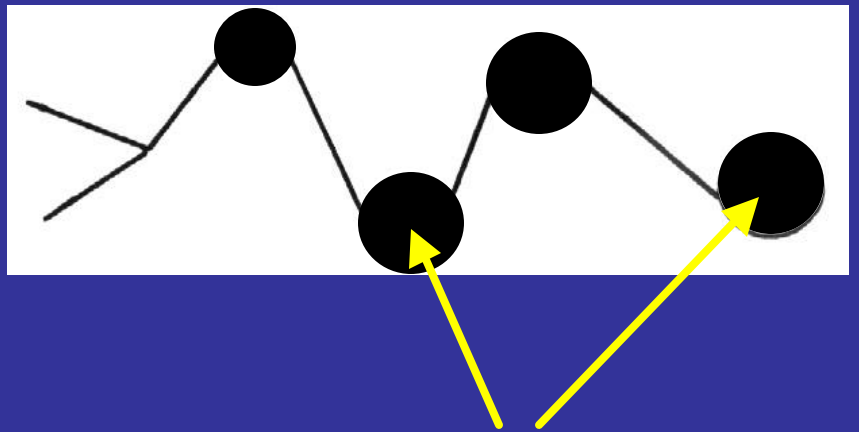
Brittle rupture ⇨ low K_{IC}

MAIN FRACTURE MECHANISMS FOR PmB's

Crack deflection

The crack deviates from one particle to the next

Particles are pulled-out

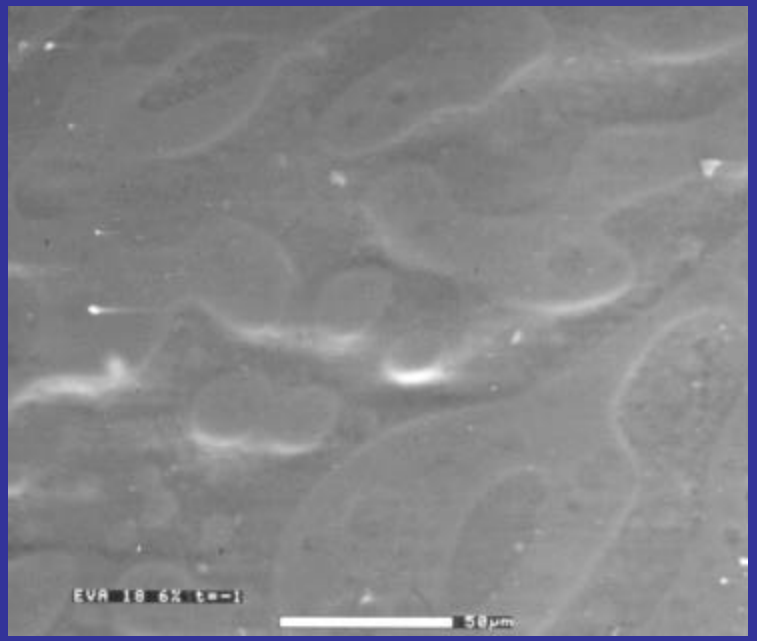


Polymer-rich nodules

CRACK DEFLECTION MECHANISM

6% EVA-28 blend

(K_{IC} = 74 kPa.$m^{1/2}$)



EVA and EMA nodules are glassy at the testing temperature

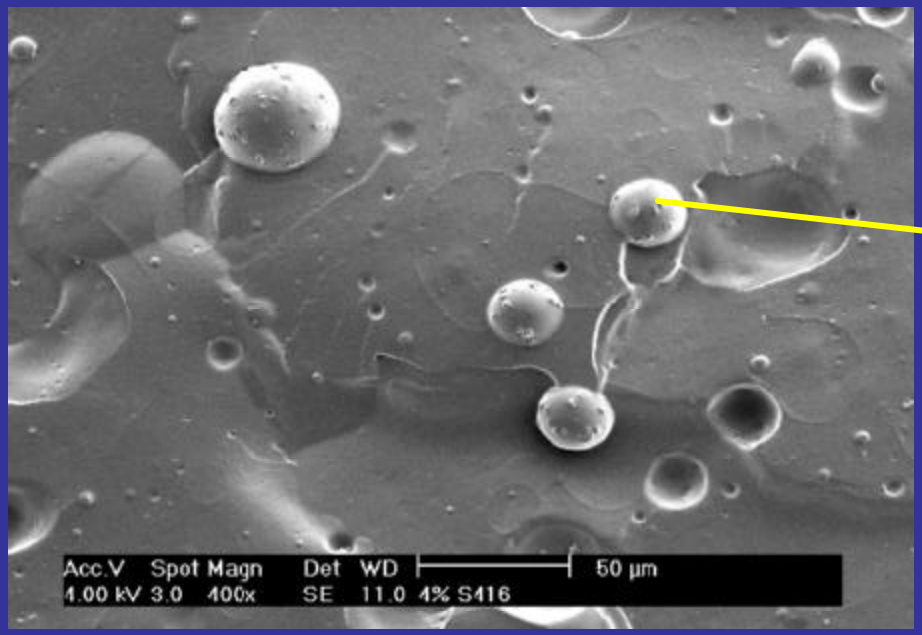
Polymer-rich particles are pulled-out without deformation

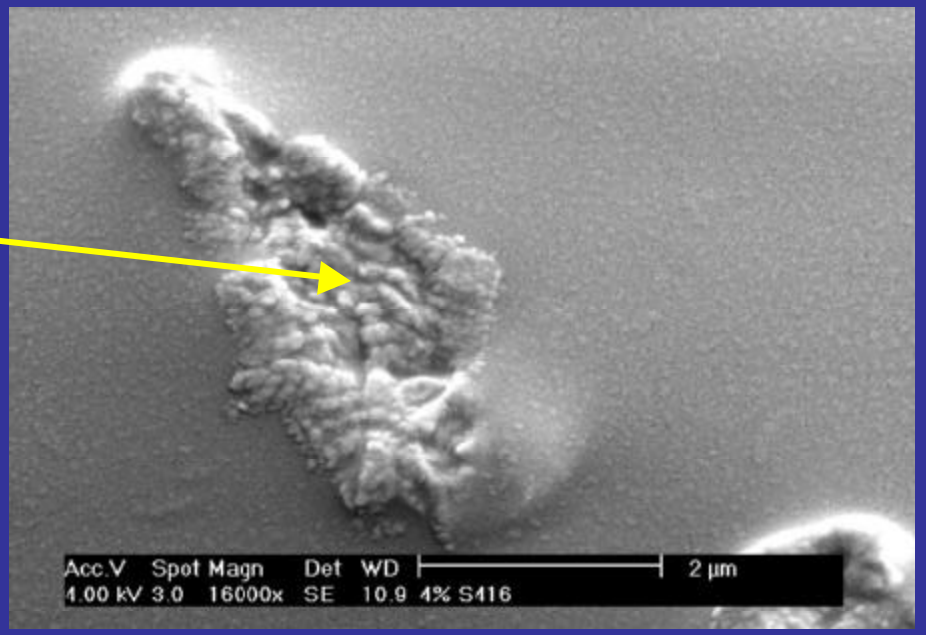
The fracture mechanism is governed by the (poor) adhesion between phases

⇨ **low K_{IC}**

CRACK DEFLECTION MECHANISM

Blend based on 4% wt. SBS*$_2$ (K_{IC} = 107 kPa.m$^{1/2}$)





Fracture faces show particle pull-out (crack deflection)

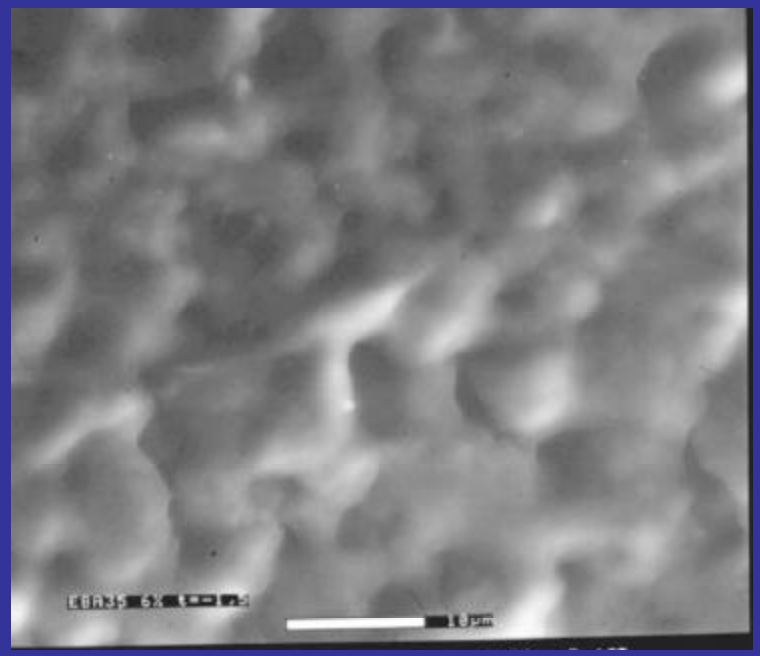
Plastic deformation of polymer-rich nodules before being pulled-out

⇨ ***High K_{IC}***

CRACK DEFLECTION MECHANISM WITH PLASTIC DEFORMATION

6% EBA-35 blend

K_{IC} = 126 kPa.m$^{1/2}$



EBA is in the rubbery state at -20°C

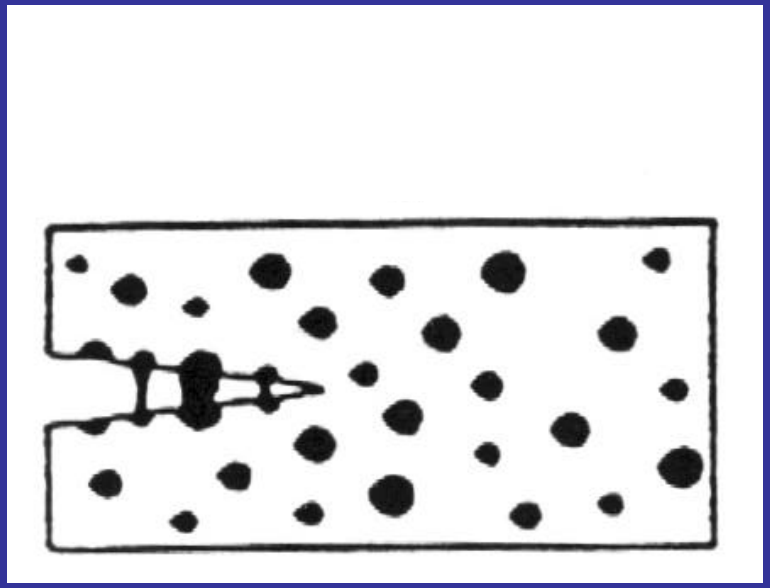
At 6%, the blend displays a finer co-continuous structure in favour of high toughness

⇨ ***high K_{IC}***

CRACK FRONT BRIDGING MECHANISM

The crack propagates through the polymer-rich nodules which are stretched

The interfacial adhesion between the phases has to be high

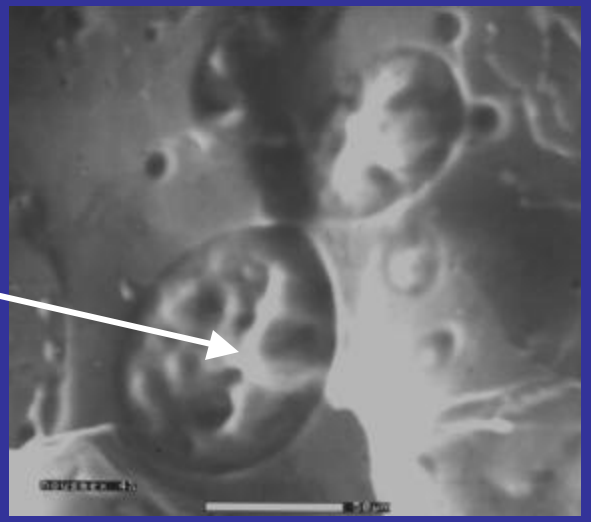


CRACK FRONT BRIDGING MECHANISM

4% SBS*$_1$ blend

K_{IC} = 85 kPa.m$^{1/2}$

Stretching of the polymer-rich nodules



The polymer-rich phase looks to be stretched (crack-bridging)

Good adhesion between phases

⇨ ***high K_{IC}***

CRACK FRONT BRIDGING MECHANISM

4% Crosslinked binder

K_{IC} = 113 kPa.m$^{1/2}$

Polymer-rich nodules are small

The dispersion of nodules is homogeneous

Fine interpenetration between the bitumen-rich matrix and polymer-rich domains

⇨ ***High K_{IC}***

FRACTURE PROPERTIES : CONCLUSIONS

- Polymer addition increases the bitumen fracture toughness at low temperature
- At low polymer content, the improvement is higher with elastomers than with plastomers due to :
  - different adhesion between phases
  - glass transition temperature
  - morphology of the blend
- These results need to be validated for mixes.

SUMMARY

- Rheological properties at low temperature are mainly controlled by the bitumen matrix properties

- Contrary to experience, rheological tests do not discriminate between binders

SUMMARY (Cont'd)

- Polymer addition increases the bitumen fracture toughness at low temperature
- At low polymer content, the improvement is higher with elastomers than with plastomers
- The fracture test allows to discriminate the low temperature thermal properties of asphalt binders
- Fracture mechanics is a good research tool, not a specification tool yet

CONCLUSIONS (CONT'D)

- **More work is needed :**
  - Effect of testing condition parameters need to be studied such as temperature with respect to T_g,...
  - These results need to be validated for asphalt mixes

- **Do not forget that**
  - These conclusions are valid only for unaged binders
  - These conclusions could be different for aged binders
  - These conclusions could be different for highly modified binders

THANK YOU !