Numerical and experimental study of the thermophysical properties of composite materials

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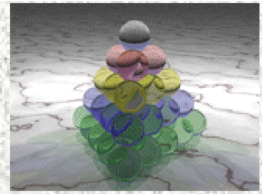


SUMMARY

1. **Presentation of CERTES laboratory**

2. **Composite materials : polymer matrix / conducting particles**
   - **PP/Cu**
   - **PP/ Al**
   - **PVC / Carbon Nanotubes**

3. **Composite materials : polymer matrix / insulating/ coated particles**
   - **EVA / glass particles**
   - **EVA / silver coated glass particles**
   - **HDPE / silver coated PA particles**

4. **Composite materials : polymer matrix/ brass spheres**
   - **Numerical study (calculation of the effective conductivity)**
   - **Thermal conductivity measurements**
   - **Validation (analytical models and Exp. Measurements)**

5. **Conclusion**

1. Presentation of the CERTES Laboratory

RESEARCH FIELDS: Energetic, measurements, modelization, materials.

- **Characterization of the thermal properties of composite materials**

  - *macroscopic scale*
  - *versus of Temperature*

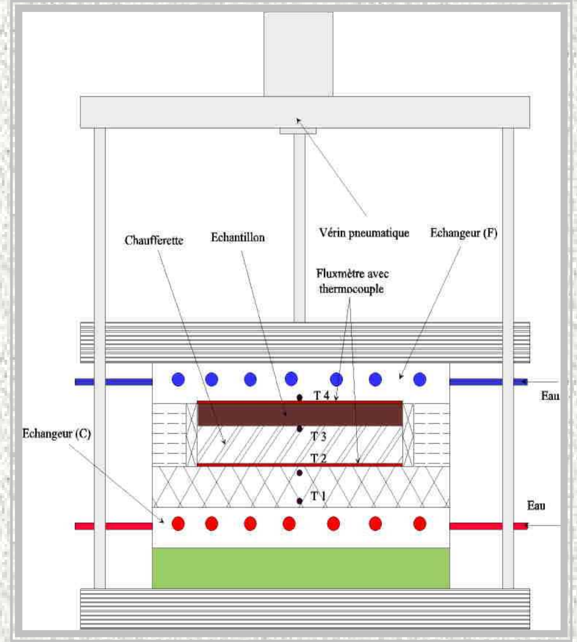
- **Development of a complementary methods for the characterization**

  - **Electrical and dielectric conductivity measurements**
  - **Mechanical measurements (Collaborations)**
  - **Structural Studies (SEM)**

Y. Candau (PR1)
M. Fois (MCF)
A. Boudenne (MCF)
L. Ibos (MCF)
M. Karkri (MCF)
A. Mazioud (MCF)
V. Feuillet (MCF)
R. Tlili (PhD student)
S. Sary Bey (PhD student)
A. Trigui (PhD student))

Composite materials characterization

PCG
Thermal conductivity (PCM)



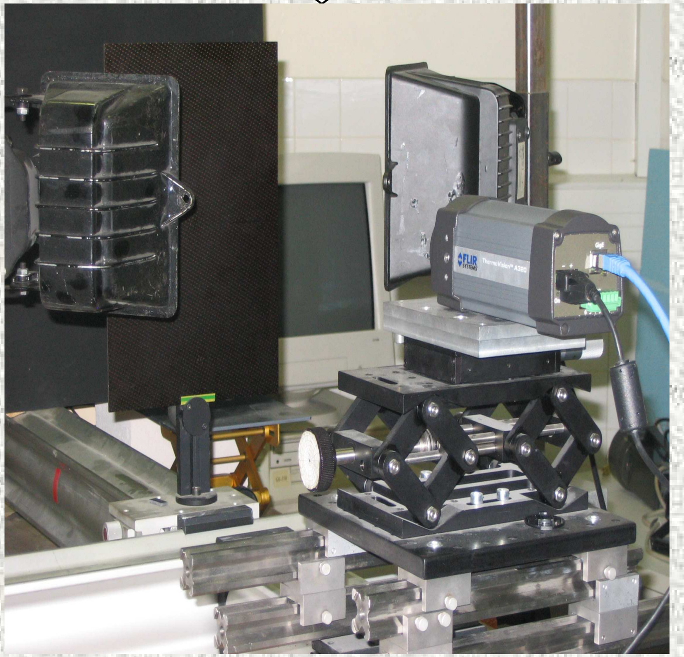
DICO :
Thermal conductivity and diffusivity

D.S.C
Specific heat capacity

Composite materials characterization

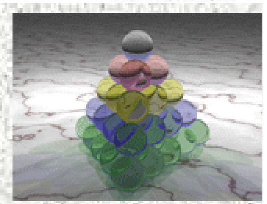
Thermal emissivity

T. IR
Flash Method



Electrical conductivity

SUMMARY



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Collab. Lab Rhéologie des Matières Plastiques, StEtienne

Electrical percolation point $$\log(\sigma_c / \sigma_m) = B(1 - e^{-\alpha\phi})^n$$

The inflexion point ϕ_i is identified to the percolation concentration ϕ_c by : $$\phi_i = \phi_c = \frac{\log(n)}{\alpha}$$

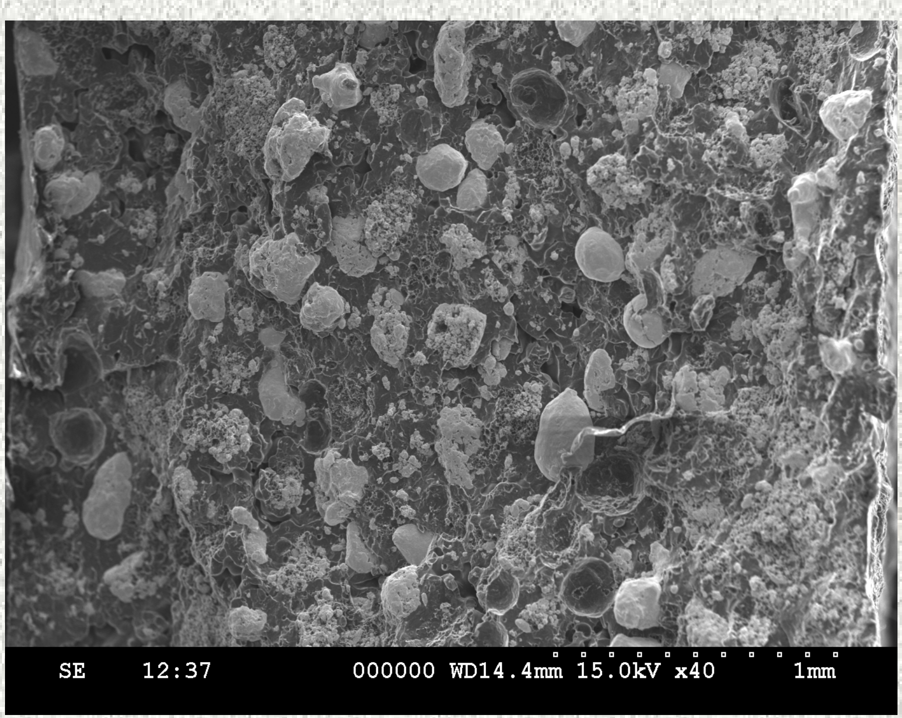


Fig. 1: SEM micrograph for 30% of Cu *(230µm)* volume fraction in Polypropylene matrix

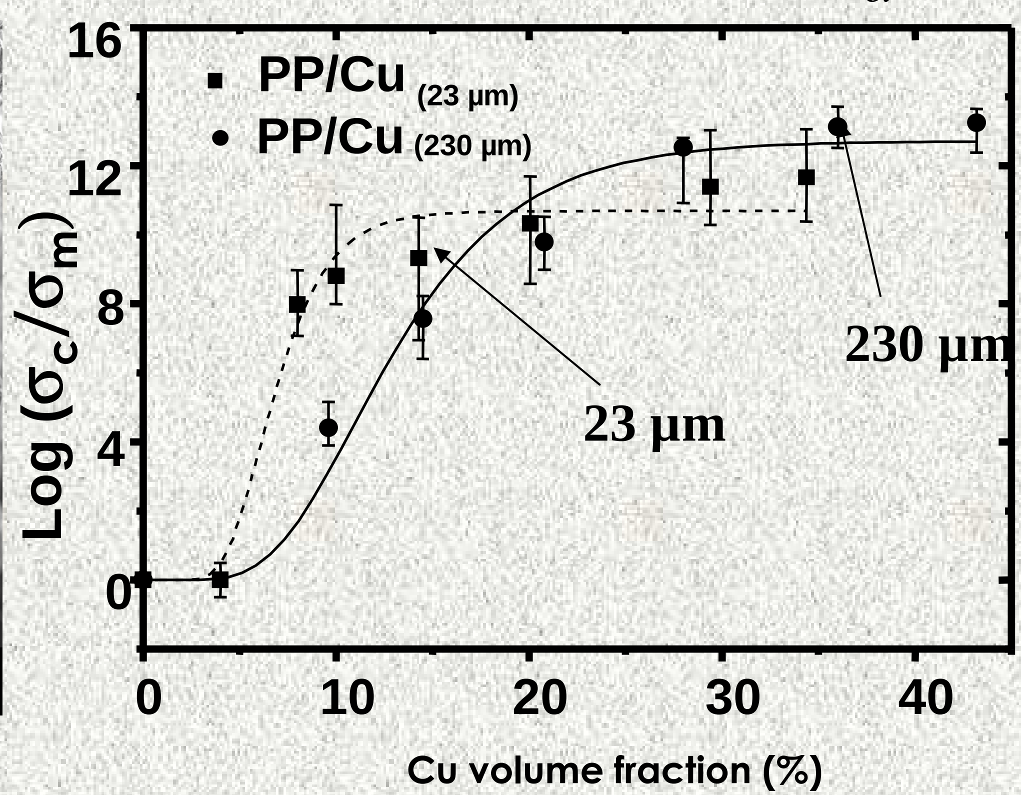


Fig. 2: Electrical conductivity of PP/Cu composites versus fillers volume fraction

Non linear raise in the composite thermal conductivity and diffusivity

$k_{PP} = 0.23$ W.m^{-1}.K^{-1}
$k_{Cu} = 389$ W.m^{-1}.K^{-1}

$a_{PP} = 1.67 \times 10^{-7}$ m^2.s^{-1}
$a_{Cu} = 1.14 \times 10^{-4}$ m^2.s^{-1}

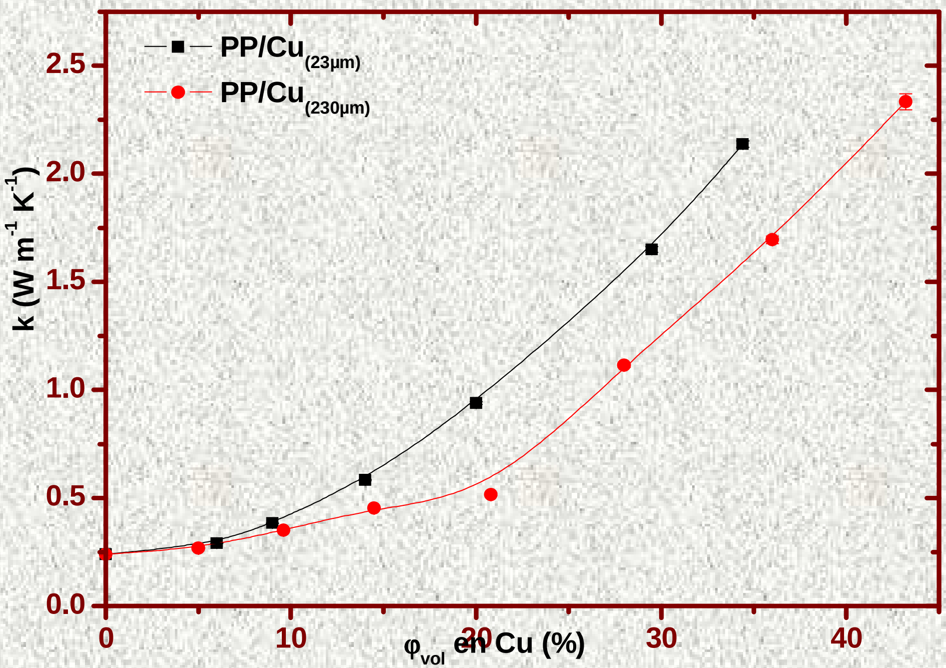


Fig. 3: Thermal conductivity of PP/Cu composites versus fillers volume fraction, measured at 25 °C

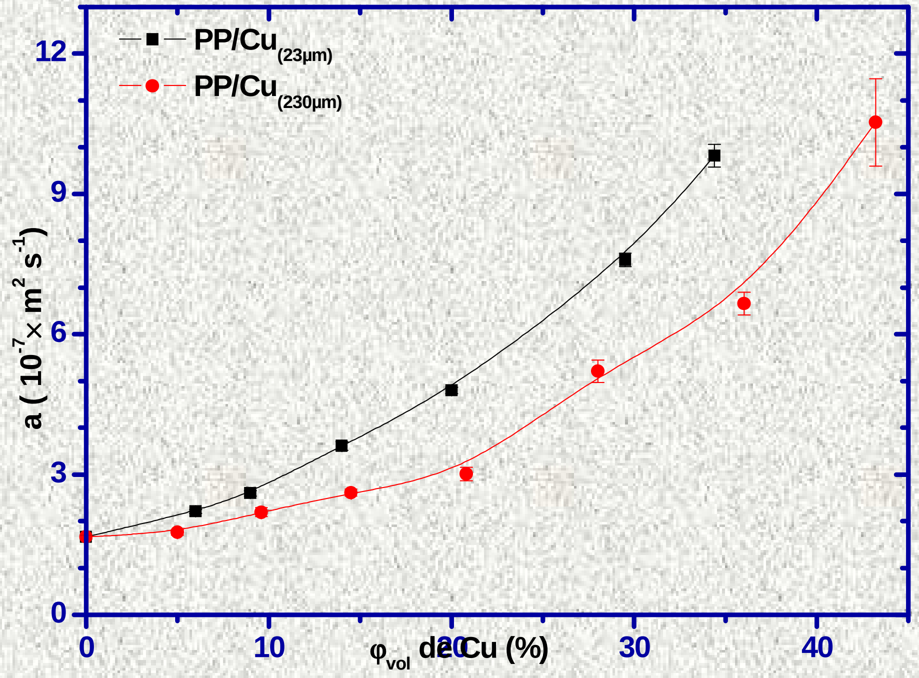


Fig. 4: Thermal diffusivity of PP/Cu composites versus fillers volume fraction, measured at 25 °C

- Oxidized Aluminum filler particles
- Electrical / insulator composites, but increase of K
- Opposite effect : thermal conduction is better by increasing the filler size

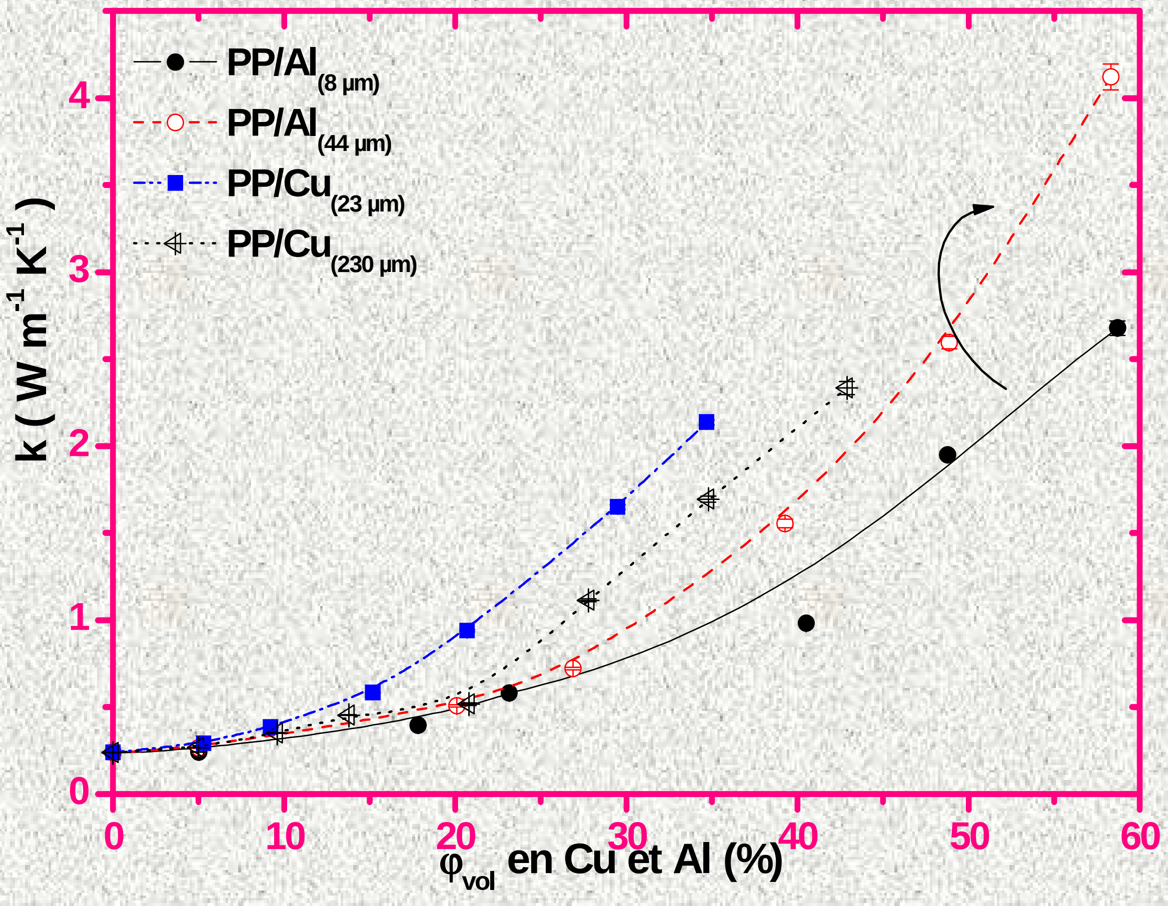


Fig. 5 : Comparison: PP/Cu and PP/Al - Oxidation Effect

Heat conductivity of Composite PVC / Multi walled Carbon Nanotubes : Collab. Institute of Macromolecular Chemistry, Kiev

1. The thermal conductivity decrease for $\varphi < \varphi_C$
2. Minimum for $\varphi = \varphi_C$
3. Linear increase for $\varphi > \varphi_C$: The composite remains thermally insulator

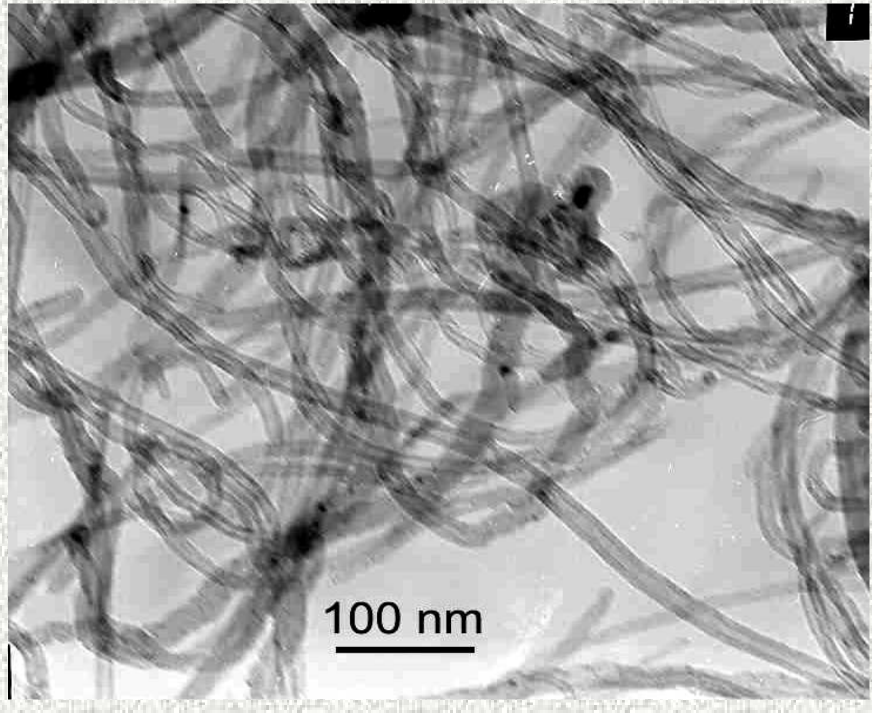


Fig. 6: Transmission electron microscopic image of multi-walled carbon nanotubes.

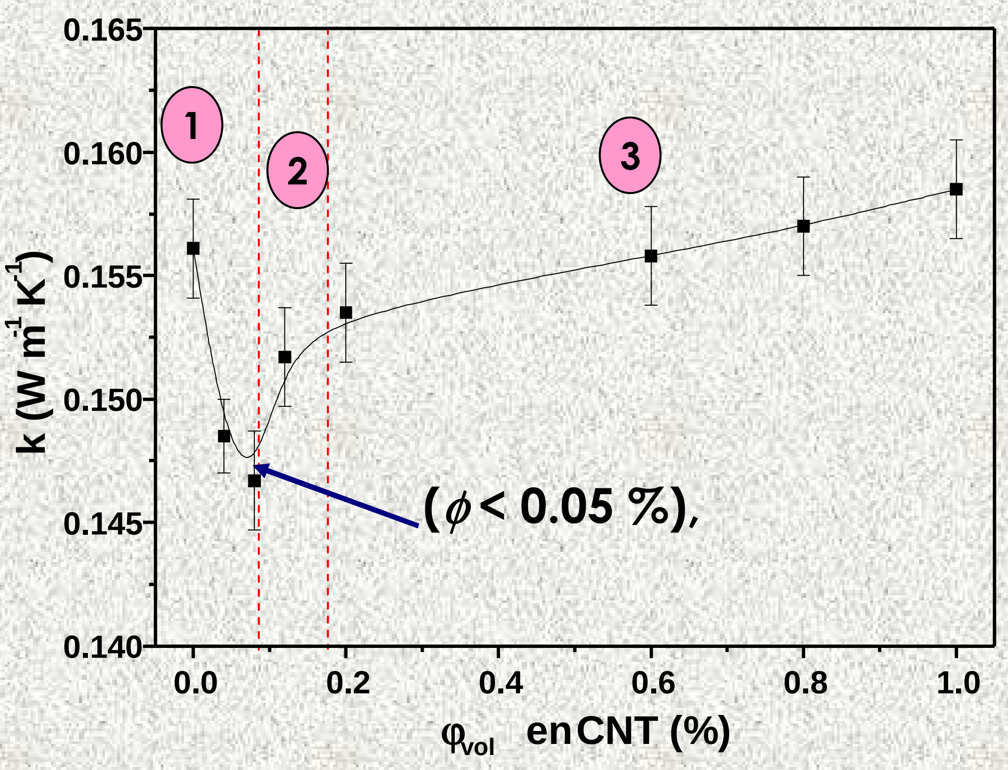
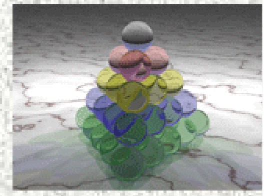


Fig. 7: Thermal conductivity of composites as a function of CNT volume concentration.

SUMMARY



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Composite EVA/ glass particles and EVA/ silver coated glass particles

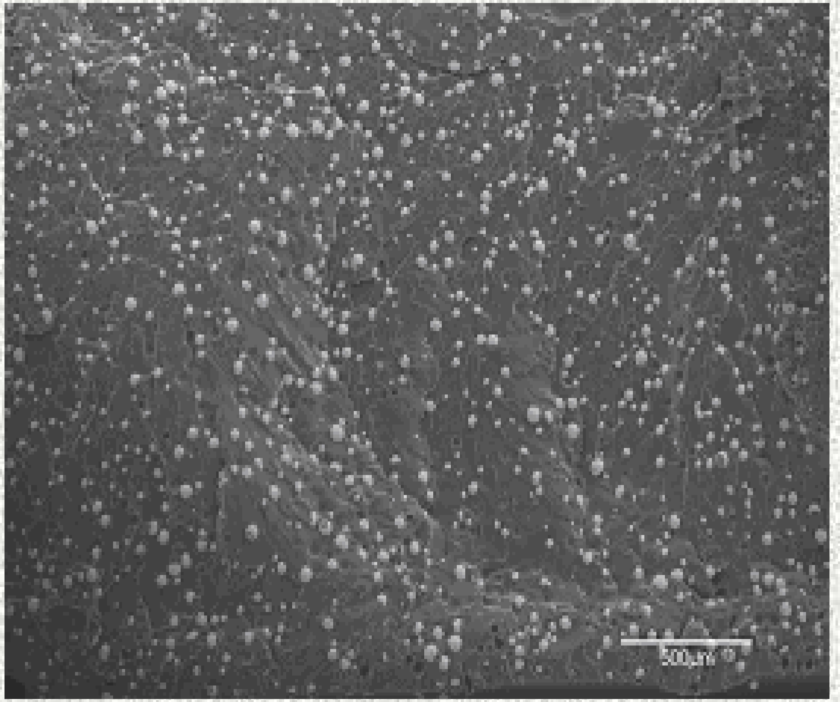


Fig. 8: EVA + SiO_2/Ag 47µm ($\varphi_{vol.}$ = 17%)

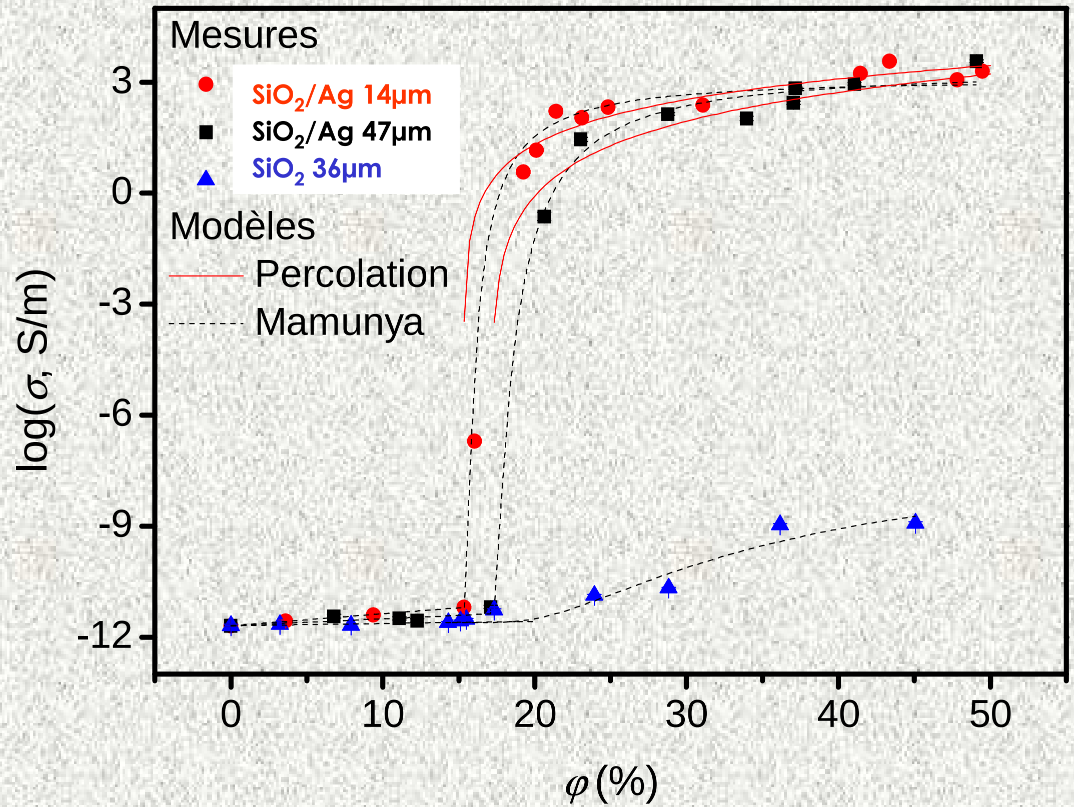
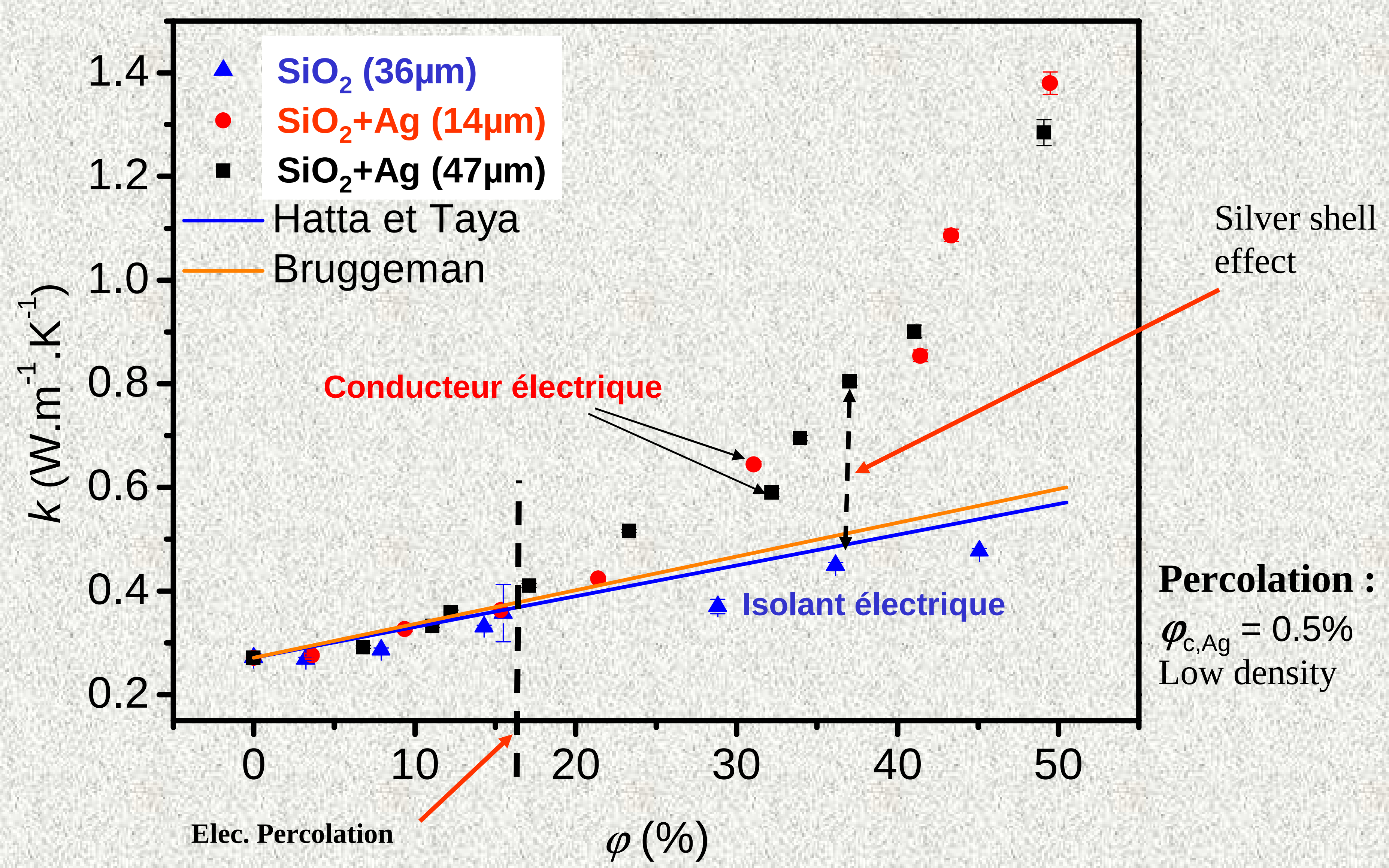


Fig. 9: Electrical conductivity versus fillers volume fraction

Fig.10: Thermal conductivity of EVA/ glass particles Composite : *Silver shell* effect



Percolation :

$\varphi_{c,Ag}$ = 0.5%

Low density

Composite HDPE/ silver coated PA particles

Collab. Polymer Institute, Bratislava

- Most of particles are well covered by silver; however some particles are insufficiently covered by silver.
- The silver shell The thickness was estimated from SEM measurements as lower than 1 µm.

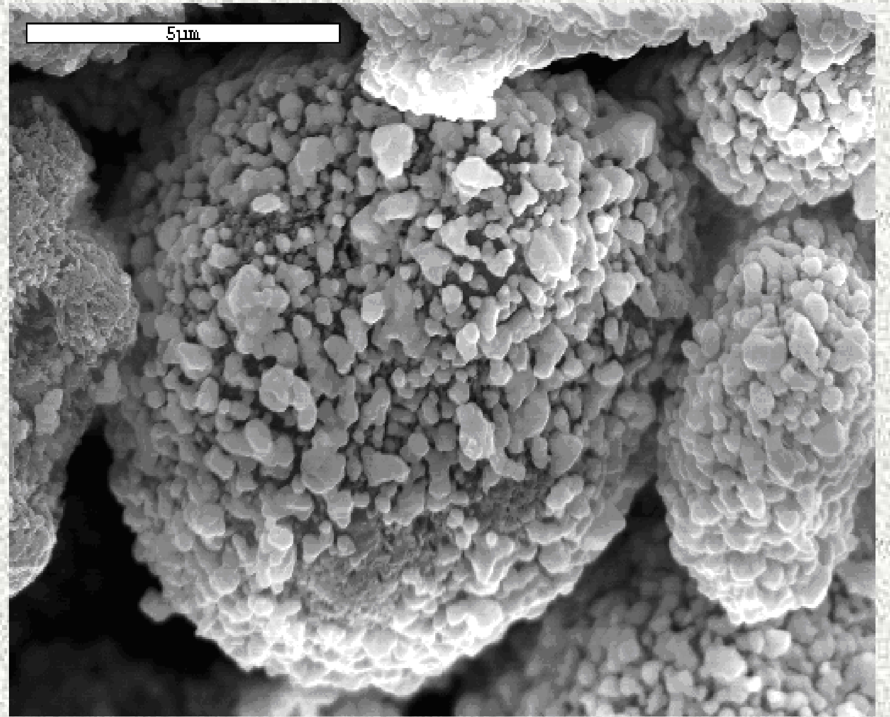


Fig. 11 : SEM micrographs of Ag coated PA particles (good coating)

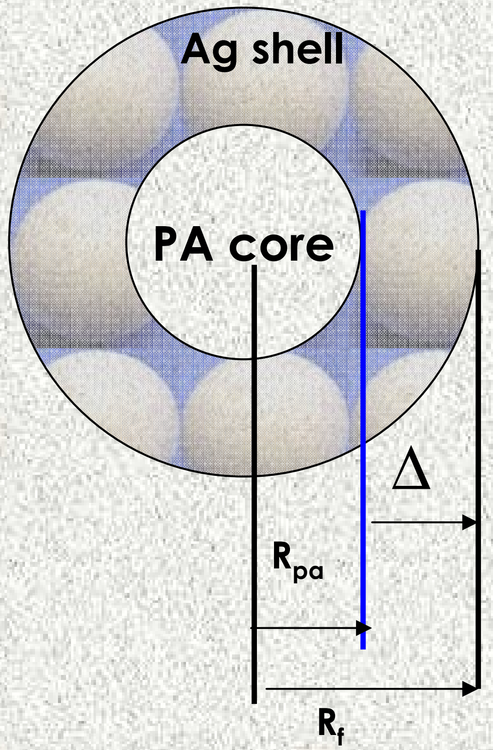


Fig. 12 : The model geometry of the coated particle.

3. Composite material : polymer matrix / insulating/ coated particles

- PA particles size : between 6 and 12 µm
- Percolation threshold (ϕ = 5% and Ag =1%)
- thermal conductivity Increase at low density

Electrical conductivity

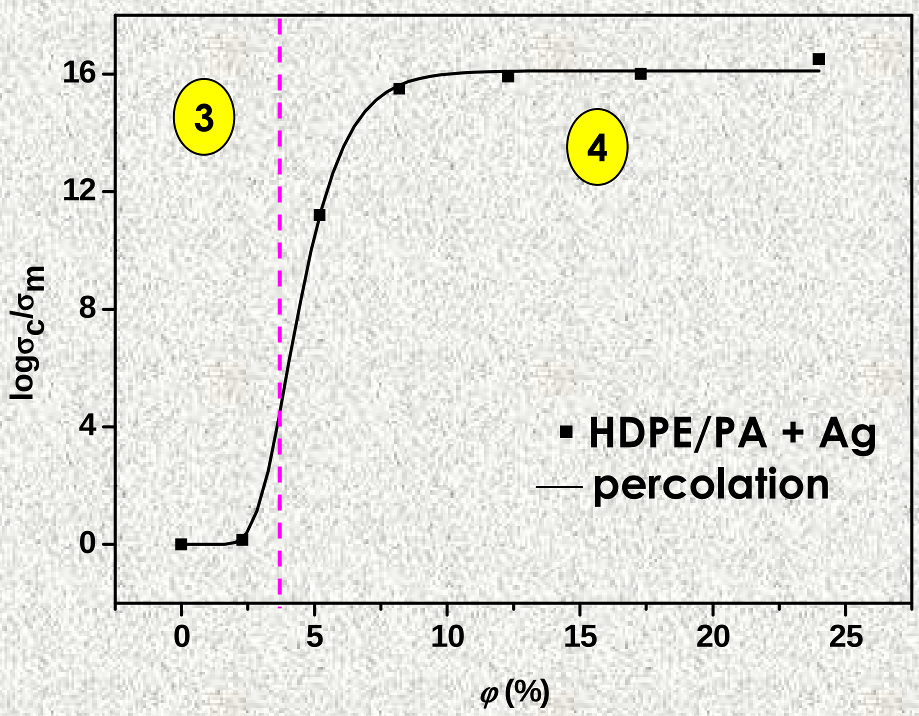


Fig. 13 : Electrical conductivity versus fillers volume fraction

Thermal conductivity

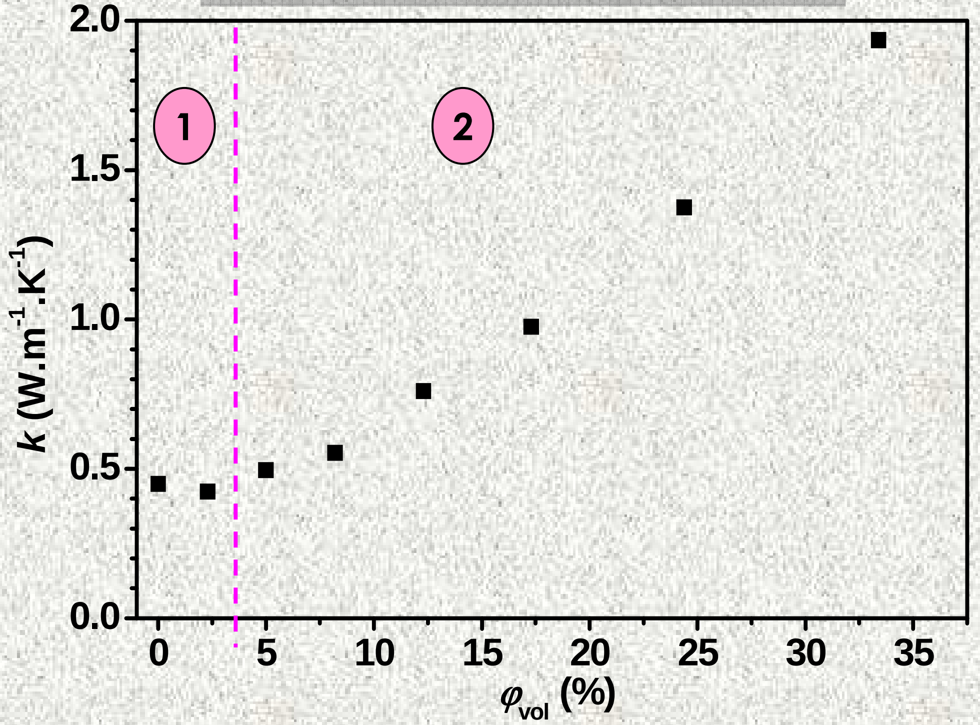


Fig. 14 : Thermal conductivity versus fillers volume fraction

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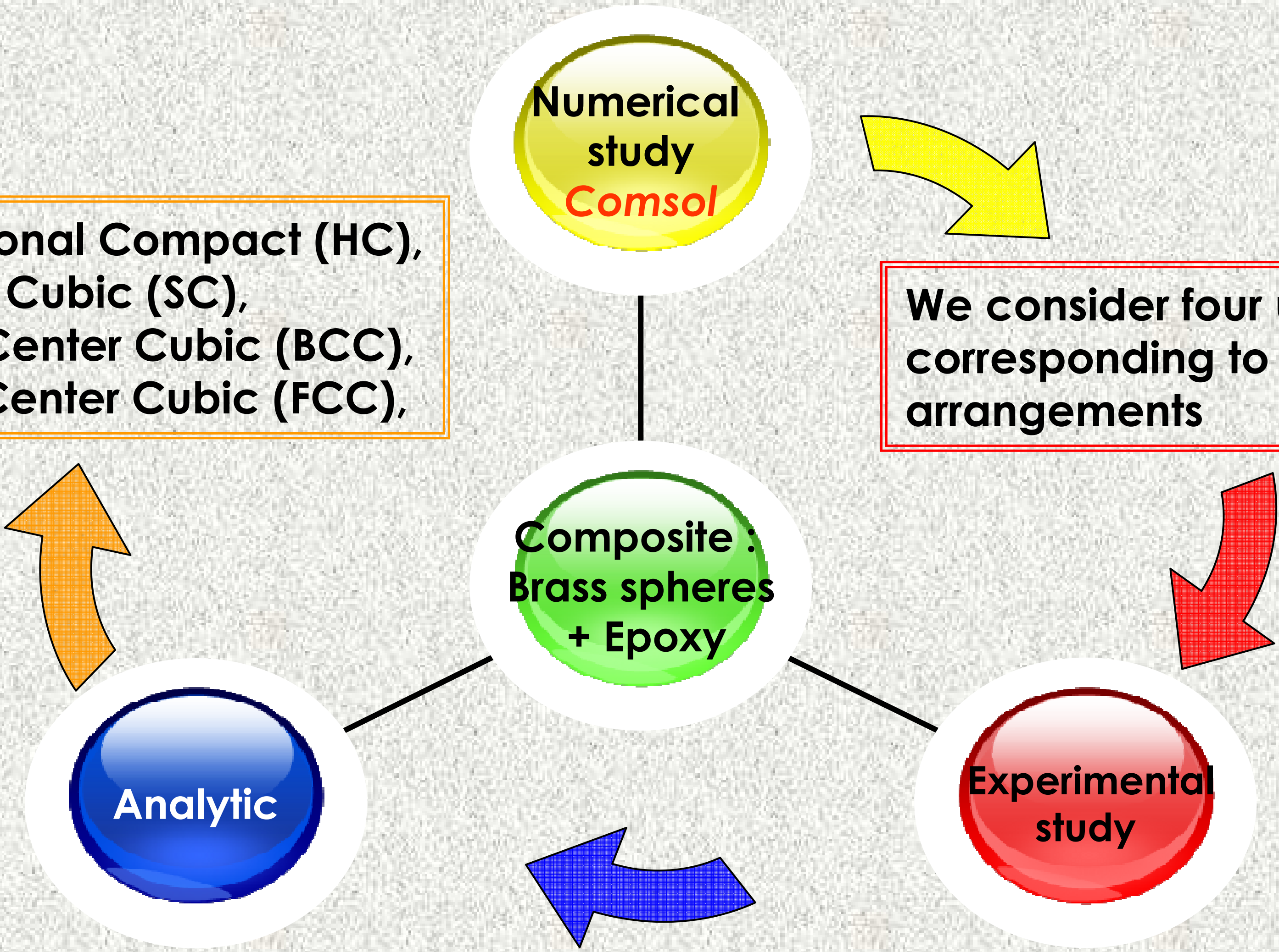
4. Composite material : polymer matrix/ brass spheres

Collab. B. Garnier, LTN

Hexagonal Compact (HC),
Simple Cubic (SC),
Body-Center Cubic (BCC),
Face-Center Cubic (FCC),

We consider four unit cell corresponding to the basic arrangements



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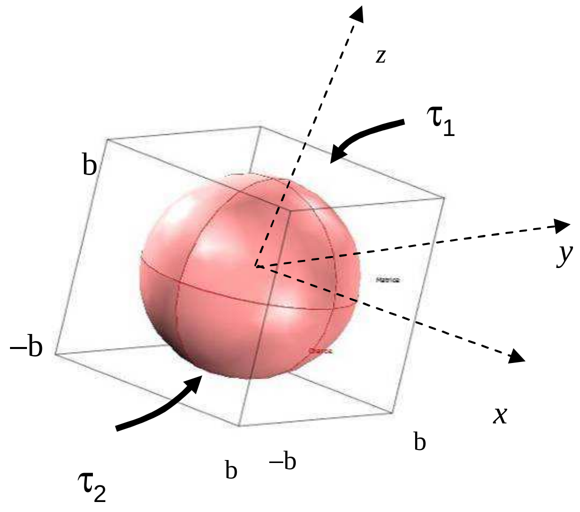
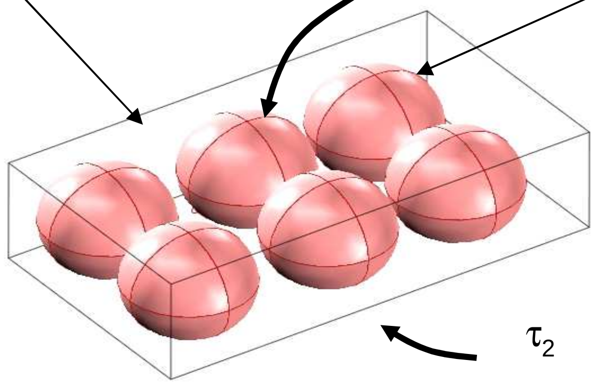
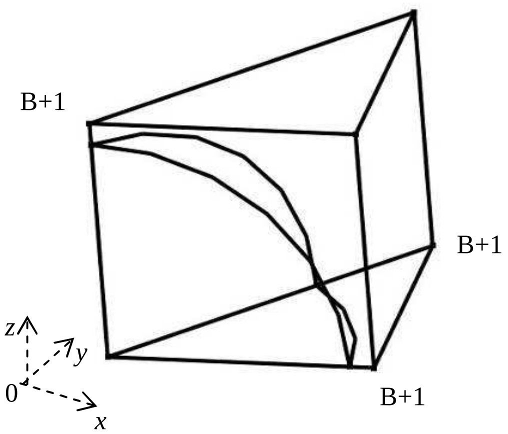




Simple Arrangement

Physical model Comsol

Fig. 13 : Computed elementary cell



Dimensionless parameters

$$D = \lambda_m / \lambda_f \qquad B = (2b - 2r)/2r$$

$$C = r_c \lambda_m / r \qquad Q = \int_0^1 \left(\int_0^{Y=X} \frac{\partial T}{\partial Z}\bigg|_{Z=B+1} dY \right) dX$$

$$E = \lambda_{eff} / \lambda_m$$

Numerical study : Simple Cubic (SC) arrangement

$$Q = \int_0^1 \left(\int_0^{Y=X} \left. \frac{\partial T}{\partial Z} \right|_{Z=B+1} dY \right) dX$$

The Thermal Effective Conductivity

$$E = 2Q/(1+B)$$

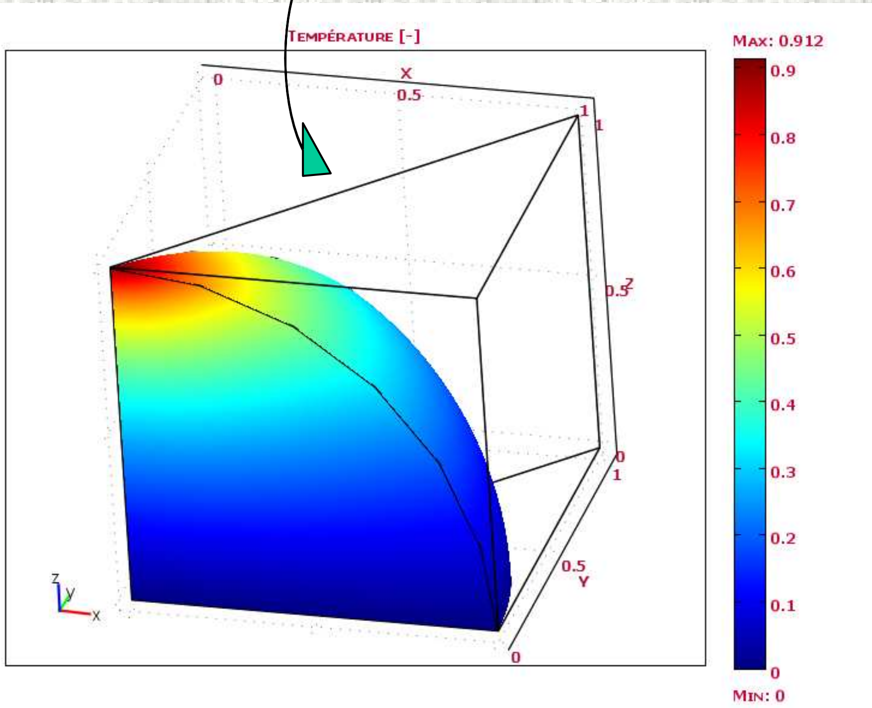


Fig. 16 : Temperature profile (SC) model

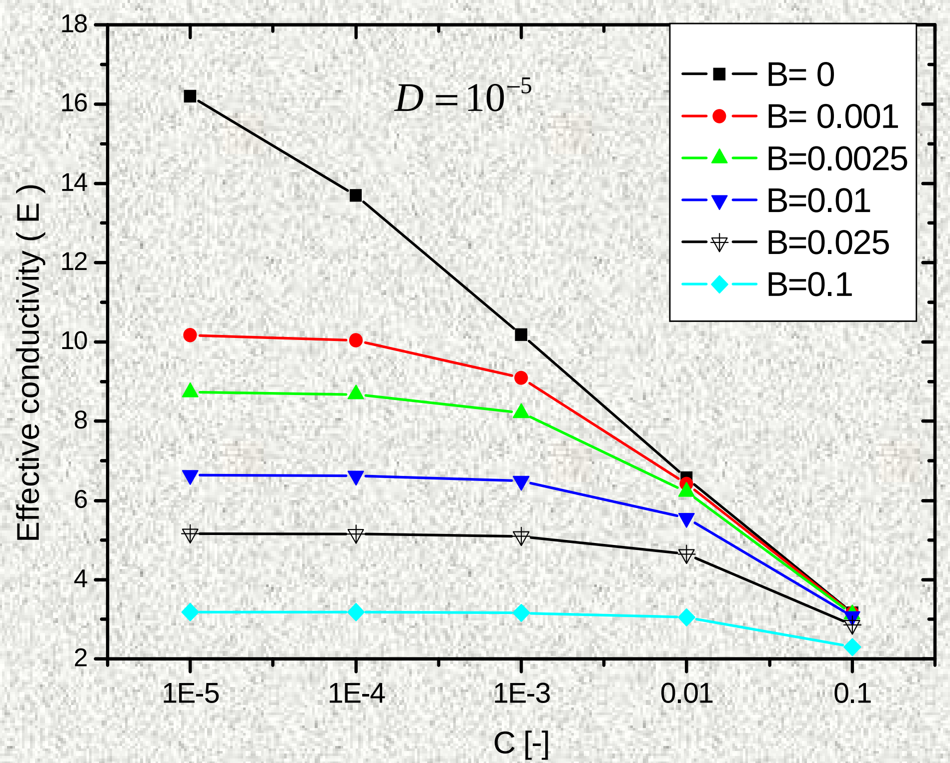


Figure 17 : Evolution of the ETC versus C et B, CS

Numerical study : Face-Center Cubic (FCC), arrangement

$$Q_T = \int_0^1 \left(\int_0^{Y=X} \left.\frac{\partial T}{\partial Z}\right|_{Z=B+1} dY \right) dX$$

The Thermal Effective Conductivity

$$E = 2Q_T$$

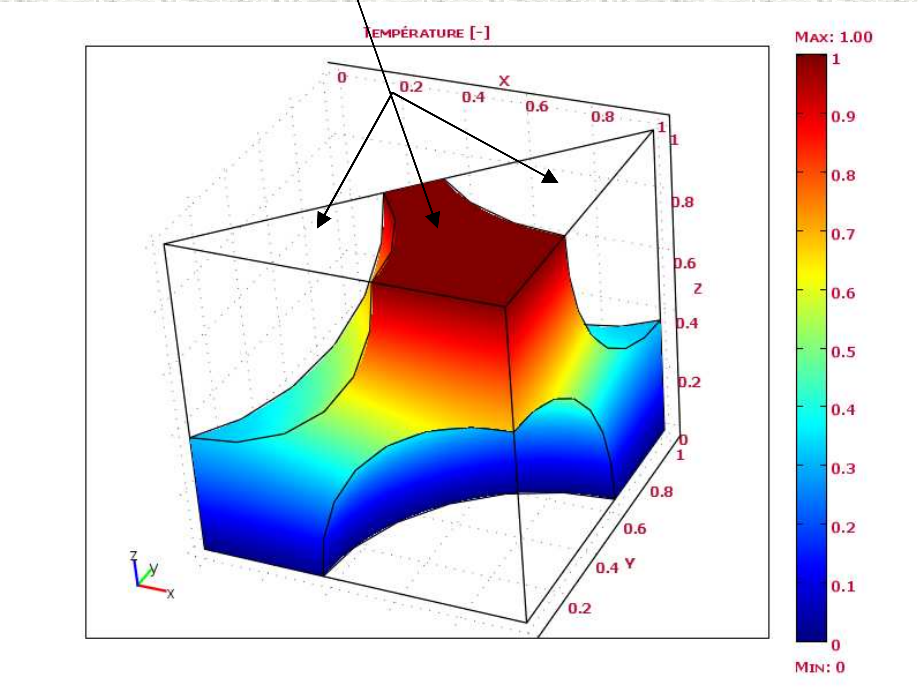


Fig. 18 : Temperature profile (FCC) model

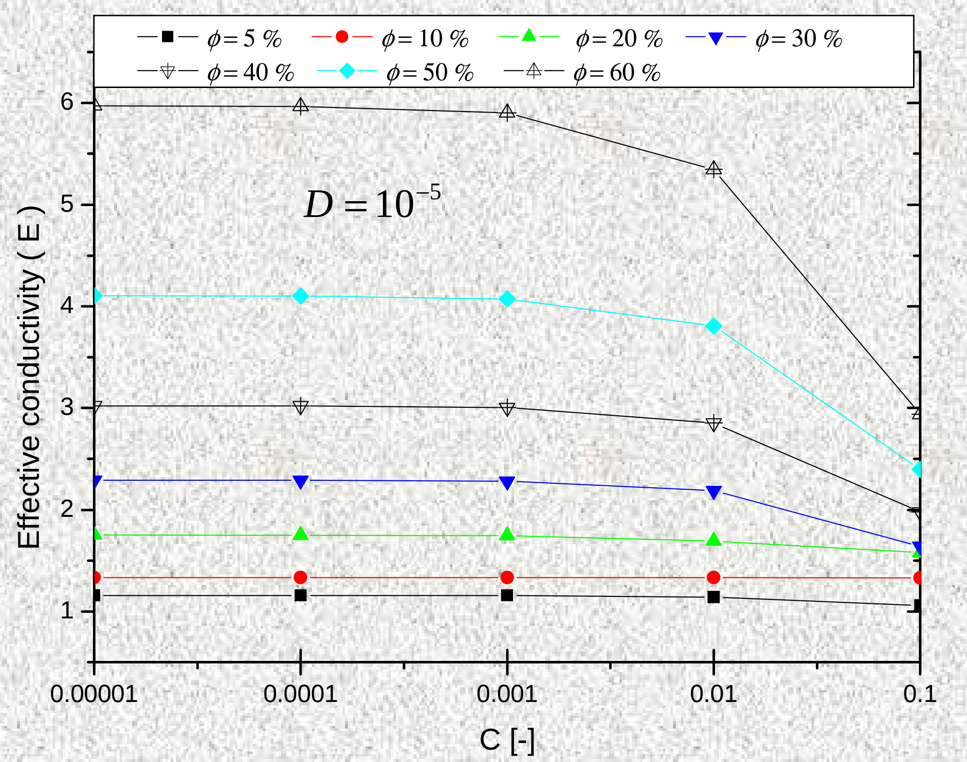


Fig. 19 : Effect of the thermal contact resistance on the ETC

Comparison : HC, SC, FCC and BCC models

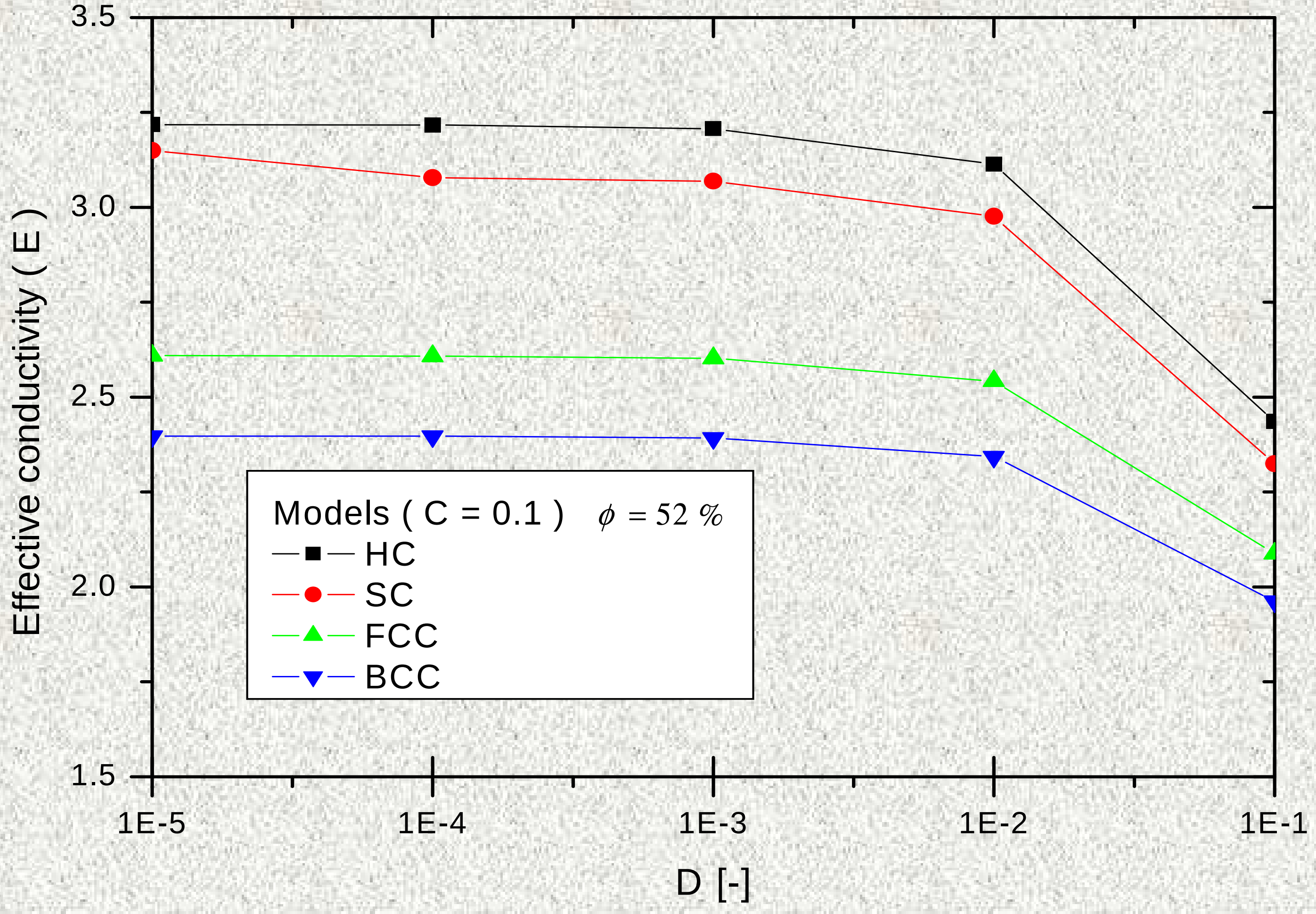


Fig. 20 : Effect of the D parameter on the ETC

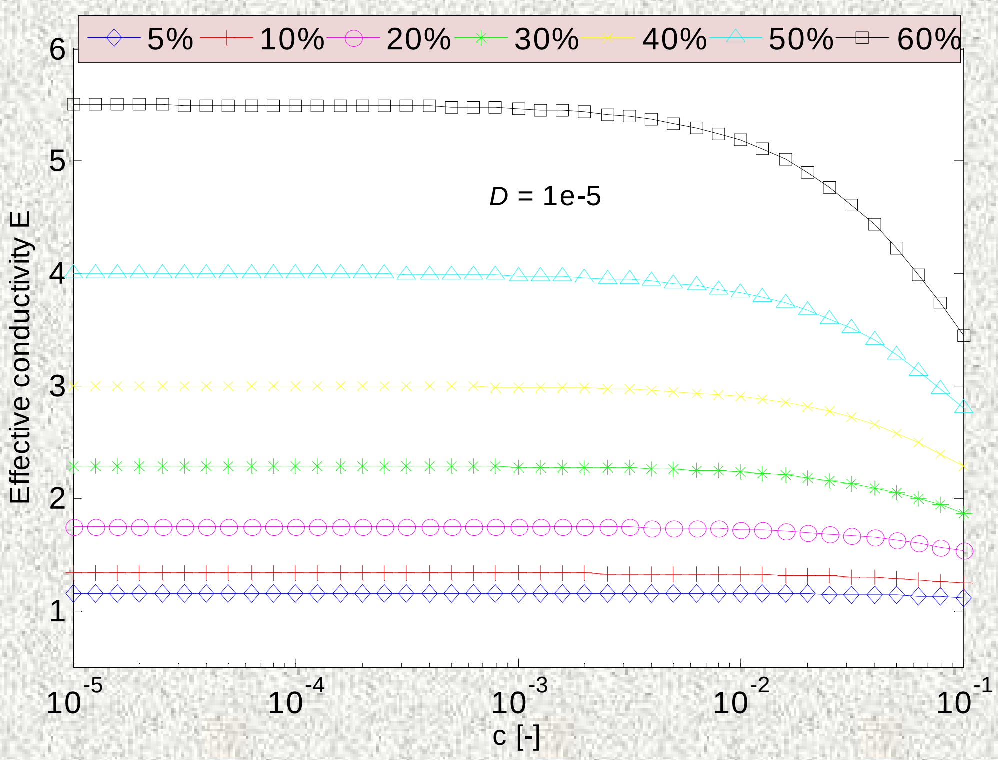
4. Composite material : polymer matrix/ brass spheres

Analytical and Numerical predictions models :

$$E = \frac{1 + (d-1)\phi\,\overline{\beta}}{1 - \phi\,\overline{\beta}}$$

$$\overline{\beta} = \frac{\frac{1}{D} - \left(1 + \frac{C}{D}\right)}{\frac{1}{D} + (d-1)\left(1 + \frac{C}{D}\right)}$$

Hashin & Sthrikman model



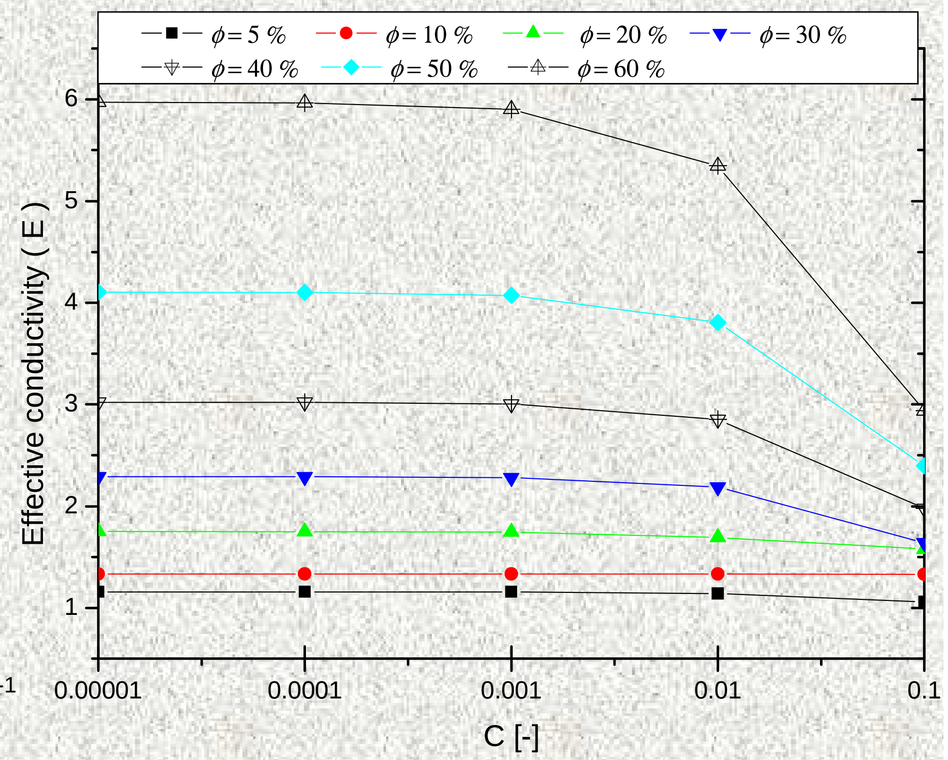


Fig. 21 : Effect of the thermal contact resistance on the ETC (FCC)

Experimental study : SC, HC, FCC arrangement

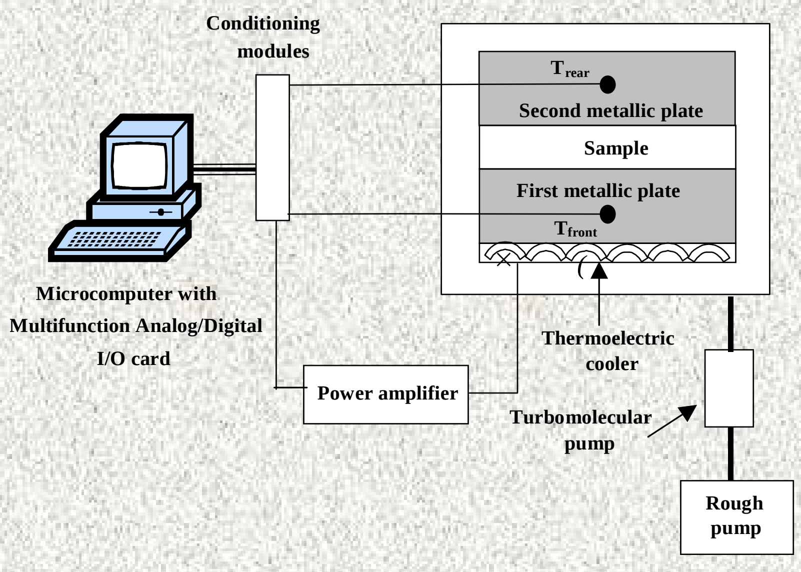
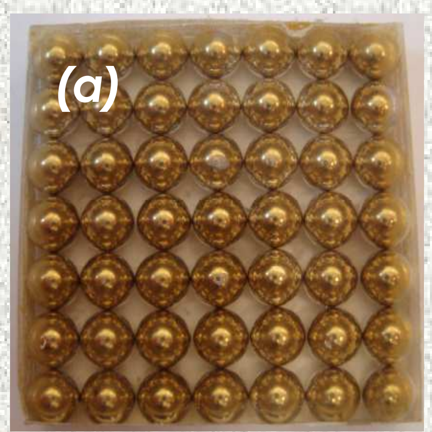


Fig. 22: Sample (c): resin +brass spheres of diameter 6.35 mm

Model	E_{ex}
Sample (a)	4.93
Sample (b)	5.85
Sample (c)	9.66



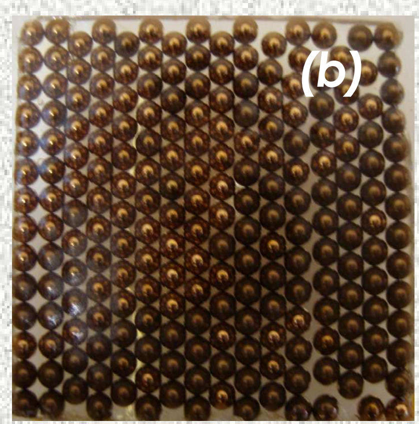


Fig. 23 : Sample (a) and (b), resin +brass spheres of diameter 6.35 mm (a) and 3.18 mm (b)

Experimental and Numerical results

Model	α_{ex} ($10^{-7}m^2s^{-1}$)	ϕ (%)	B	E_{ex}	E_c	$(E_{ex}-E_c)/E_c$ (%)
SC : Sample (a)	4.21 ±0.21	49	0.031	4.93	4.96	0.6
HC: Sample (b)	2.93± 0.12	55	0.022	5.85	6.04	3.14
FCC: Sample (c)	11.97 ± 0.66	57	-	9.66	9.52	1.52

Table 1 : The measured parameters

The control of ϕ, may be very important to control the composite effective thermal conductivity

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Effet de la taille et de la forme des charges

- **A φ constant, *k* et *a* augmentent**

Diminution très importante du seuil de percolation électrique pour des nanotubes sans augmentation de la conductivité thermique

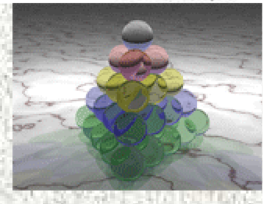
5. Conclusion

Effet de la surface des charges

- **Charges métalliques oxydées (PP/Alu):**
  - ✓ augmentation de la conductivité thermique
  - ✓ matériau isolant électrique
- **Charges isolantes métallisées :**
  - ✓ augmentation importante des conductivités thermique et électrique
  - ✓ intérêt : diminution de la quantité de métal au seuil de percolation donc une réduction de la masse volumique

Etude Numérique (composites conducteurs)

- ✓ au-delà de $D = 10^{-3}$, la CTE augment très peu.
- ✓ Influence des paramètres prépondérants (*B* et C)
- ✓ alternative aux systèmes énergivores



Merci pour votre attention