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Elaboration of dense ceramics and glass ceramics piezoelectric coatings by selective laser treatments

POLYTECH MONS

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Research context

Selective Laser Sintering of ceramic powder coatings for electric applications

- Manufacture of 2D or 3D components
- Alternative route to conventional sintering in the manufacture of hybrid circuits

Advantages:

- Selectivity
- Rapidity and versatility of process

Application to $BaTiO_3$ (piezoelectric components, capacities,…)

Presentation plan

I. Theoretical aspects

I.1 Laser treatment of a surface

I.2 Sintering of a coating

II. Experimental results

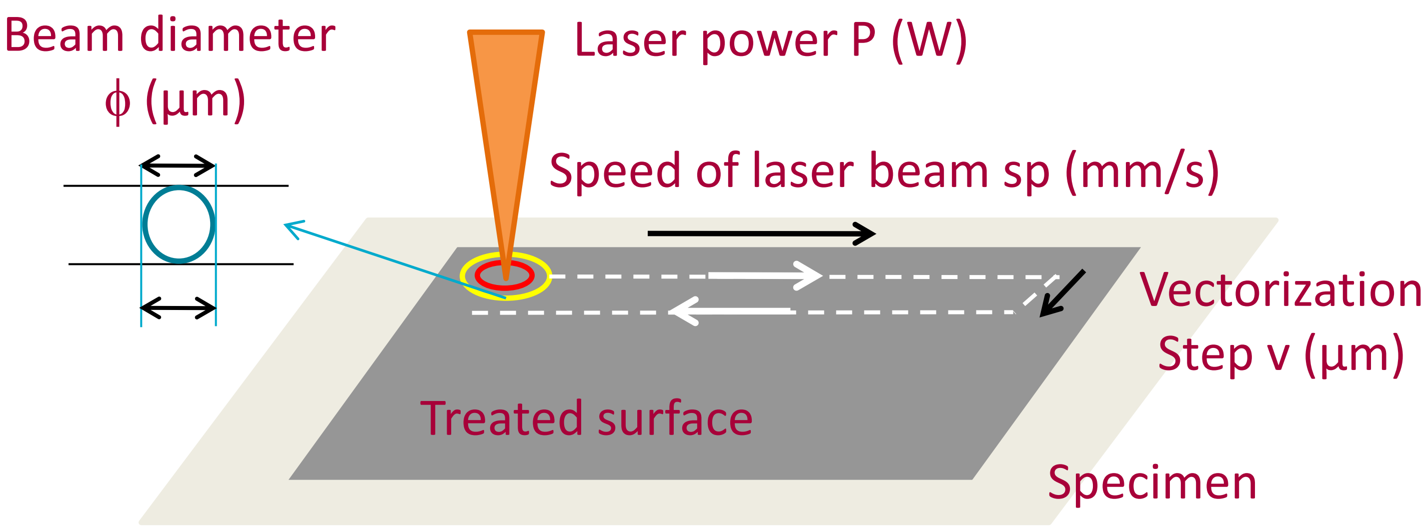
II.1 Treatment of $BaTiO_3$ compacts

II.2 Treatment of $BaTiO_3$ coatings

Conclusions and outlooks

I.1 Laser treatment of a surface

Scan parameters



Powder density (W/mm^2) : $P_w = P / \phi^2$

Interaction time (s) : $t = \phi / sp$

Energy density (J/mm^2) : $J = P / (sp \cdot \phi)$

$P_w = J / t$

I.1 Laser treatment of a surface

Scan parameters

Laser characteristics:

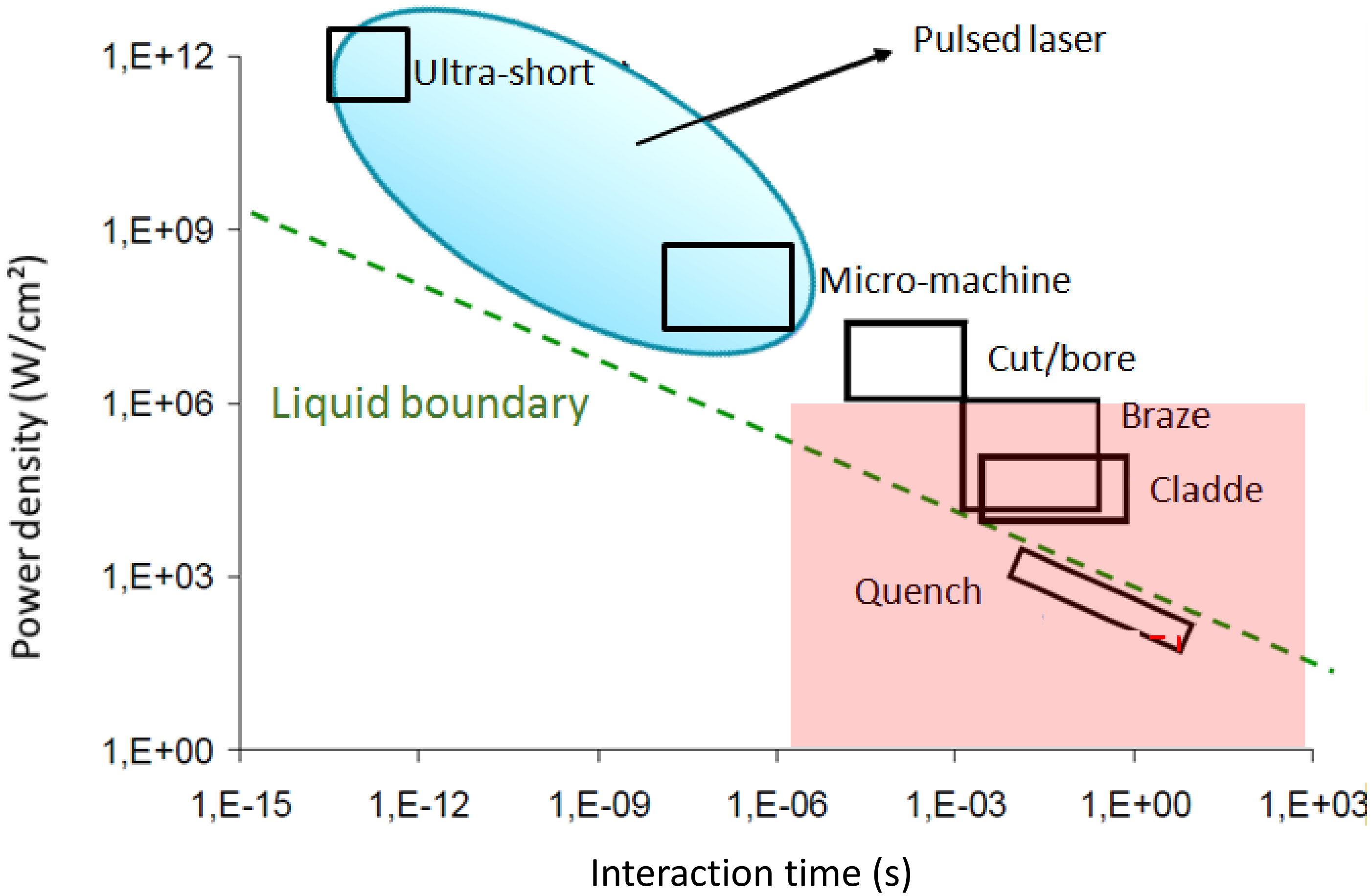
Laser Trumark Station 5000 (Trumpf)

- Solid-state laser (Nd-YVO_4)
- P_{max} = 22,8 W (cw)
- λ = 1064 nm
- Beam: TEM_{00} (Gaussian)
- ϕ = 45 µm
- Sp = 0 to 1500 mm/s
- P_w max = 10^6 W/cm^2
- t min = 30 µs

I.1 Laser treatment of a surface

Scan parameters

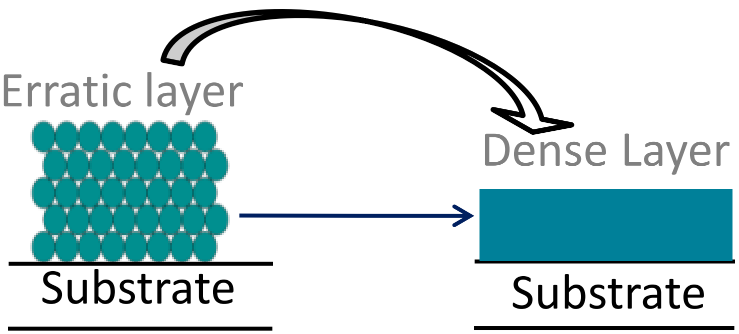
Sintering ?



Laser Cladding: Jan Gedopt, Marleen Rombouts *Lasercentrum Vlaanderen*

I.2 Sintering of a coatings

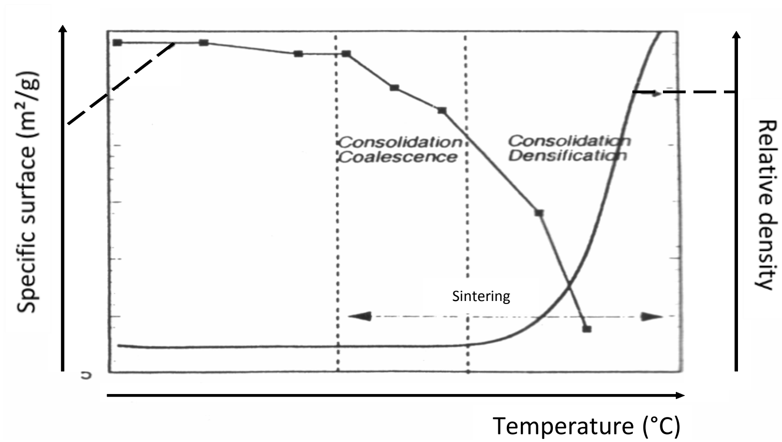
Mechanisms



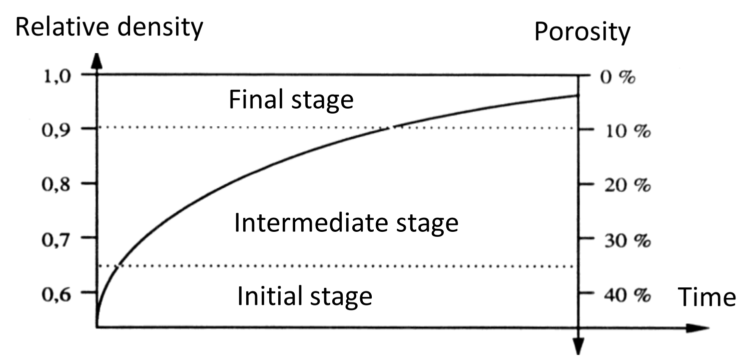
Sintering steps → Consolidation
Sintering steps → Densification
Sintering steps → Grain growth

Parameters

Temperature



Time



I.2 Sintering of a coatings

Specific problems due to laser heating

- Mastering of the real temperature seen by the powder grains

  → function of the absorption efficiency

- Short interaction time

  → thermal gradient between surface and center of the grains (> 1000°C / µm)

  → not compatible with slow kinetics (i.e solid state densification by diffusion)

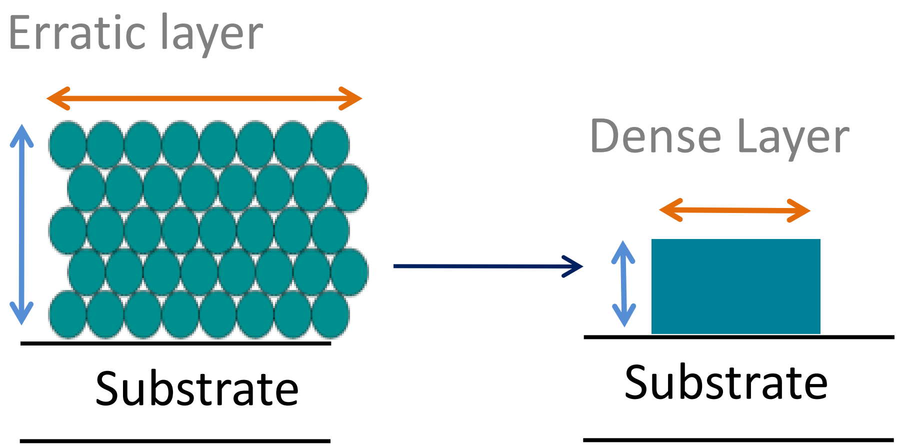
- Fast local heating and cooling

  → thermal chocks

I.2 Sintering of a coatings

Specific problems due to coating / substrate interaction

1. Shrinkage during sintering



<u>Investigation:</u>

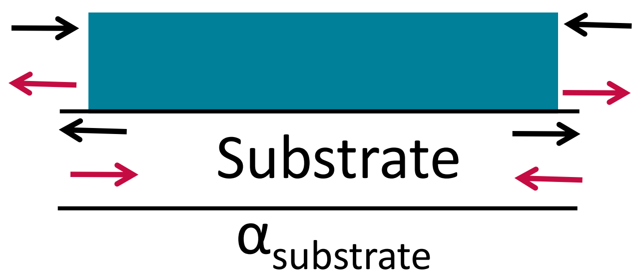
↗ initial compacity of the green coating

→ Mixture of 2 granulometric distributions of powders (micro and nano)

I.2 Sintering of a coatings

Specific problems due to coating / substrate interaction

2. Dilatometric mismatch



Depending on the respective compositions of the coating and of the substrate

$\alpha_{\text{ substrate}} < \alpha_{\text{ coating}}$: coating in tension

$\alpha_{\text{ substrate}} > \alpha_{\text{ coating}}$: coating in compression

$\alpha_{Al_2O_3}$ (25°C) $= 7{,}1\ 10^{-6}\ K^{-1}$

α_{BST} (25°C) $= 8{,}0\ 10^{-6}\ K^{-1}$

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Conclusions and outlooks

II.2 Treatment of $BaTiO_3$ compacts

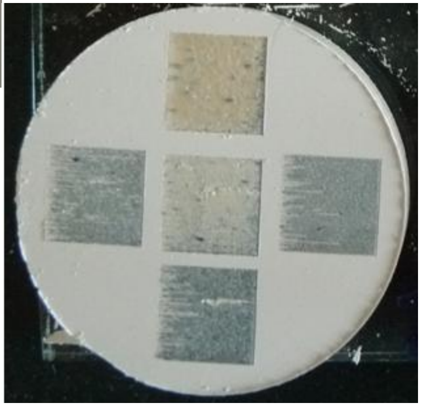
Experimental procedure

Tetragonal $BaTiO_3$ Powders :

- micro, d_{50} = 0,44 µm, d_{90} = 2,40 µm (Sigma Aldrich, 99%)
- nano, d_{50} = 50 nm (Inframat Advanced Materials, 99,95%)

Preparation of the compacts:

- mixing of powder in isopropanol
- magnetic stirring
- evaporation of isopropanol
- powder compaction (80 MPa)
- laser scan



II.2 Treatment of $BaTiO_3$ compacts

Experimental procedure

Parametric investigation :

- Ratio micro/nano : 100/0 ; 85/15; 70/30 ; O/100
- Laser power : 100 % (23 W) ↓
- Scan speed : 1500 mm/s ↓
- Vectorization step : 10, 20, 40 µm

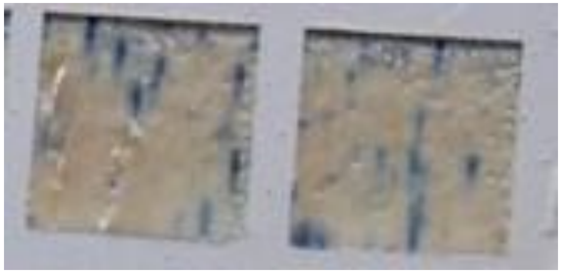
Characterization

- Optical observation → aspect of coating, selection for other analysis
- XRD → $BaTiO_3$ forms, other crystalline or amorphous phases
- SEM →Microstructure, porosities, cracks…

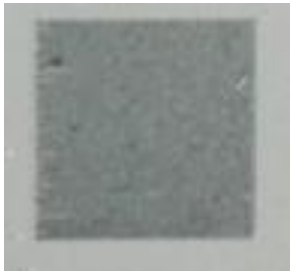
II.2 Treatment of $BaTiO_3$ compacts

Optical observation

70/30 compact: P100vect10



Sp 70 Sp80



Sp1000

Sp1500

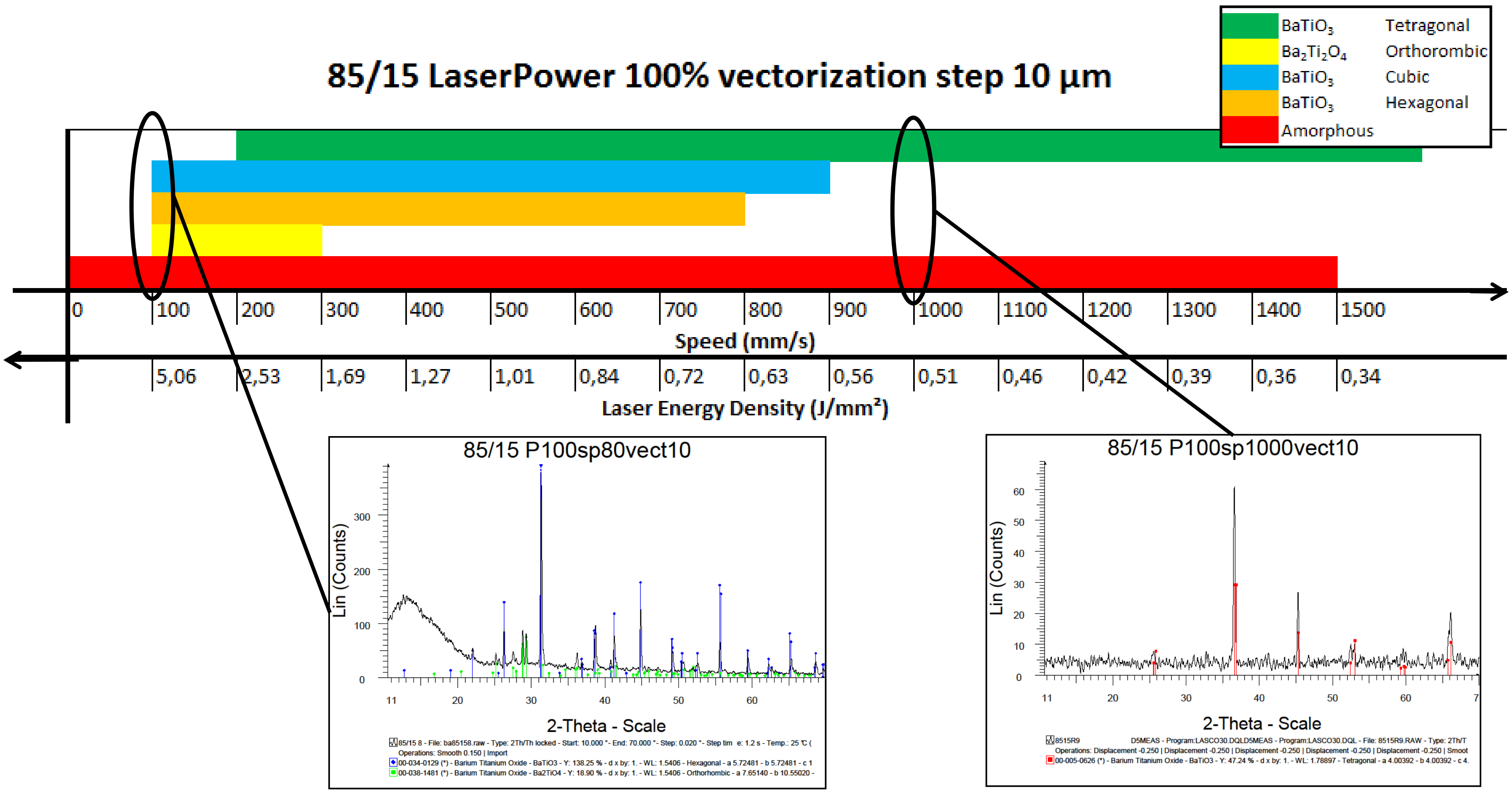
XRD analysis

Crystalline phase of $BaTiO_3$ in function of temperature

Orthorhombic $\xrightarrow{0°C}$ **Tetragonal** $\xrightarrow{130°C}$ Cubic $\xrightarrow{1460°C}$ Hexagonal $\xrightarrow{1650°C}$ liquid

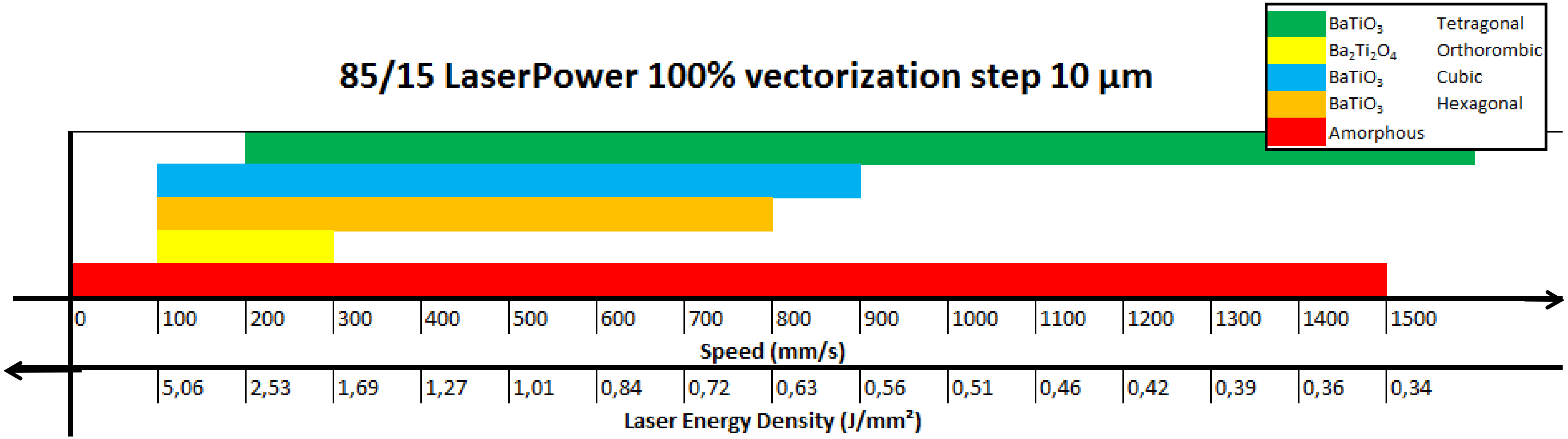
II.2 Treatment of $BaTiO_3$ compacts

Influence of scan speed → XRD



II.2 Treatment of $BaTiO_3$ compacts

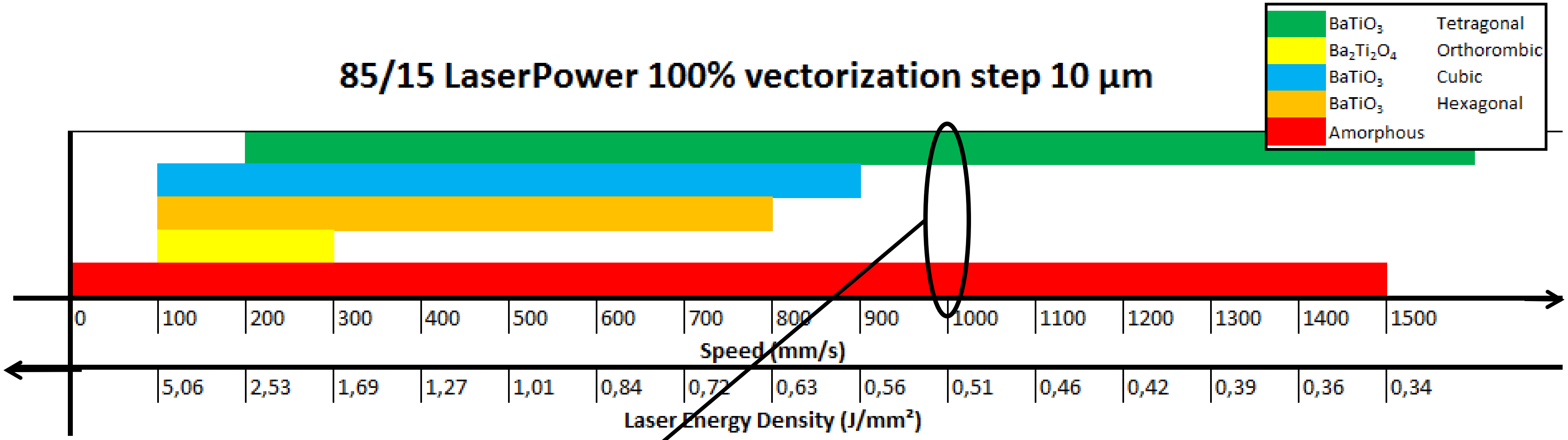
Influence of scan speed → XRD



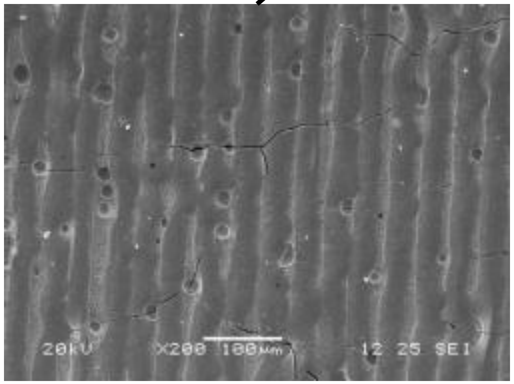
1) Only tetragonal $BaTiO_3$ for sp > 900 mm/s
2) High temperature forms of $BaTiO_3$ when sp ↘
3) Amorphous phase for all sp but very low amount when sp ↗ 1000mm/s

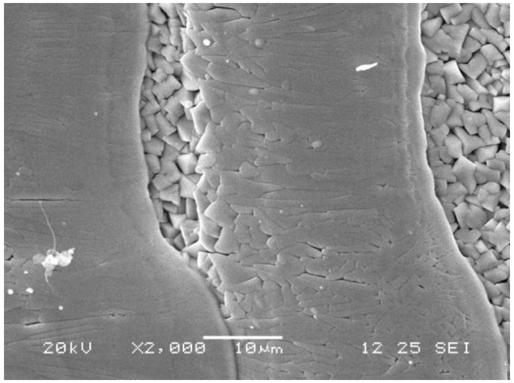
II.2 Treatment of $BaTiO_3$ compacts

Influence of scan speed → SEM



Alternation of dense and less dense stripes



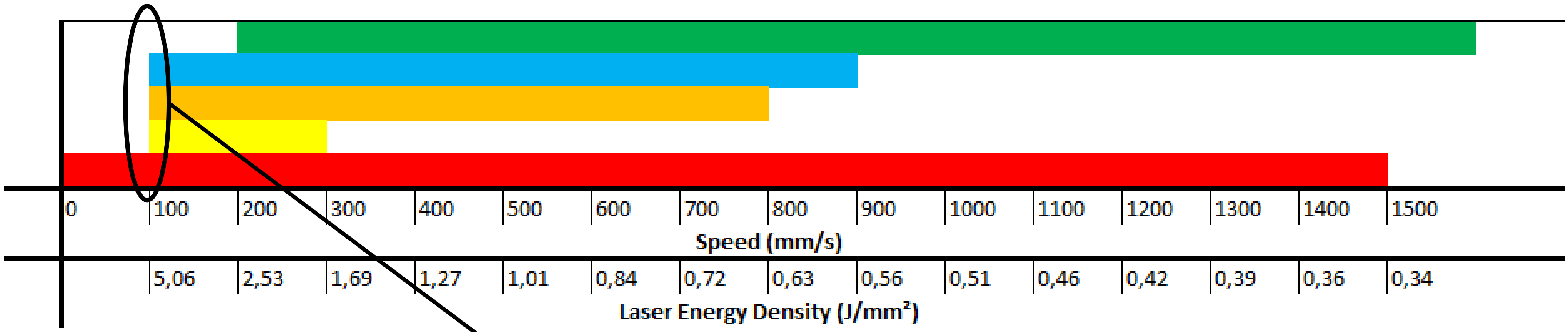


Elongated and Equiaxis grains

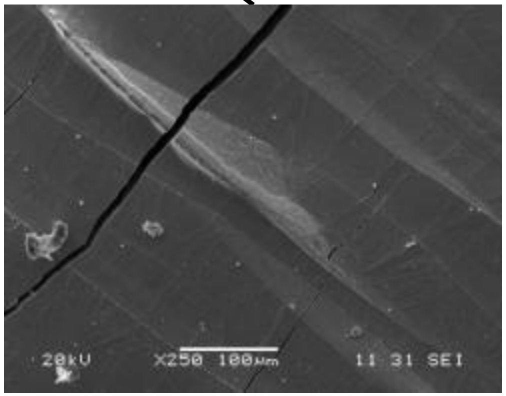
II.2 Treatment of $BaTiO_3$ compacts

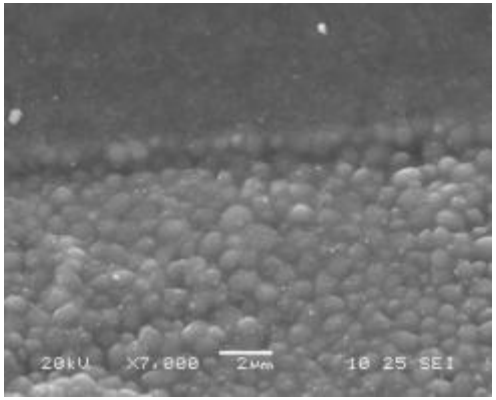
Influence of scan speed → SEM

85/15 LaserPower 100% vectorization step 10 µm



Surface dense and glassy





Region of grains embbeded in the amorphous phase

II.2 Treatment of $BaTiO_3$ compacts

Influence of scan speed → Discussion

Speed influences J and then :

The amount of liquid under laser TT (↓ when sp ↑)

→ low sp = melting nano grains and significant part of micro grains

→ high sp = melting of nano grains and very near surface of micro grains

The temperature of the center of the micro grains (↓ when sp ↑)

→ low sp = phase transformation of $BaTiO_3$

Remarque : due to fast cooling no reverse transformation of BaTiO3

→ high sp = no phase transformation of $BaTiO_3$

II.2 Treatment of $BaTiO_3$ compacts

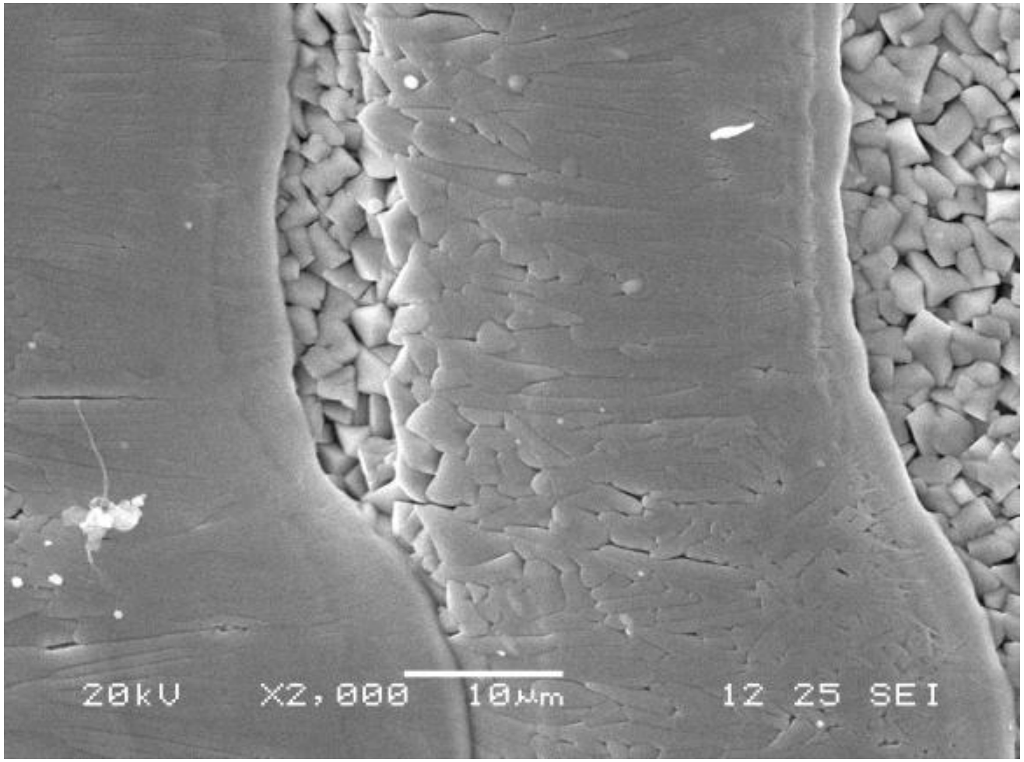
Influence of scan speed

→ Discussion

High sp microstructure :

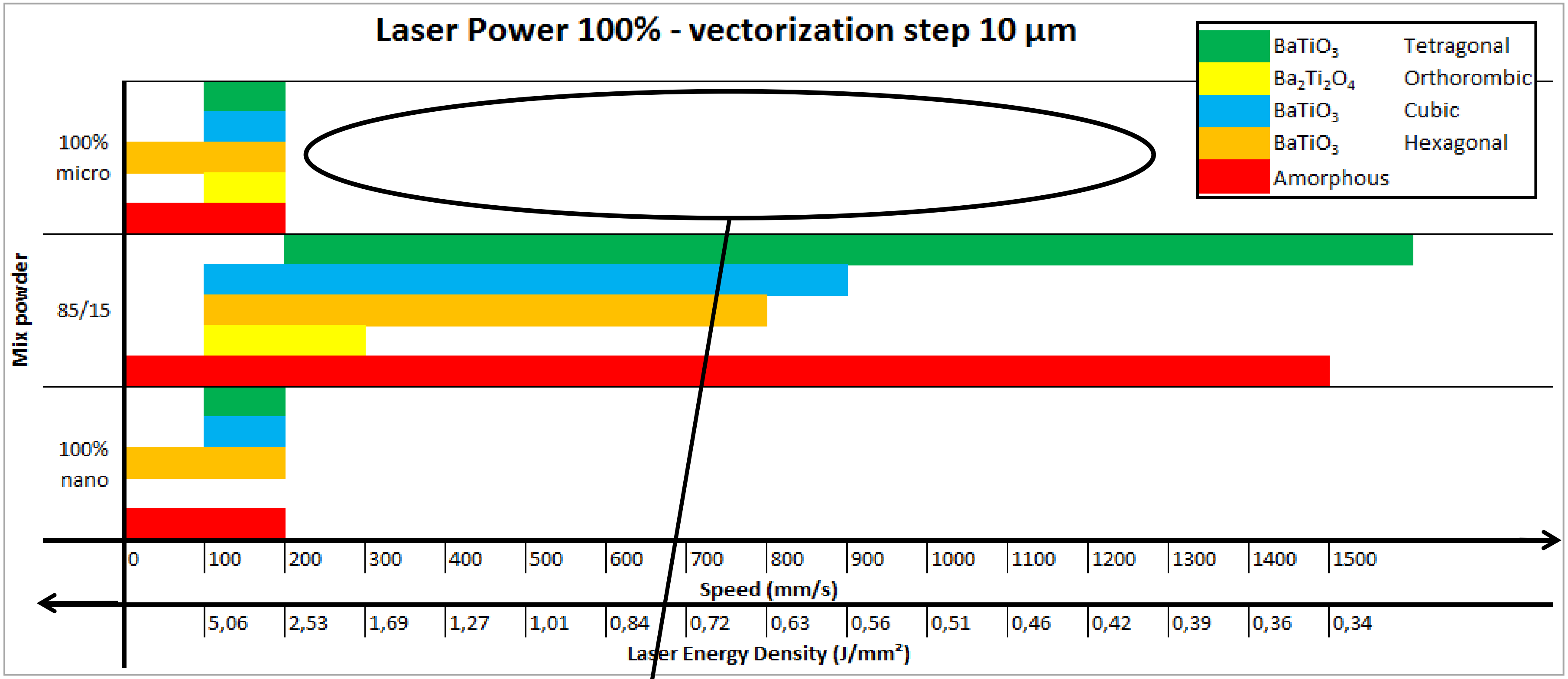
Low ratio liquid / solid

→ Grain growth in the liquid phase of the unmelted grains



II.2 Treatment of $BaTiO_3$ compacts

Comparison mix micro + nano / pure micro → XRD

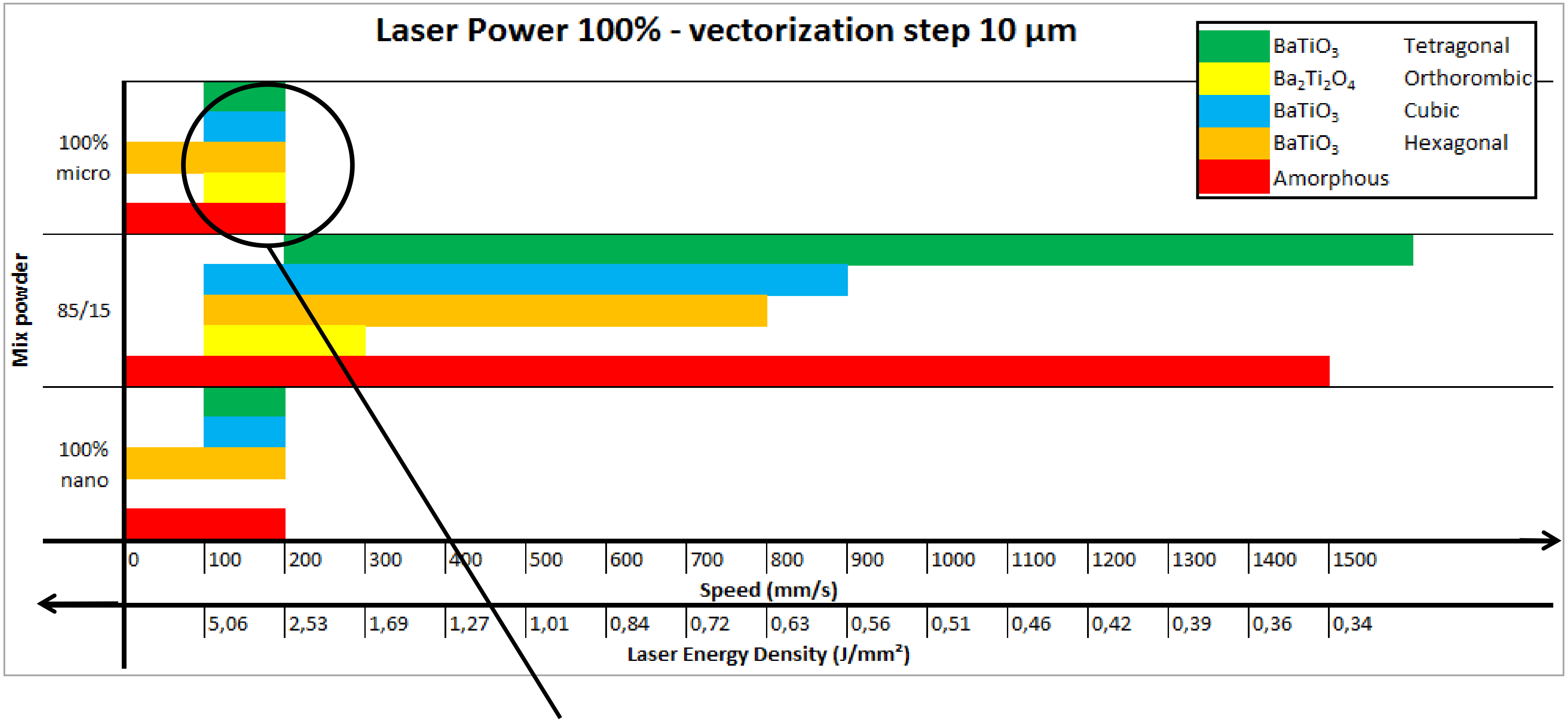


No cohesion of compacts for sp > 200 mm/s

→ Weak melting of the grains = low amount of liquid

II.2 Treatment of $BaTiO_3$ compacts

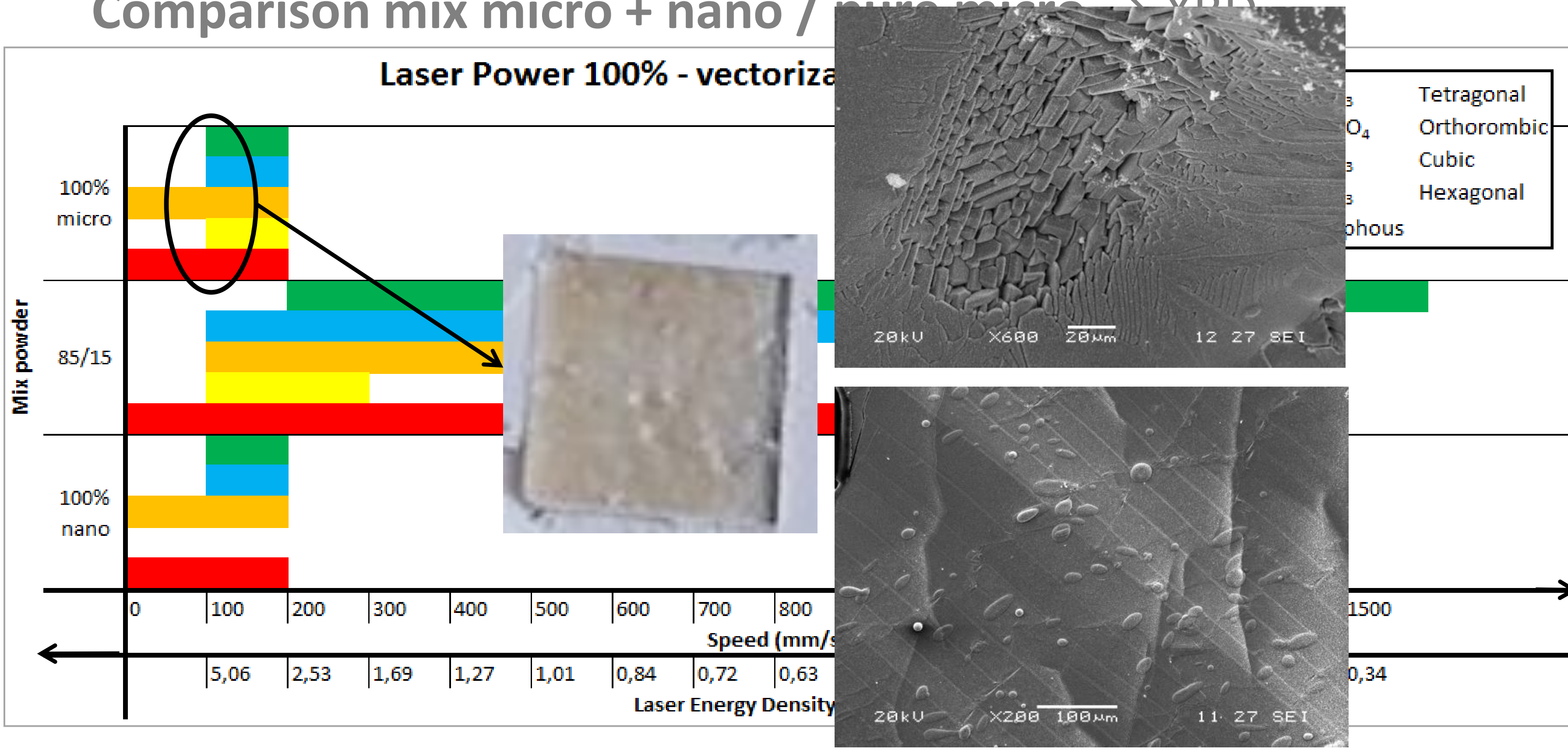
Comparison mix micro + nano / pure micro → XRD



For sp < 200 mm/s → melting of a part of the grains
→ phase transformation of the unmelted grains

II.2 Treatment of $BaTiO_3$ compacts

Comparison mix micro + nano /

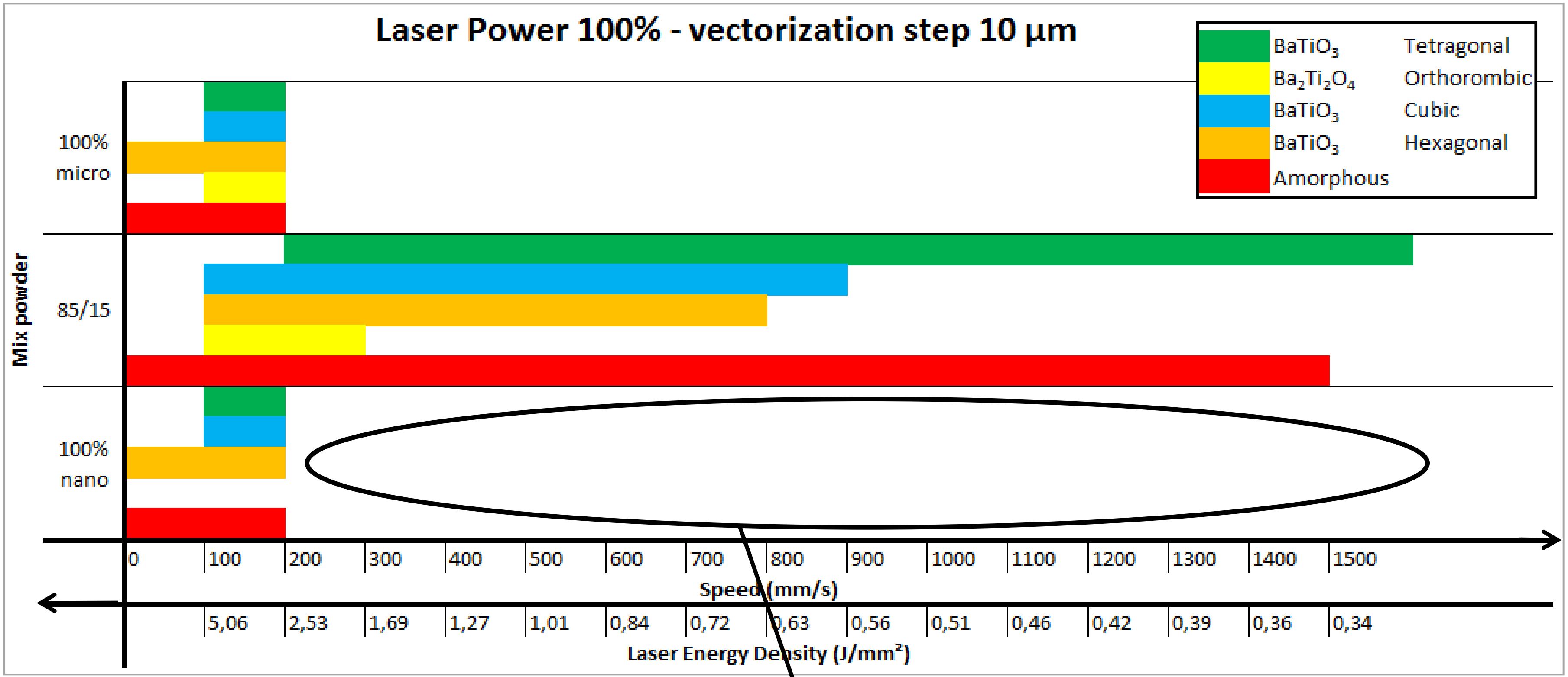


For sp < 200 mm/s → Coating extremely melted

→ In some areas large grains growth

II.2 Treatment of $BaTiO_3$ compacts

Comparison mix micro + nano / pure nano → XRD



No cohesion of compacts for sp > 200 mm/s

→ efficiency of the laser radiation absorption ?

→ grains agglomerates ?

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Experimental procedure

Coating may be assimilated to a 2D specimen (targeted thicknesses = 10µm)

Shrinkage limited to thickness

Heat transfers to substrate

Dilatometric mismatch vs substrate

Transposition of scan conditions obtained on compacts ?

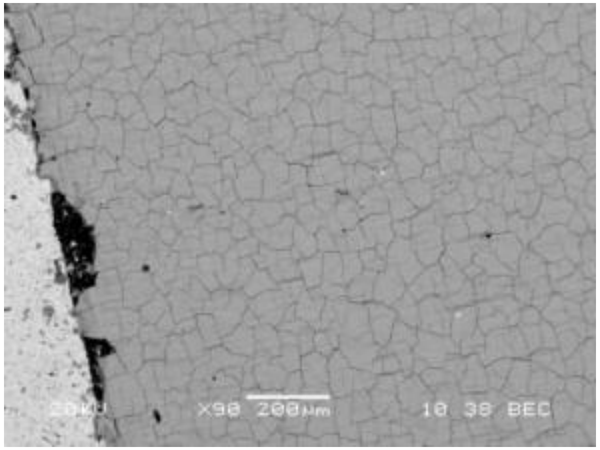
Preparation of the coating:

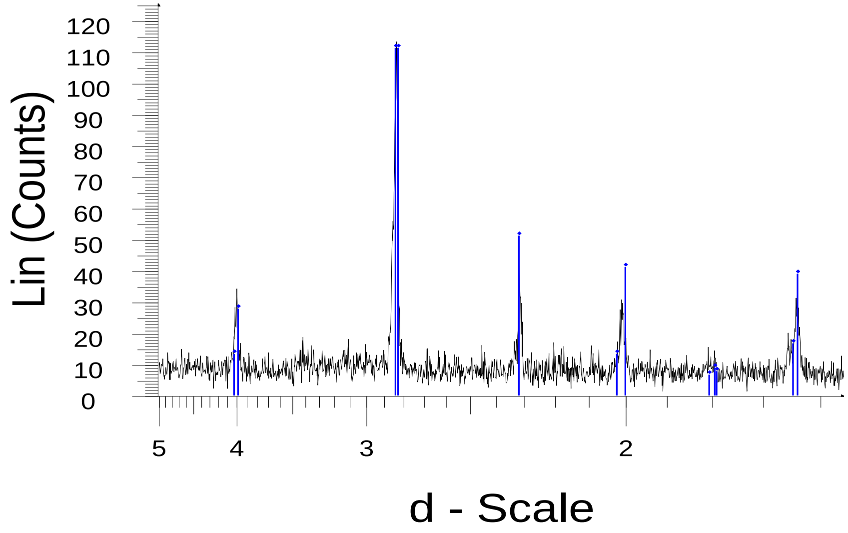
- Dispersion of the powder in water with addition of PAA
- Ball milling
- Suspension spraying with an airbrush system on a alumina substrate
- Laser scan

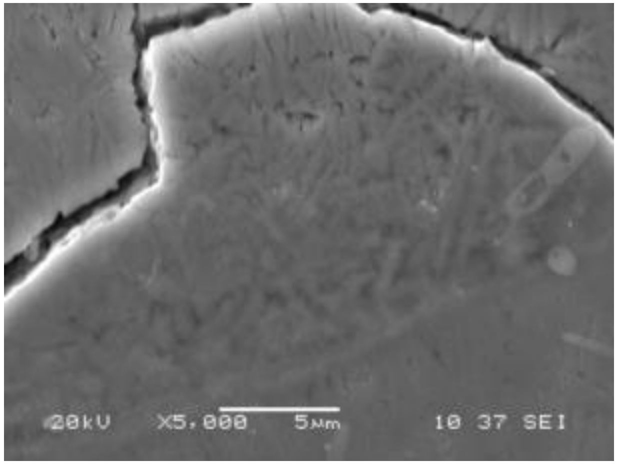
II.1 $BaTiO_3$ coating

First results

Scan laser condition : P100sp200vect20







- → Dense tetragonal $BaTiO_3$ coating with growth of initial grain powder
- → Good adhesion to substrate
- → Coating cracked (differential shrinkage + rapid cooling)

Presentation plan

Surface treatment by Laser/ research context

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Conclusions et perspectives

Conclusions:

Laser scanning of powder compacts show difficulties to obtain an homogeneous surface

In all scan conditions a partial melting occurs during treatment.

The amount of liquid produced is function of :

- The energy density J
- The granulometric distribution

Best results are obtained at high scan speed (> 1000 mm/s) on a powder mixture nano + micro :

- Low ratio liquid/solid (mainly melting of nano grains)
- Growth of the unmelted grains in the liquid phase
- No phase transformation of the tetragonal $BaTiO_3$
- Low amount of residual amorphous phase

Conclusions et perspectives

Conclusions:

First tests of laser scanning of powder coatings have been performed

A good adhesion to substrate is observed after laser treatment

A dense 20 µm tetragonal $BaTiO_3$ coating has been obtained but :

- Periodic cracking due to differential shrinkage
- Large amount of residual vitreous phase

Outlooks:

<u>Compacts :</u>

- Optimization of micro / nano ratio
- Further investigation of the sintering mechanisms

<u>Coatings :</u>

- Optimization of powder deposition (higher compacity)
- Further investigation of the scan condition

Thank you for your attention

Questions?