Advances and problems in the modeling of powder materials sintering with direct heating by electric current

Alexander Laptev [1,2]

Kim Vanmeensel [2]

1) Donbass State Engineering Academy, Kramatorsk, Ukraine

2) Katholieke Universiteit Leuven, Leuven, Belgium

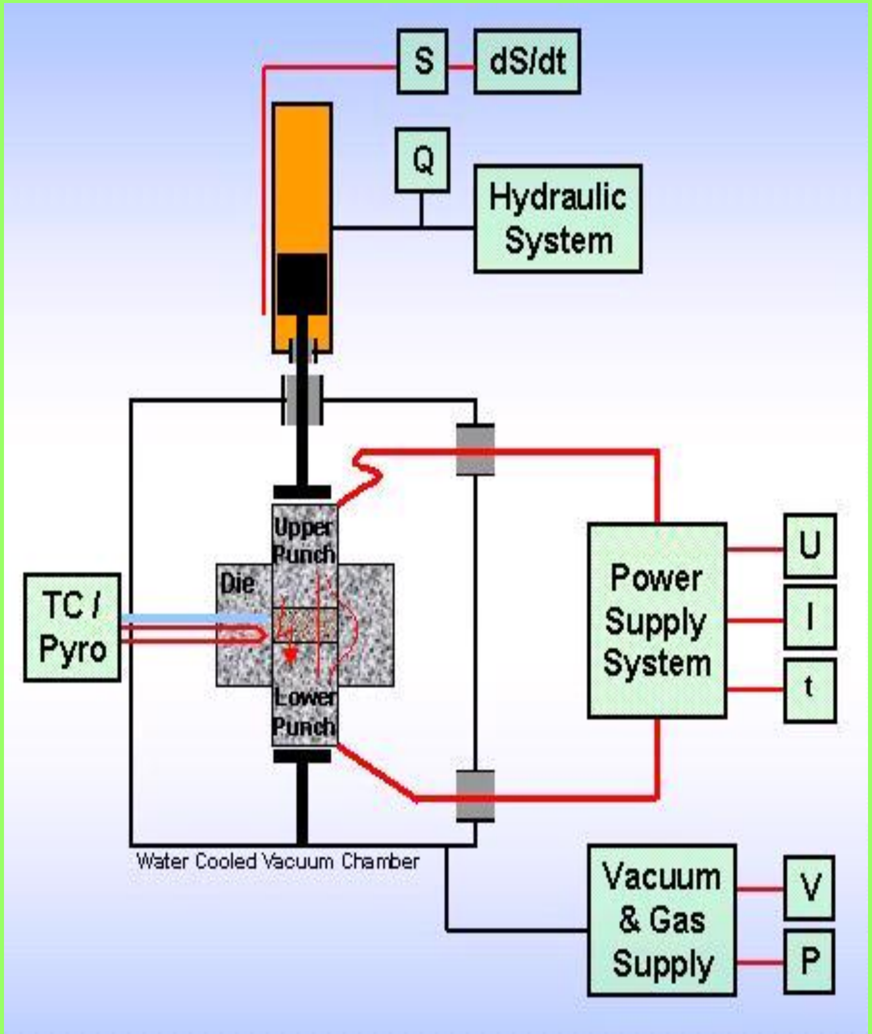
Outline

- Introduction. Where this work was done?
- Why modeling is needed?
- Mathematical model of process.
- Input data.
- Contact resistances.
- Modeling examples. Conclusions.
- Outlook.

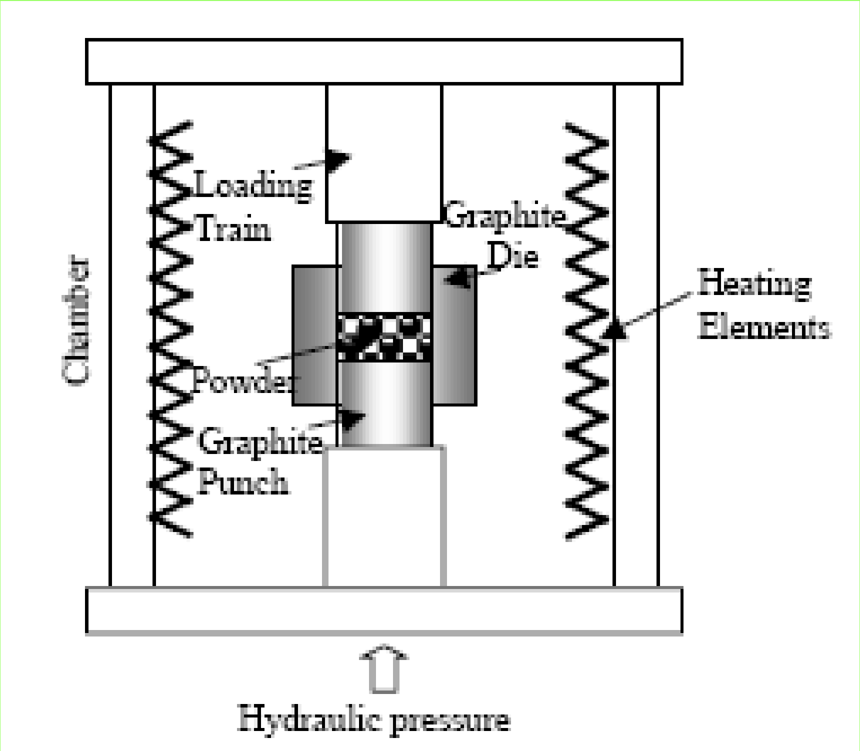
What is meant by sintering with direct heating by electric current ?

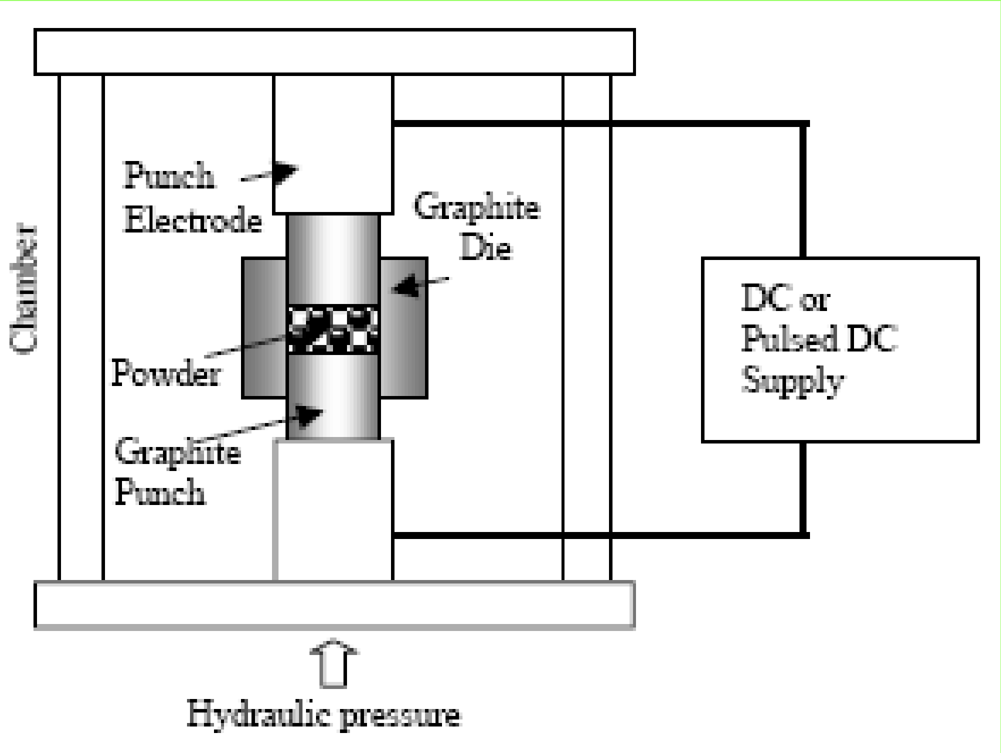
Technology of sintering with direct heating by electric current

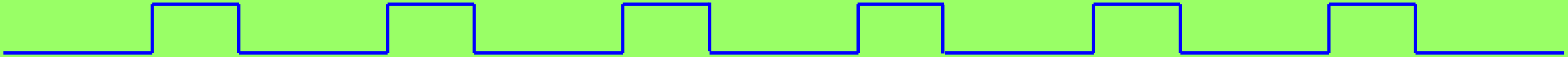
- **Process of rapid consolidation of metallic, ceramic powders and composites into fully dense material – SPS, FAST, PECS**
- **Powder is loaded into a die, made from a conductive material (graphite, steel, ceramic), and consolidated by a combination of mechanical pressure and temperature produced by direct electrical (Joule) heating with the rates up to 1000°C/min.**



SPS vs Hot Pressing

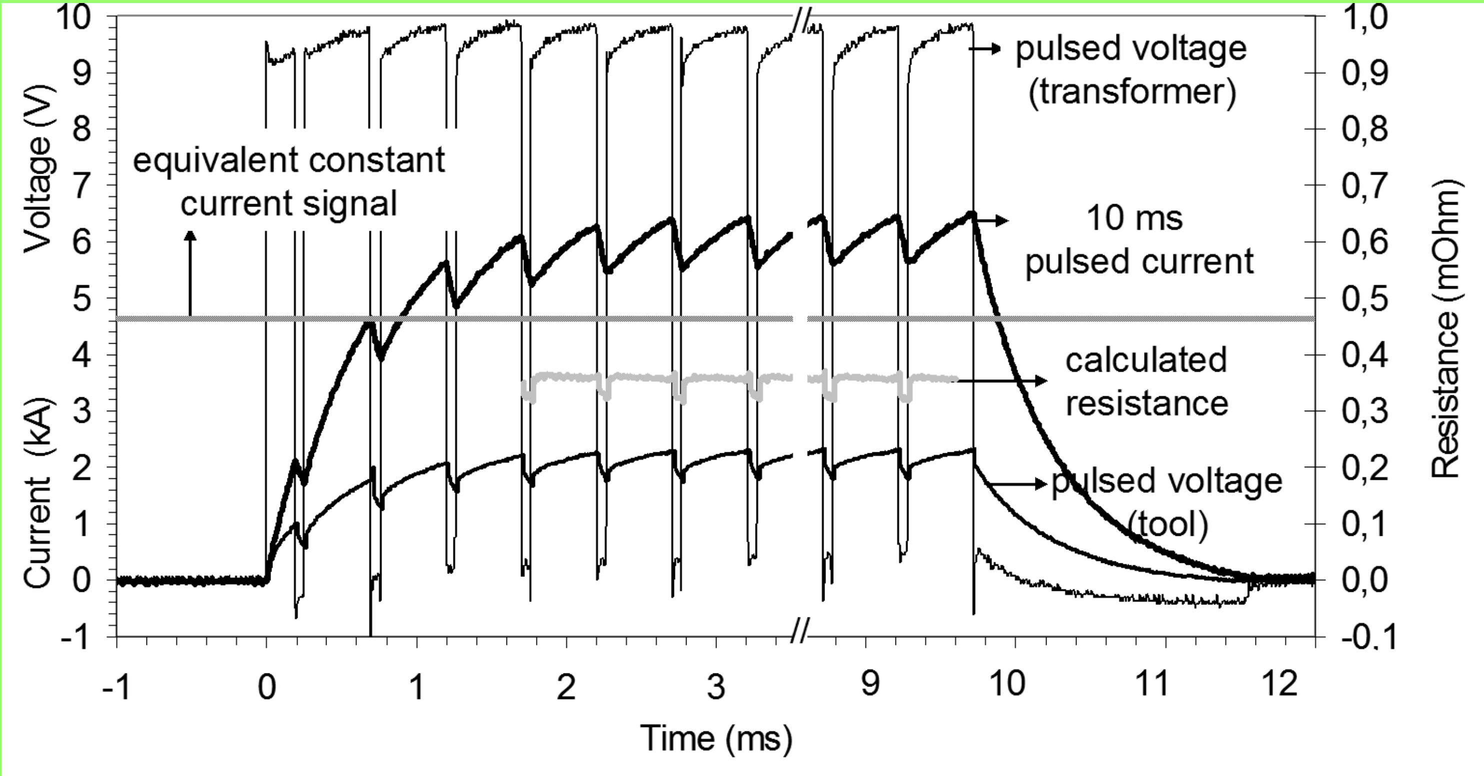






Direct pulsed current

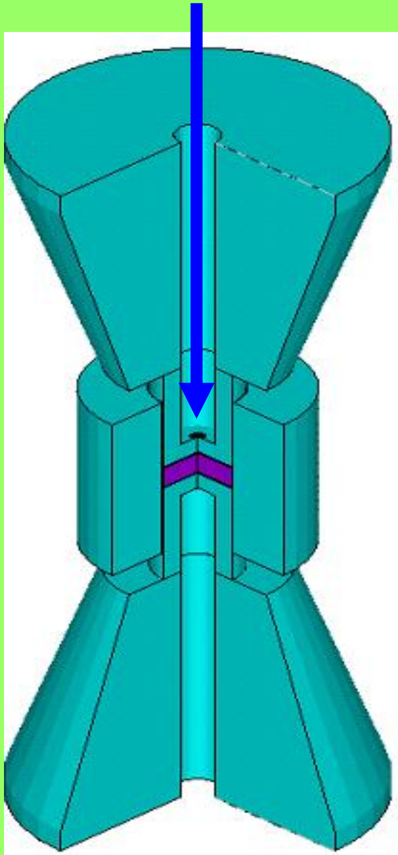
Pulse structure at Spark Plasma Sintering



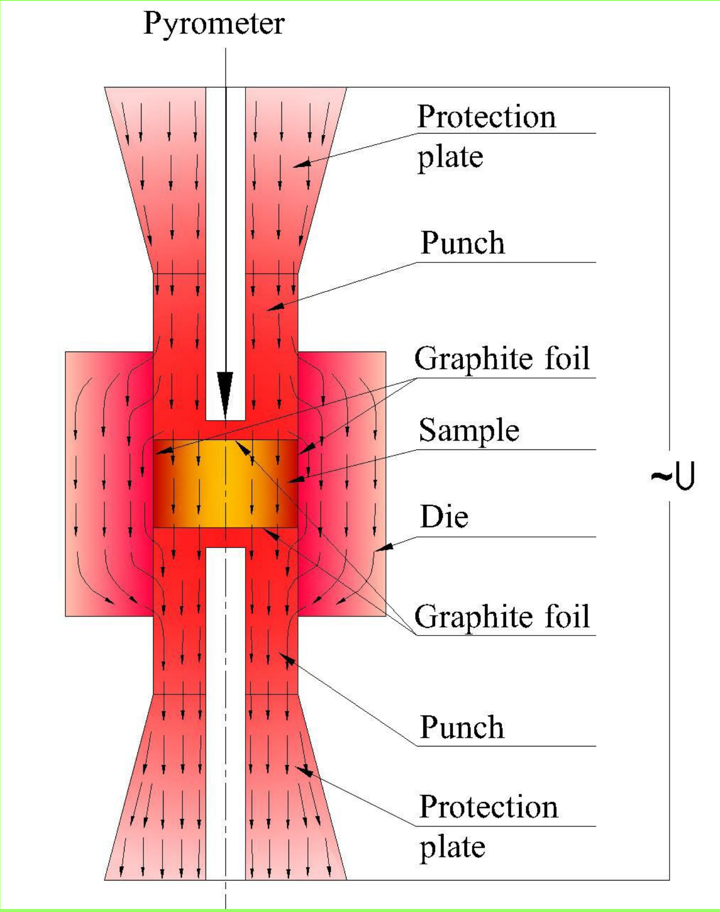
- Pulsed DC current
- Pulse time: 0-255 ms
- Pause time: 0-255 ms

Tool design and current paths during SPS

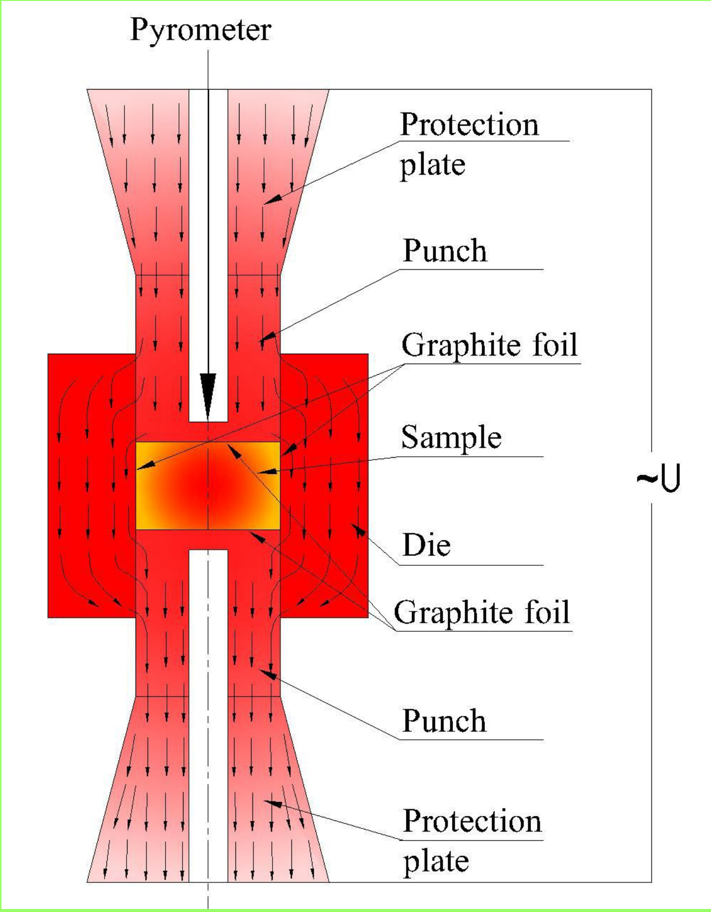
Pyrometer



Graphite tool



Conductive powder



Non conductive powder

Advantages of spark plasma sintering technique:

a) very quick consolidation comparing with traditional hot pressing methods (HIP, HP)

~ additional diffusion mechanisms

b) possibility to minimize grain growth

~ strength

c) Short processing time (high heating rates, short dwell, fast cooling).

~ productivity

☺ Consolidation of nanomaterials with improved properties within much shorter time periods

Problems using spark plasma sintering technique:

a) temperature control within samples

b) possible inhomogeneous temperature distribution (size and shape limitation)

c) possible inhomogeneous distribution of density and other properties

Where this work was done?

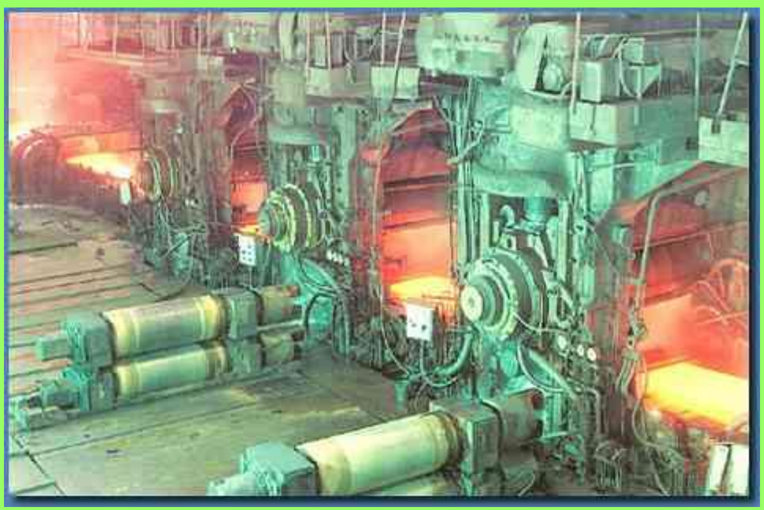
1. Donbass State Engineering Academy, Donbass, Kramatorsk, Ukraine
   Department of Materials Science, Department Materials Working by Pressure

2. Katholieke Universiteit Leuven, Leuven, Flanders, Belgium
   Department of Metallurgy and Materials Engineering


KU Leuven
DSEA
500 Miles
500 Km
Reykjavik
ICELAND
Norwegian Sea
Arctic Circle
Barents Sea
North Atlantic Ocean
Torshavn
NORWAY
SWEDEN
FINLAND
Baltic Sea
Oslo
Stockholm
Helsinki
Tallinn
ESTONIA
St. Petersburg
North Sea
U. K.
Glasgow
Edinburgh
IRELAND
Belfast
Dublin
Limerick
Cork
Waterford
Liverpool
Manchester
Leeds
Sunderland
Leicester
Birmingham
London
DENMARK
Goteborg
Copenhagen
Riga
LATVIA
LITHUANIA
Vilnius
Minsk
BELARUS
Moscow
Jaroslavl
Rostov
Gorkiy
Kazan
Volga
Tol Yatti
Kuybyshev
Saratov
Volgograd
Sverdlovsk
Perm
Chelyabinsk
Izevsk
Ufa
The Hague
Amsterdam
NETHERLANDS
Rotterdam
Hamburg
Gdansk
Berlin
POLAND
Warsaw
Poznan
Lodz
Lubin
Wroclaw Breslau
Krakow
GERMANY
Leipzig
Bonn
Frankfurt
BELGIUM
Luxembourg
LUX.
Paris
Nantes
Karlsruhe
Strasbourg
Nuernberg
Stuttgart
Muenchen
Prague
CZECH
Ostrava
Brno
SLOVAKIA
Vienna
Bratislava
Zurich
Bern
SWITZERLAND
AUSTRIA
Geneva
FRANCE
Bordeaux
Lyon
Bayonne
Toulouse
Monaco
Marseille
Milano
Turin
Genova
Bologna
Venezia
Firenze
Ljubljana
SLOVENIA
CROATIA
Zagreb
BOSNIA and HERZ.
Sarajevo
YUGOSLAVIA
Belgrade
HUNGARY
Budapest
Cluj
Timisoara
ROMANIA
Bucharest
Constanta
MOLDOVA
Chisinau
Odessa
Kiev
Lviv
UKRAINE
Kharkov
Donets
Zaporozhye
Krivoy Rog
Frunze
Dnieper
Krasnodar
RUSSIA
Caspian Sea
Black Sea
GEORGIA
Tbilisi
Baku
AZERBAIJAN
ARMENIA
Yerevan
Danube
Varna
Burgas
Sofia
BULGARIA
Skopje
MACEDONIA
Tirane
ALBANIA
Thessaloniki
GREECE
Agrinion
Athens
Istanbul
Bursa
Ankara
TURKEY
Izmir
Adana
Halab
Al Mawsil
Tabriz
SYRIA
Euphrates
Baghdad
IRAQ
Basra
Damascus
Beirut
LEBANON
CYPRUS
NORTH CYPRUS
Porto
PORTUGAL
Lisbon
Valladolid
SPAIN
Madrid
Valencia
Cordoba
Sevilla
Barcelona
Gibraltar
Rabat
Casablanca
MOROCCO
Algiers
Tunis
Rome
ITALY
Napoli
Palermo
Valletta
MALTA
Mediterranean Sea
Ob'

Kramatorsk on the map of Ukraine






НКМЗ

Donbass State Engineering Academy

Kramatorsk – city in the south-east of Ukraine with population of 200 thousand people. Center of heavy machine building and ferrous metallurgy.

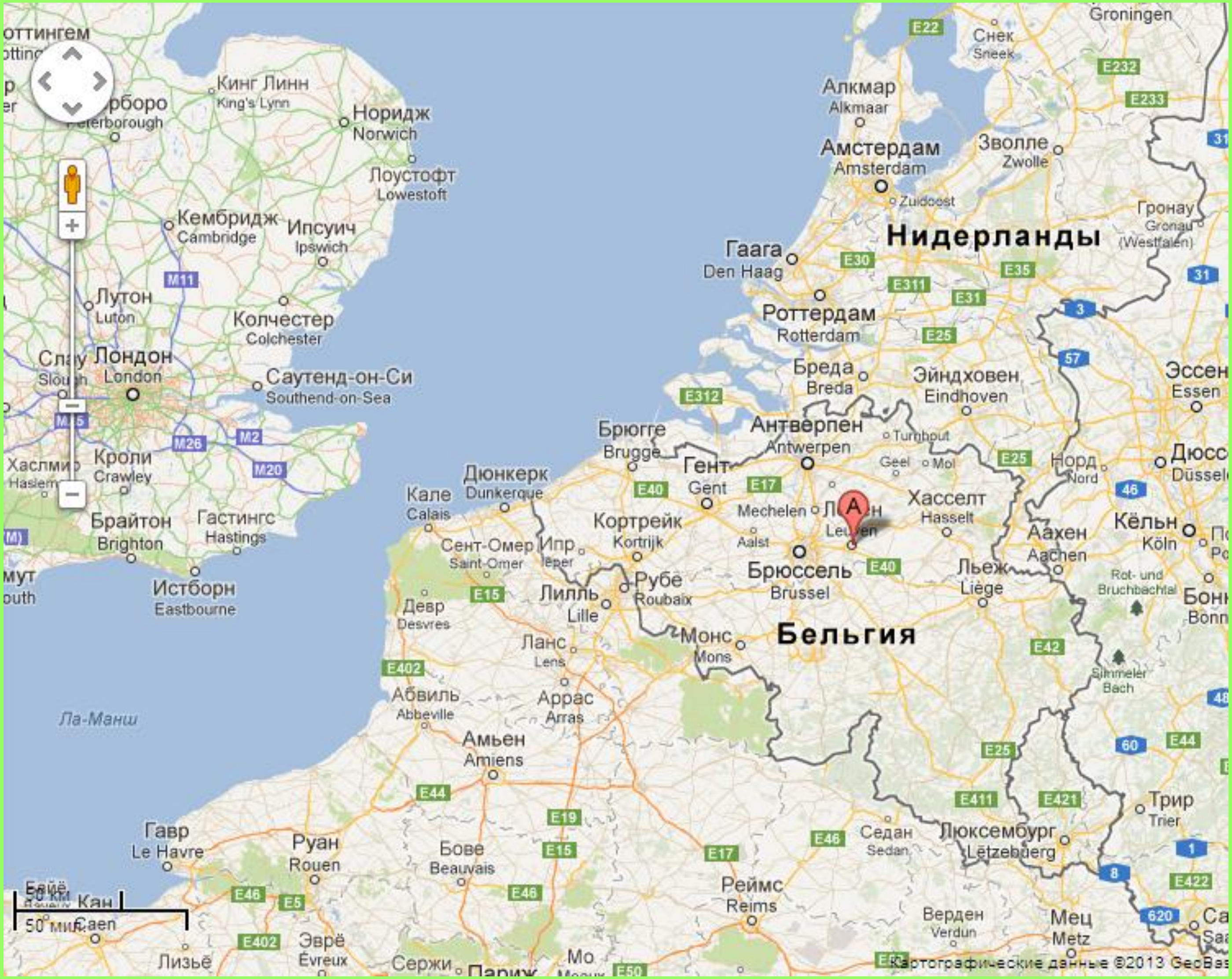
Donbass State Engineering Academy – High Education Institution of four level of accreditation.

5390 full-time students. 5070 external students.

446 teachers, 250 associate professors, 19 full professors.

4 faculties, 40 departments. Education in mechanical engineering, metallurgical engineering, electrical engineering, IT, economics, management.

Leuven on the map of Belgium



Katholieke Universiteit Leuven

Leuven – city in the Flemish part of Belgium, located at 26 km on the east from Brussels. One of the university centers of Belgium. Main office of AB InBev (Stella Artois, Leffe, Hoegaarden)

Katholieke Universiteit Leuven – the oldest and the most famous university in Belgium. Founded in 1425 by Pope Martin V.

40.000 students, 3 groups of faculties: humanitarian sciences; biomedicine; science, engineering and technology. Place 58 in the Universities Word Ranking 2012/2013. Place 38 in the group of engineering and technology (Times, Thomson Reuters).

Department of Metallurgy and Materials Engineering. Group of Advanced Ceramics and Powder Metallurgy. Professor Jef Vleugels, Professor Omer Van der Biest, PhD Kim Vanmeensel.

Faculty of engineering sciences

SPS (FAST) installation in KU Leuven

HP D 25/1 FCT Systeme (Rauenstein, Германия)

Temperature: 20-2200 °C

Load: 5-250 кН

Heating rate:

5-400 °C /min

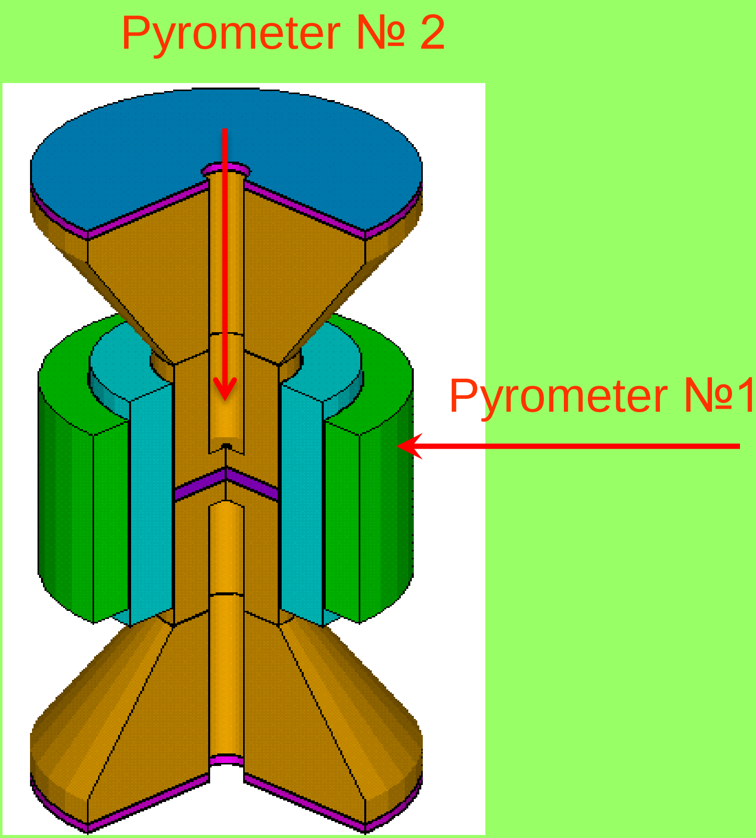
Power: 8 kW

Pulse: 0-255 мс

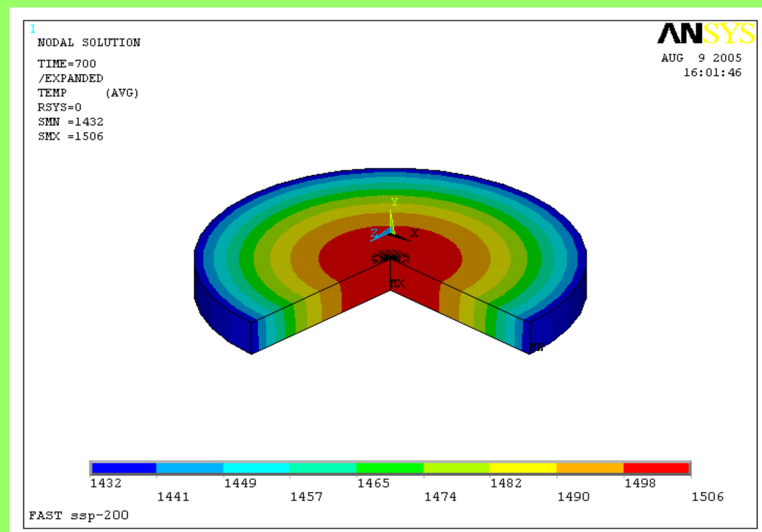
Diameter of compact (max): 80 мм

Atmosphere: vacuum, Ar, N_2, He

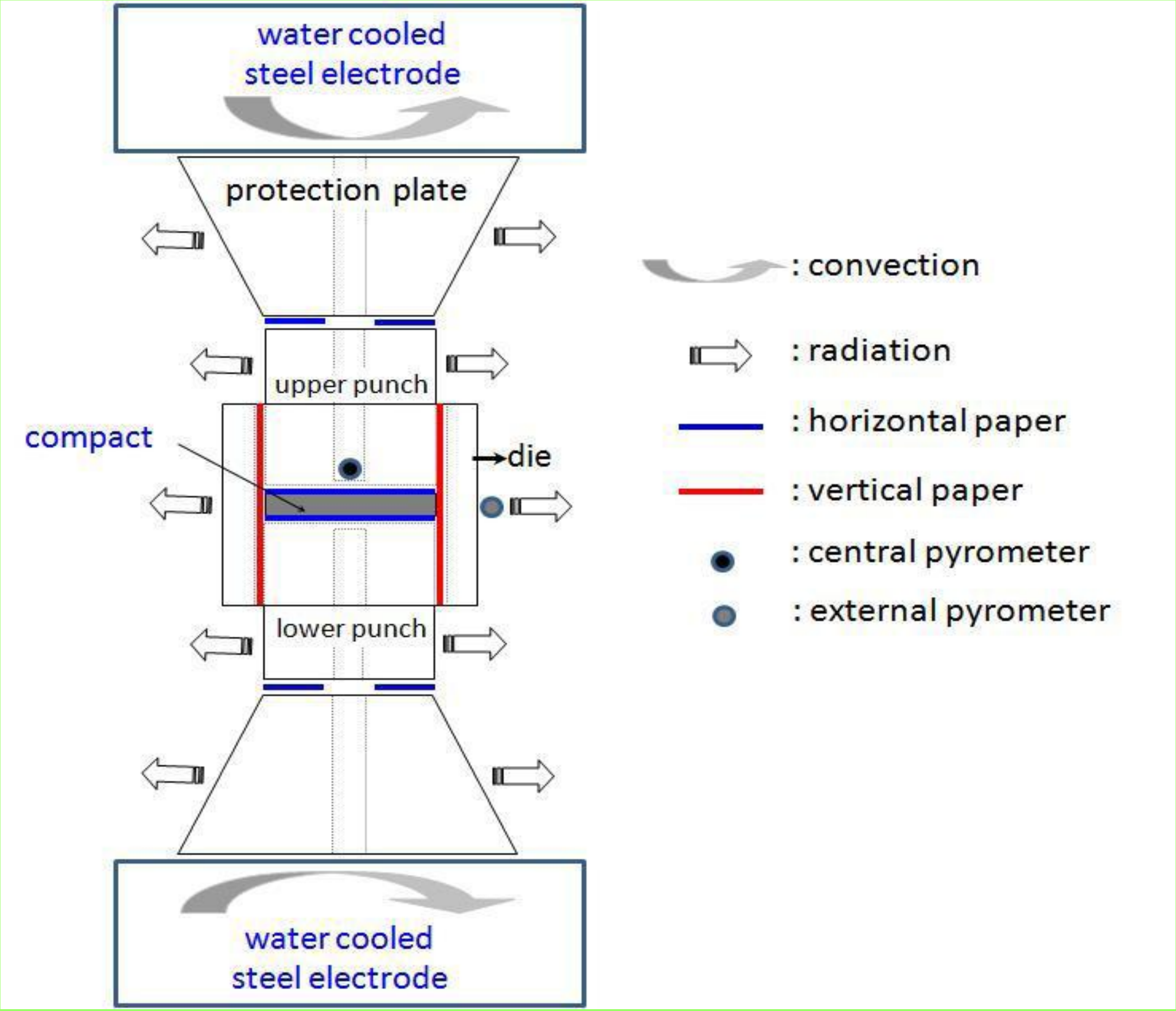
Why modeling is needed?



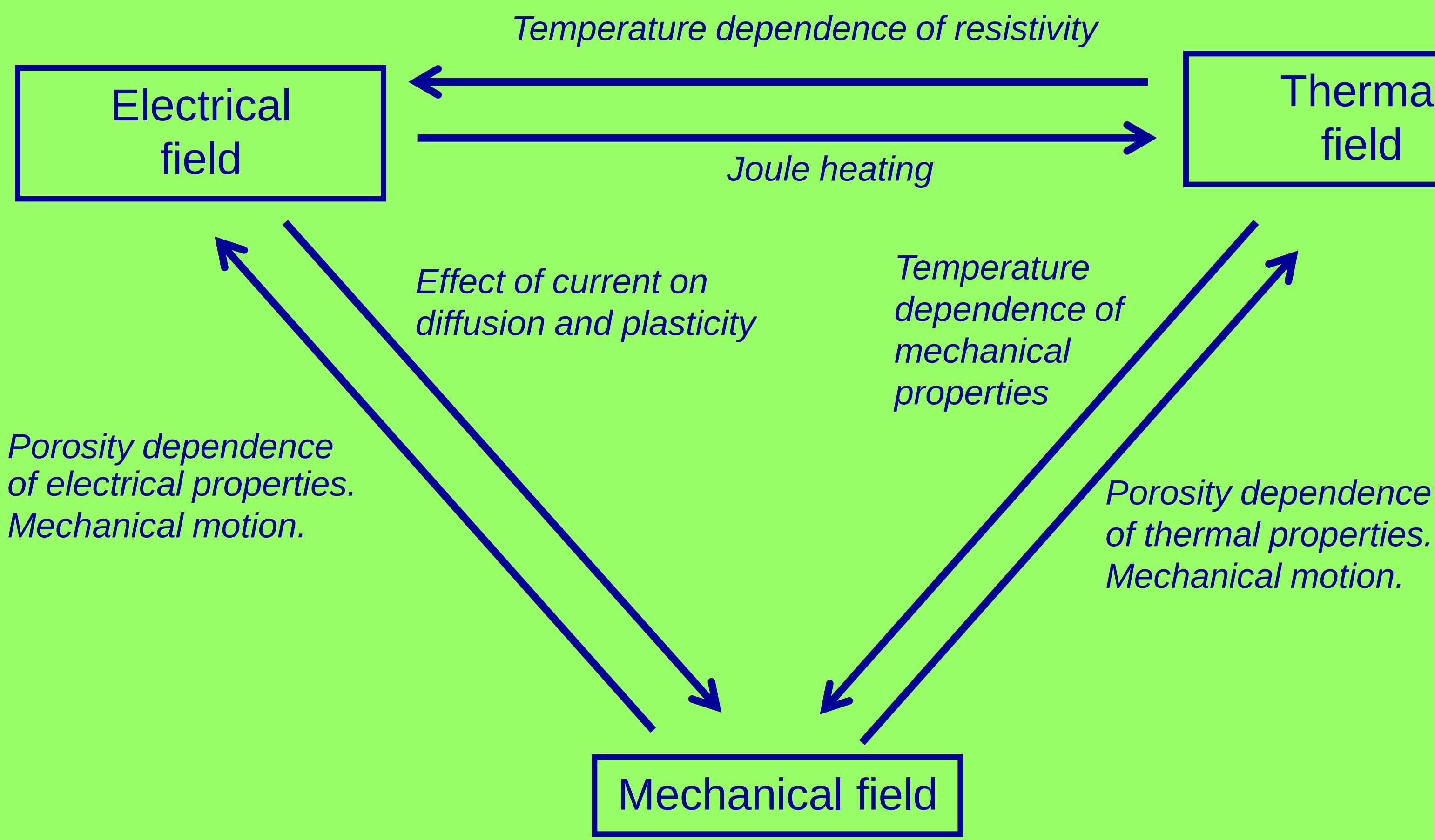
- Temperature control (especially at high heating rates)
- Temperature distribution (especially for large parts and in the case of narrow temperature window)
- Properties distribution (particularly density)
- Shape distortion (during plastic deformation)



Schematic view of tool



Physical fields, interacting during sintering with direct electrical heating



Mathematical model of process

Electrical field

Ohm's law

$$\vec{j} = \sigma(\rho, T) \cdot \vec{E}$$

Joule's law

$$w_j = \vec{j} \cdot \vec{E} = \sigma(\rho, T) \cdot \vec{E}^2$$

Thermal field

Fourier's law

$$\gamma \cdot c(\rho, T) \cdot \frac{\partial T}{\partial t} = \lambda(\rho, T) \cdot \nabla^2 T + w_j$$

Stefan–Boltzmann law

$$w_r = \sigma_S \cdot \varepsilon(T) \cdot (T_e^4 - T_a^4)$$

Mechanical field

Hook's law

$$T_\varepsilon^e = \frac{1}{3K(\rho, T)} \cdot T_{\sigma_0} + \frac{1}{2G(\rho, T)} \cdot D_\sigma$$

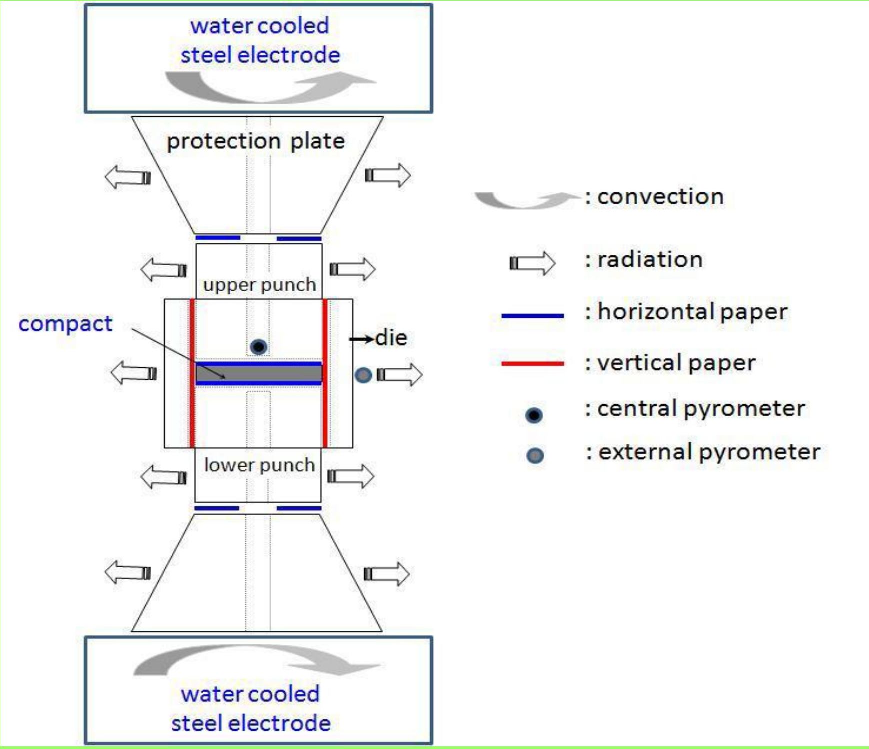
Constitutive equations

$$T_{\dot{\varepsilon}} = F(T_\sigma, \rho, T, H)$$

$$\dot{\varepsilon}_{eff} = A_0 \frac{\sigma_{eff}^n}{d^p} \exp\left(-\frac{Q}{RT}\right)$$

Mathematical model of process

Contact properties



Thermal conductivity

$$w_c = \lambda_c(T)\cdot(T_1 - T_2)$$

Electrical conductivity

$$\vec{j}_c = \sigma_c(T)\cdot(\vec{U}_1 - \vec{U}_2)$$

Heat generation

$$w_c = \sigma_c(T)\cdot\vec{U}^2$$

Convection

$$w_{conv} = h(T)\cdot(T_p - T_w)$$

Friction law (Coulomb)

$$f = \mu\cdot F_n$$

Input data

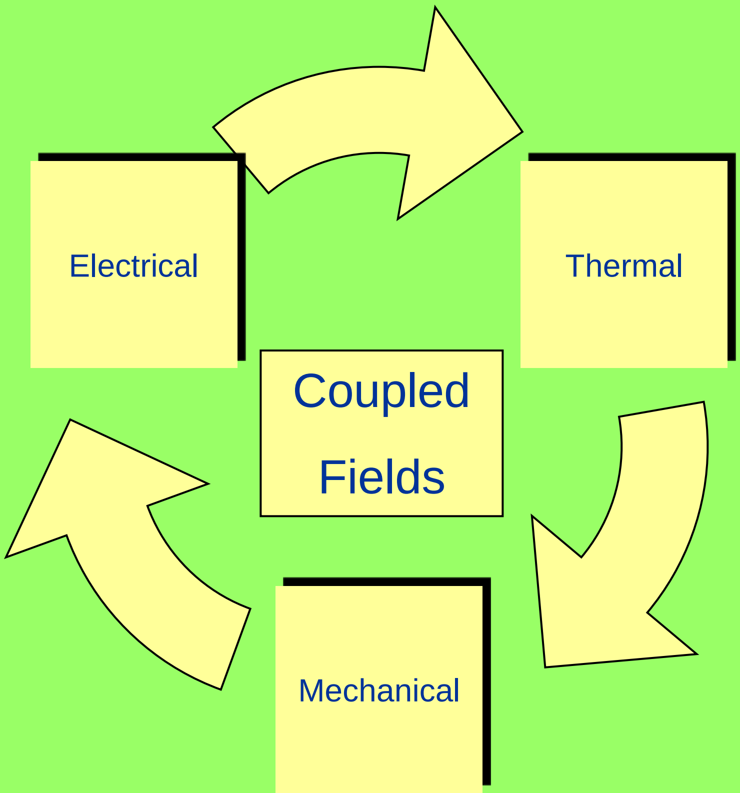
Properties of tool and sample materials in dependence on temperature and porosity

1. Electrical conductivity
2. Specific heat
3. Thermal conductivity
4. Thermal expansion coefficient
5. Mechanical properties (Young's modulus, Poisson coefficient etc.)

Contact properties (vertical and horizontal)

1. Specific electrical conductivity
2. Specific thermal conductivity
3. Radiation coefficient
4. Convection coefficient
5. Friction coefficient

FEM software used for modeling



☺

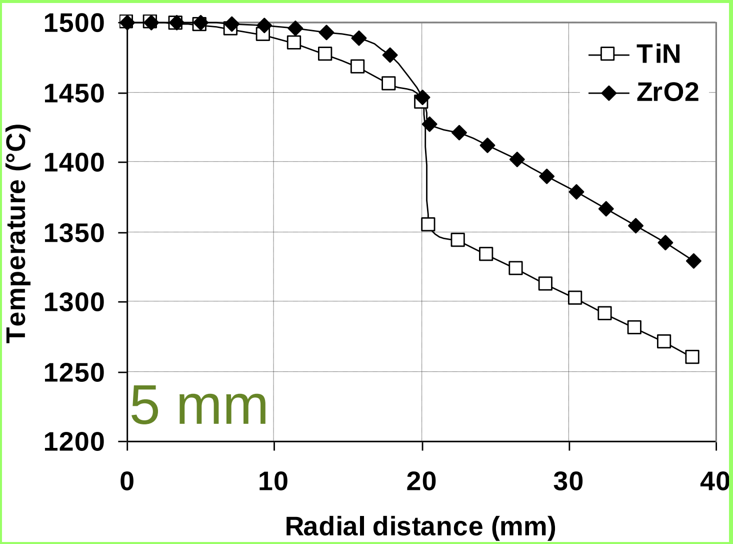
ANSYS и ABAQUS software provide coupling of electrical and thermal fields or thermal and mechanical fields separately ☹

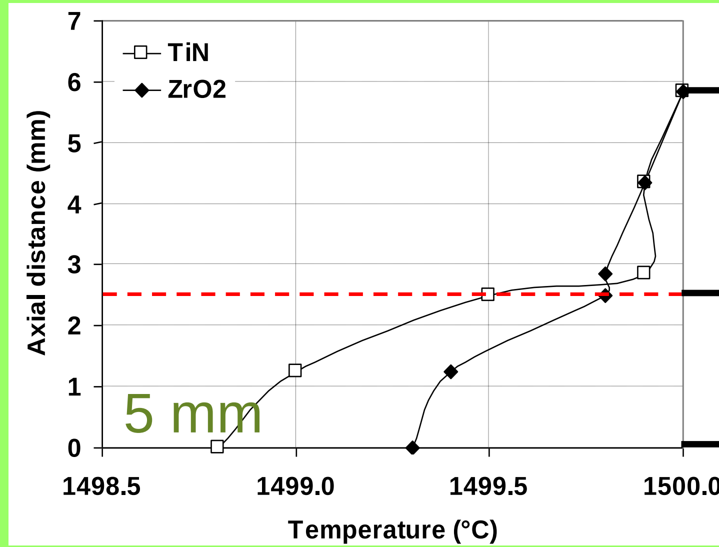
Modeling of temperature field

K. Vanmeensel, A. Laptev, J. Hennicke , J. Vleugels,
O. Van der Biest

Acta Materialia 53 (2005) 4379–4388

Temperature distribution in sample and in tool

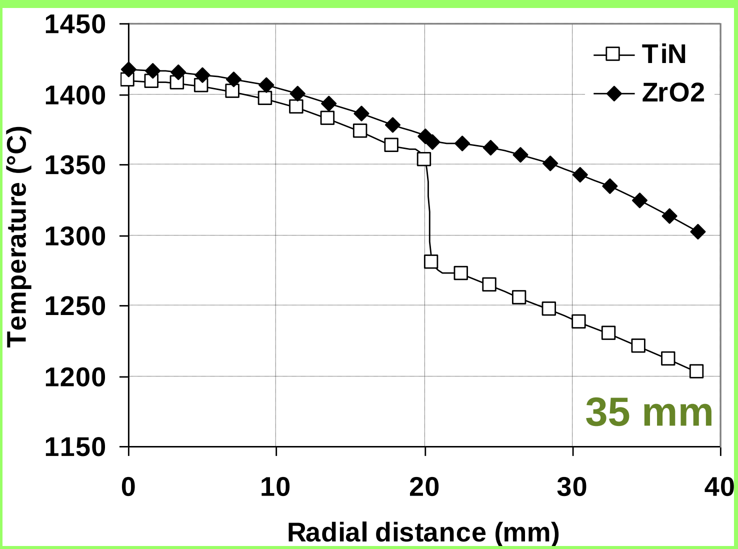


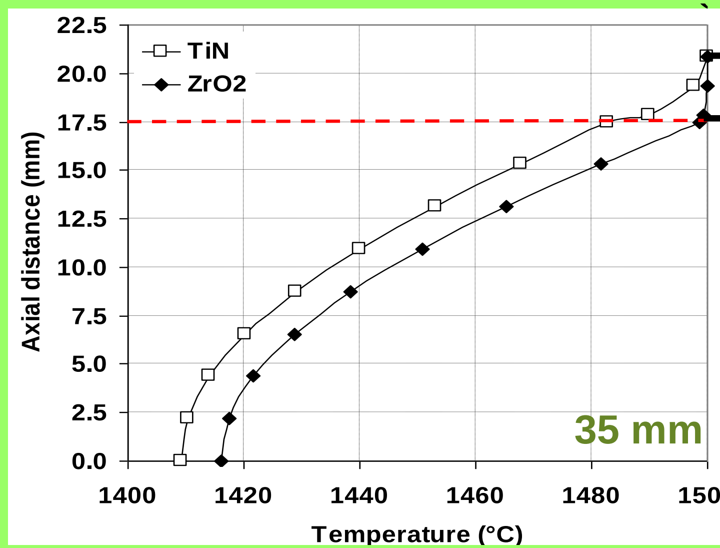


Pyrometer

Sample top

Sample centre



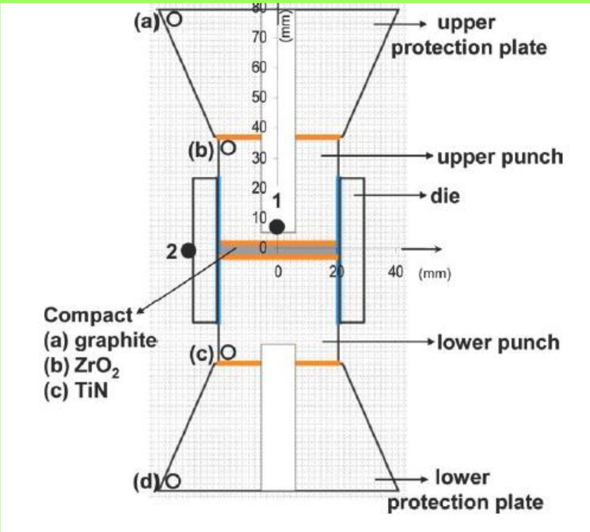


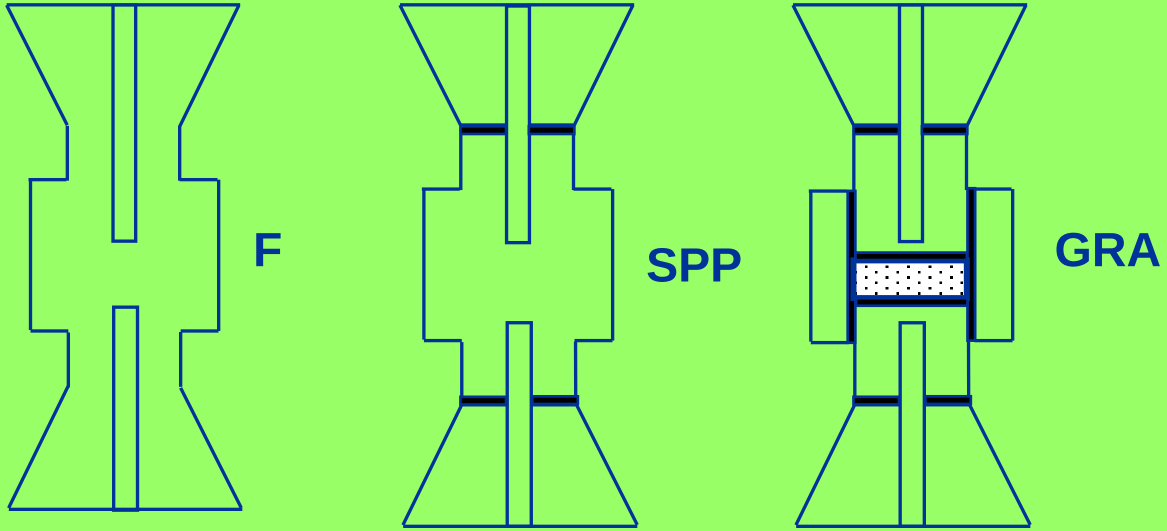
Pyrometer

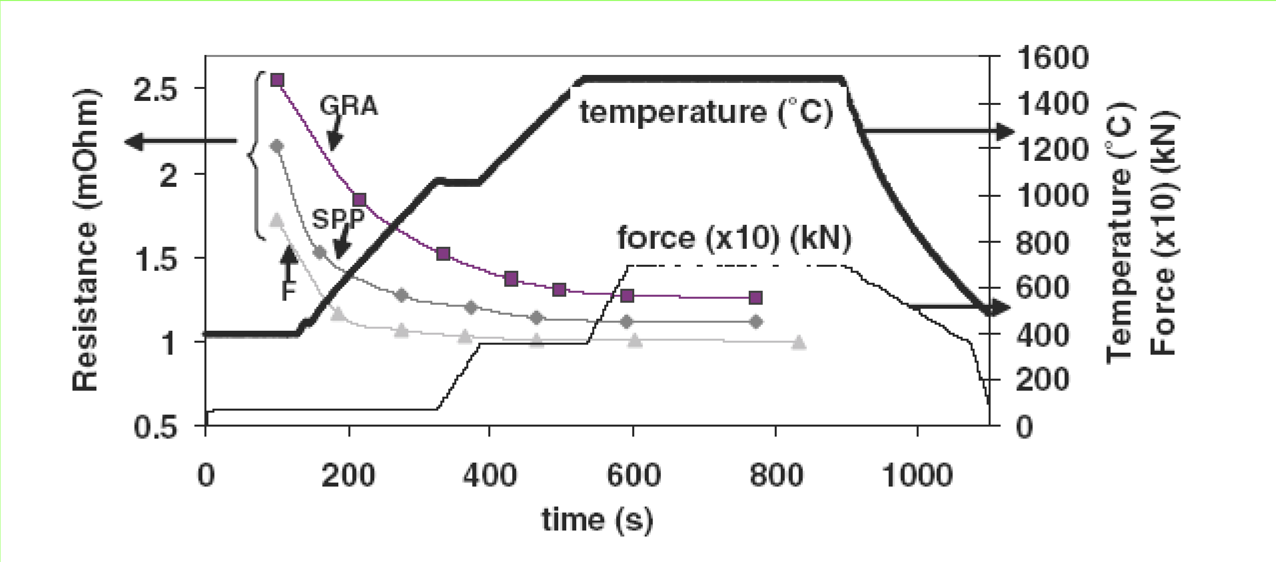
Sample top

Sample centre

Investigation of contact resistances





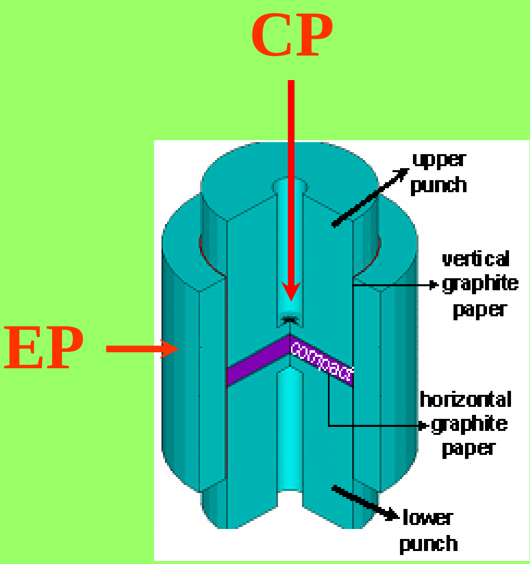


$$I_c = I_p \sqrt{\frac{2}{3}}.$$

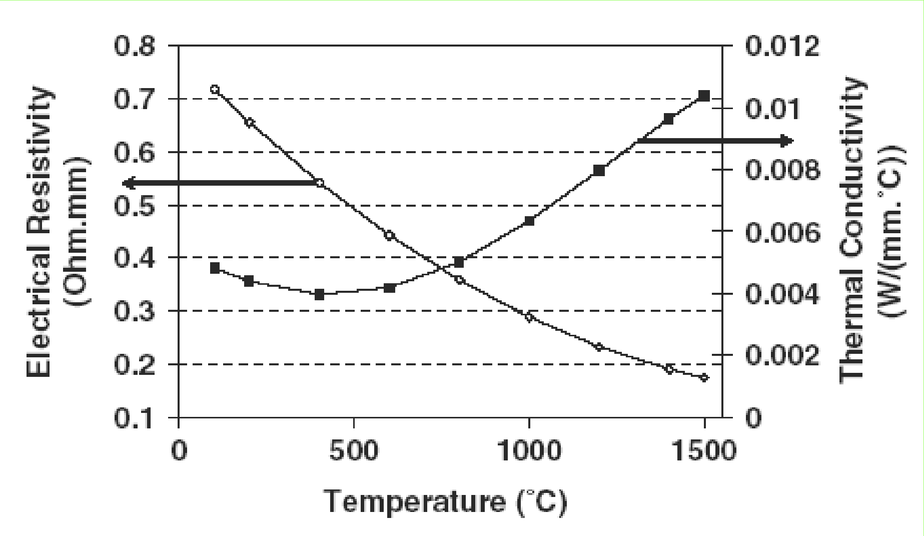
Investigation of contact resistances

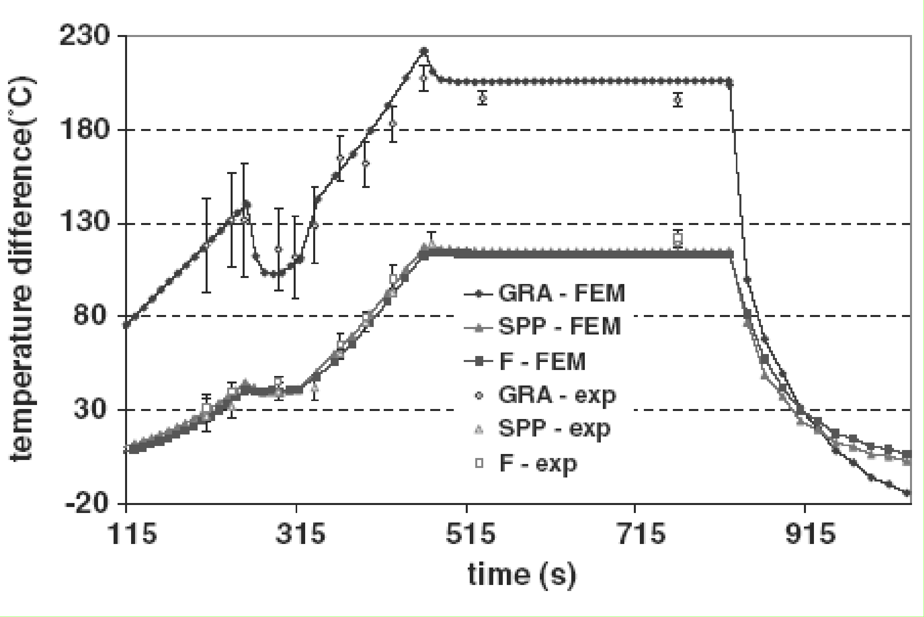
$$\lambda_p = \eta \cdot \lambda_{gr} \cdot \frac{\rho_{gr}}{\rho_p} \qquad \eta = 2.85$$

Resistance of vertical contacts approximately 7 times larger then horizontal contacts.



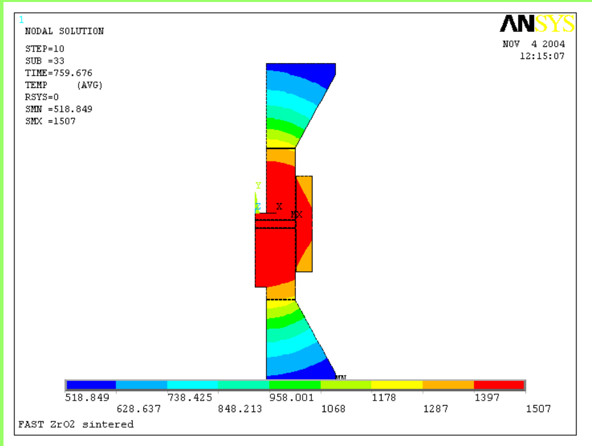
Vertical resistances have an essential influence on temperature distribution.

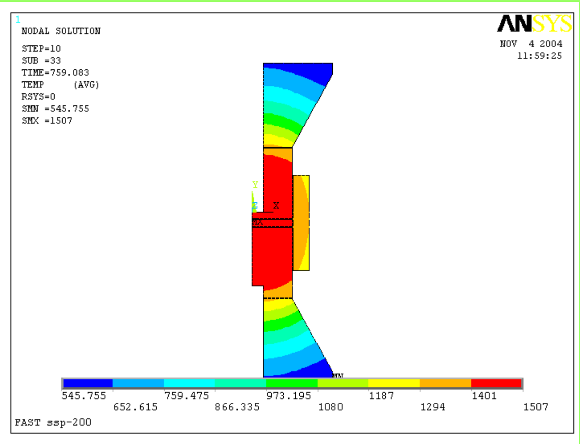


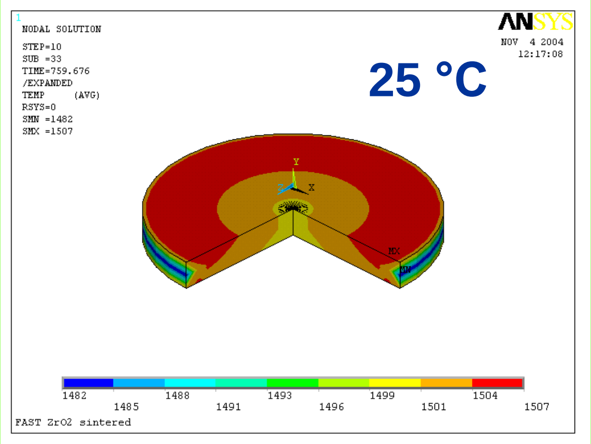


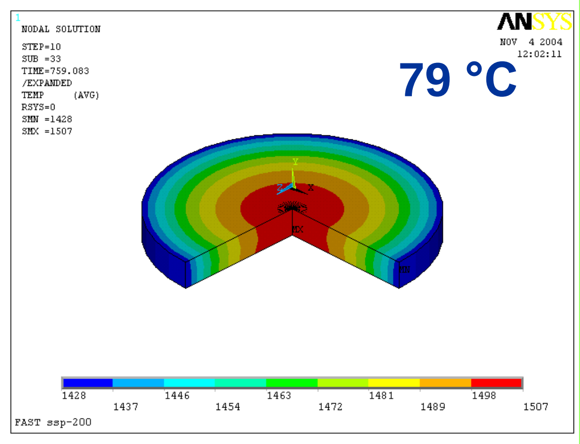
Holding at 1500°C

Influence of samples conductivity on temperature distribution at 1500°C





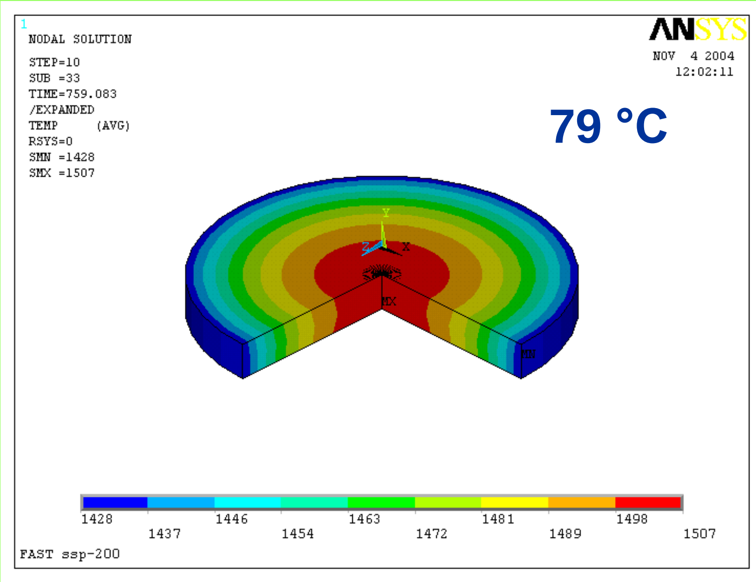


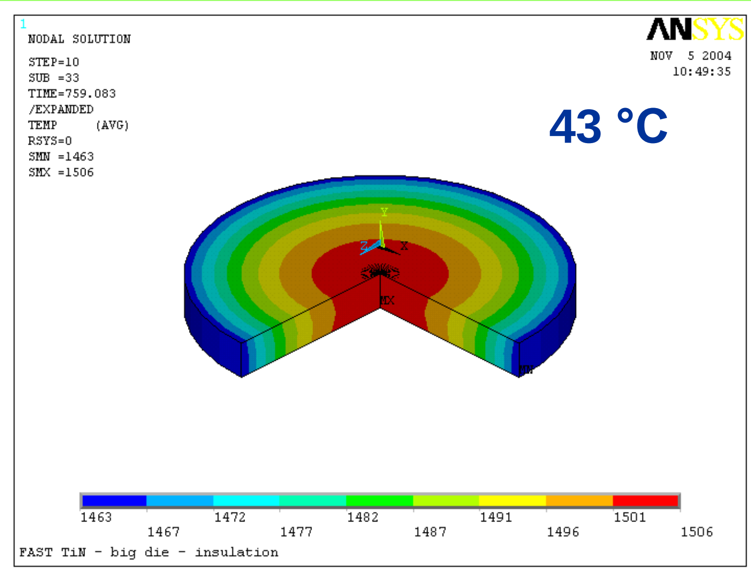


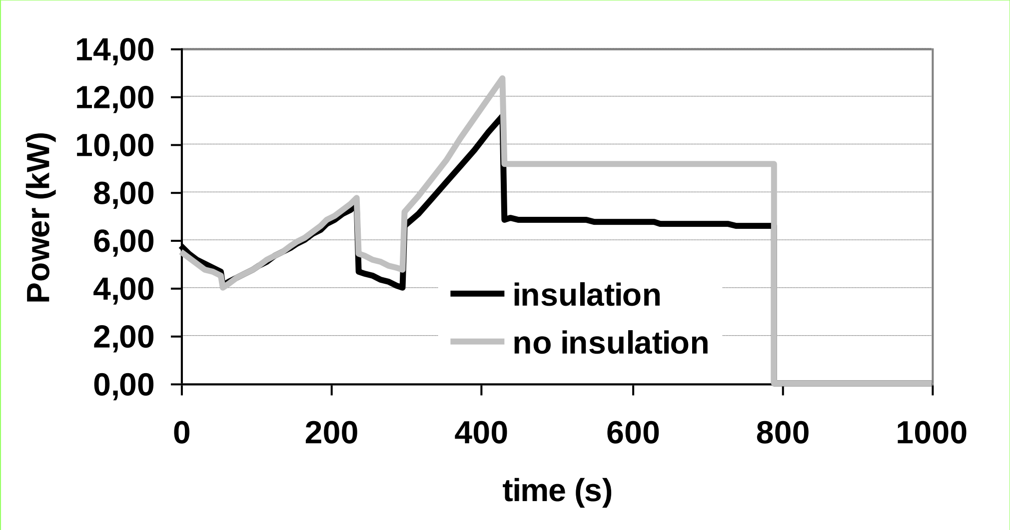
3Y-ZrO_2

TiN

Influence of die insulation at 1500°C







Main conclusions (section # 1)

- Temperature distribution during SPS within both tool and sample can be very inhomogeneous. Therefore a special attention must be paid to correct temperature control.
- Contact resistances have essential influence on temperature distribution in both tool and sample during SPS.
- Conductivity of sample material and die wall thickness determine character of temperature distribution within the sample.
- Application of thermal insulation results in essential decrease of temperature gradient and energy consumption during SPS.

Spark Plasma Sintering of ZrO_2-TiCN (60/40) composite

K. Vanmeensel, A. Laptev, O. Van der Biest, J. Vleugels

Journal of European Ceramics Society 27 (2007) 979–985

Powder composite ZrO_2-TiCN (60/40)

Y-ZrO_2

-High toughness (10 MPa.$m^{1/2}$)

-High strength (1.2 GPa)

-Moderate hardness (12 GPa)

-Electrically insulating

TiN

-Low toughness (3.5 MPa.$m^{1/2}$)

-Low strength (0.5 GPa)

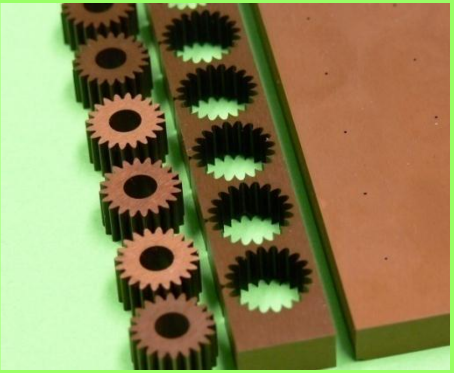
-High hardness (17 GPa)

-Electrically conductive

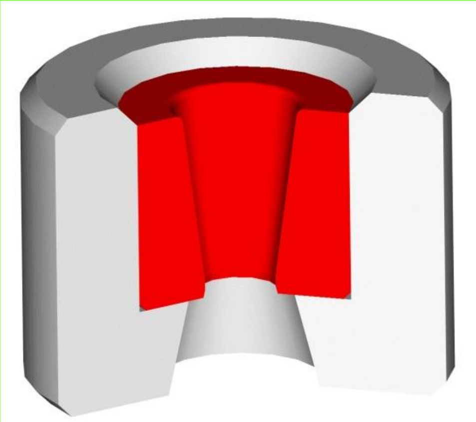
↓

ZrO_2-TiN (60/40)

ZrO_2-TiCN components manufactured by EDM



Gears

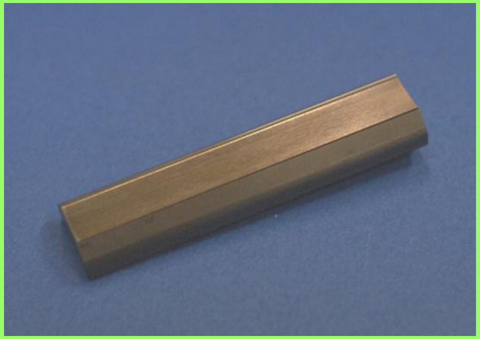


Extrusion die

Attritor disc

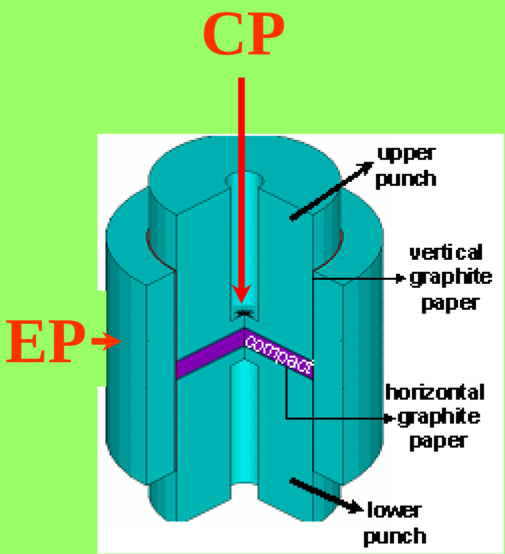
Gears

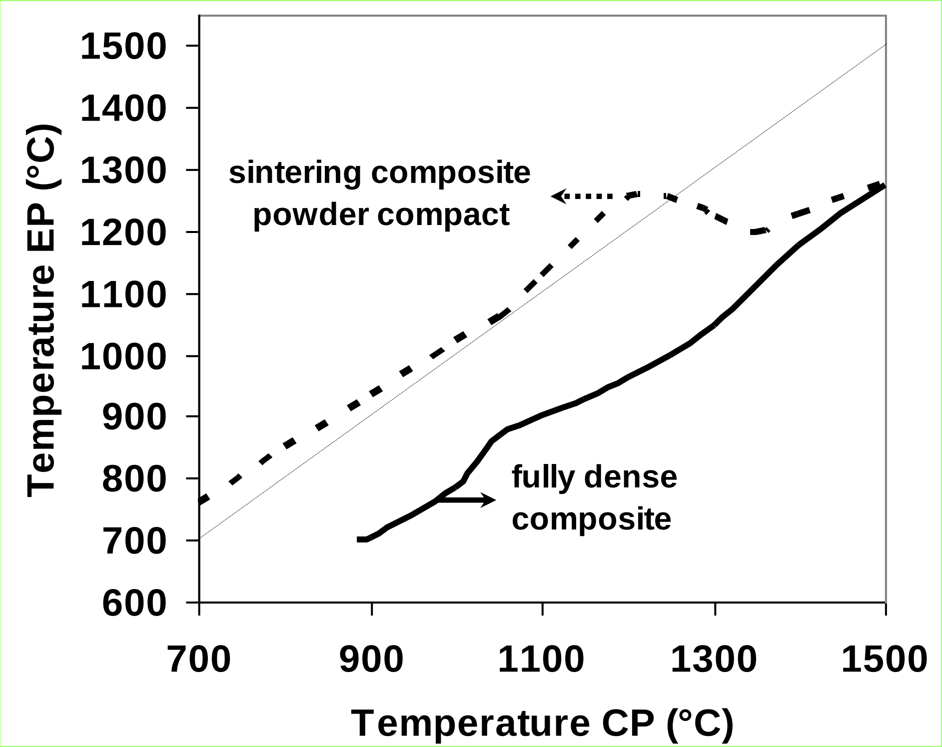
Dies



Punching tool

Development of temperature field during consolidation





Dense composite: $T_{die} << T_{compact}$

Porous composite: $T < 1250°C$: $T_{die} \sim T_{compact}$ $T > 1250°C$: $T_{die} << T_{compact}$

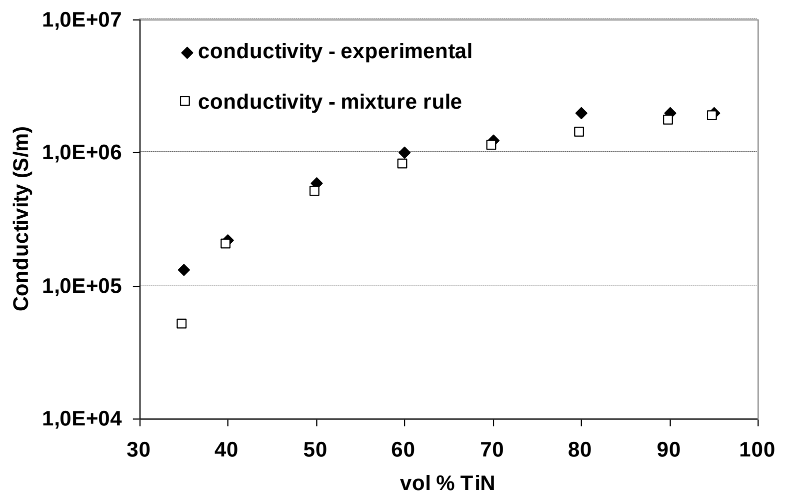
Influence of composition and porosity on properties

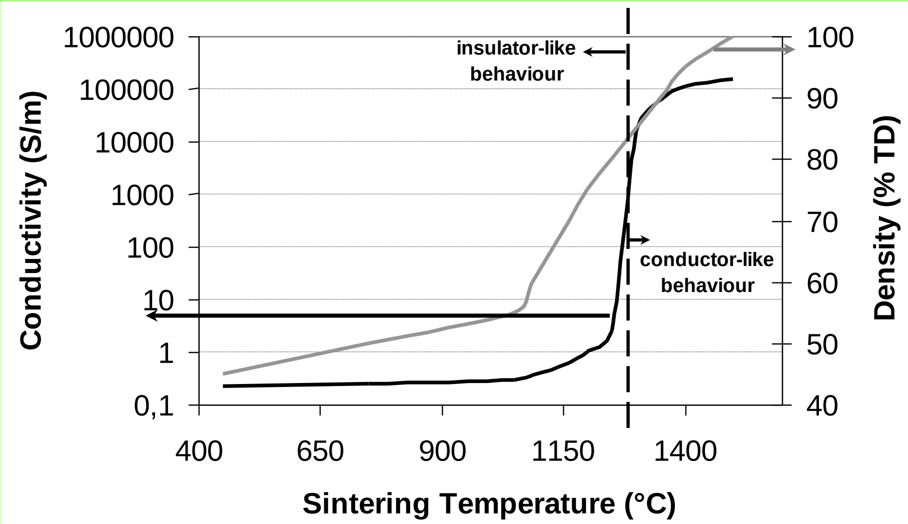
Influence of composition
(Polder – Van Santen mixture rule)

$$\rho_c = \rho_{ZrO_2} + \left(\rho_{TiN} - \rho_{ZrO_2}\right)\frac{V_{TiN}\rho_c}{\rho_c + \frac{\rho_{TiN} - \rho_c}{3}}$$

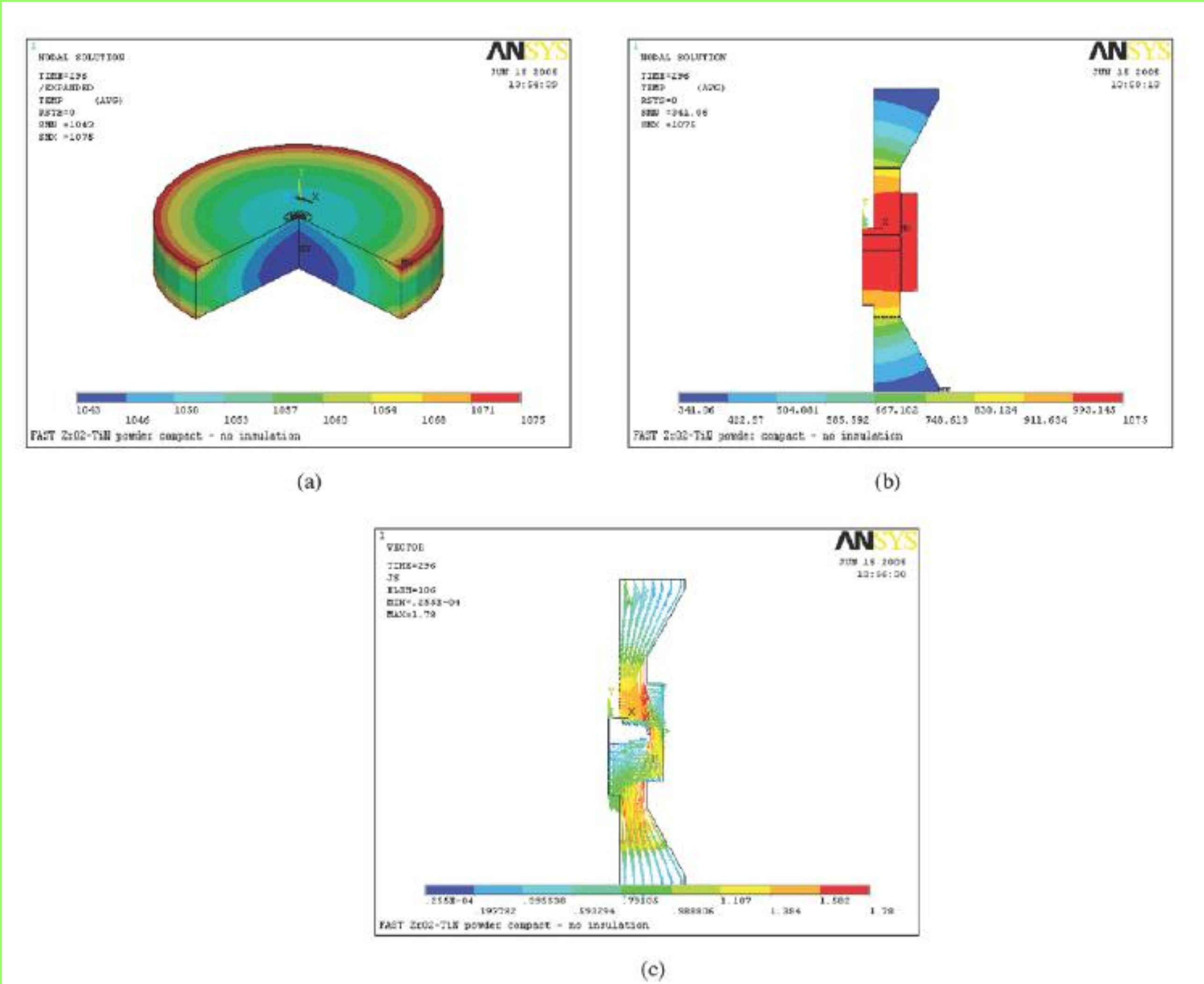
Influence of porosity
(formulae Argento – Bouvard)

$$k_{eff} = k_s\left(\frac{\rho - \rho_0}{1 - \rho_0}\right)^{t(1-\rho_0)}$$





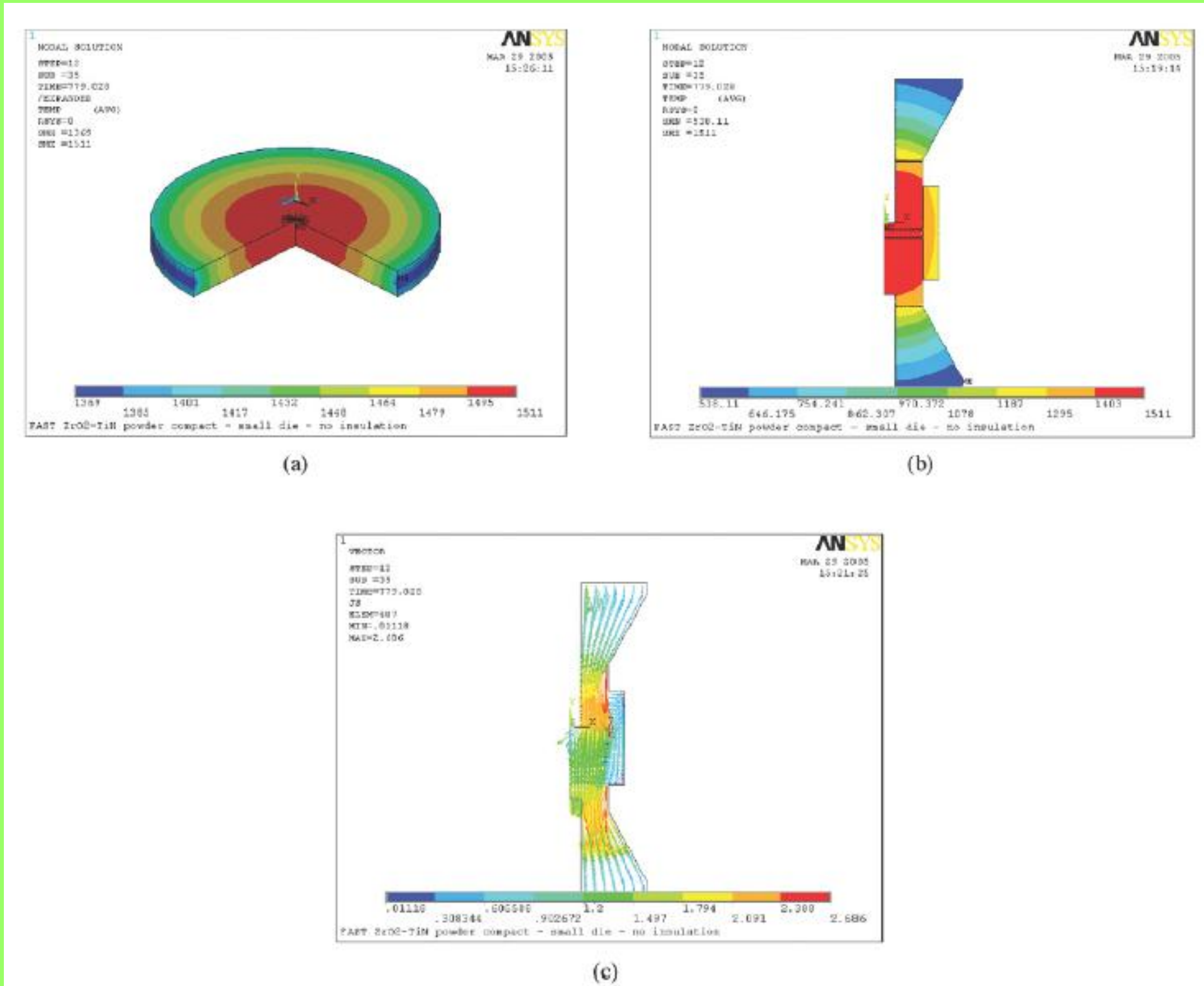
Temperature and current distribution at 1050°C



D=40 mm

ΔTmax=52°C

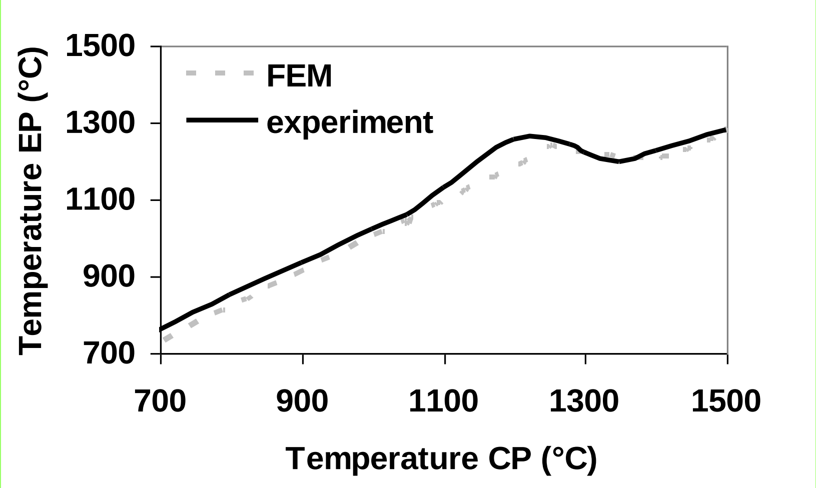
Temperature and current distribution at 1500°C



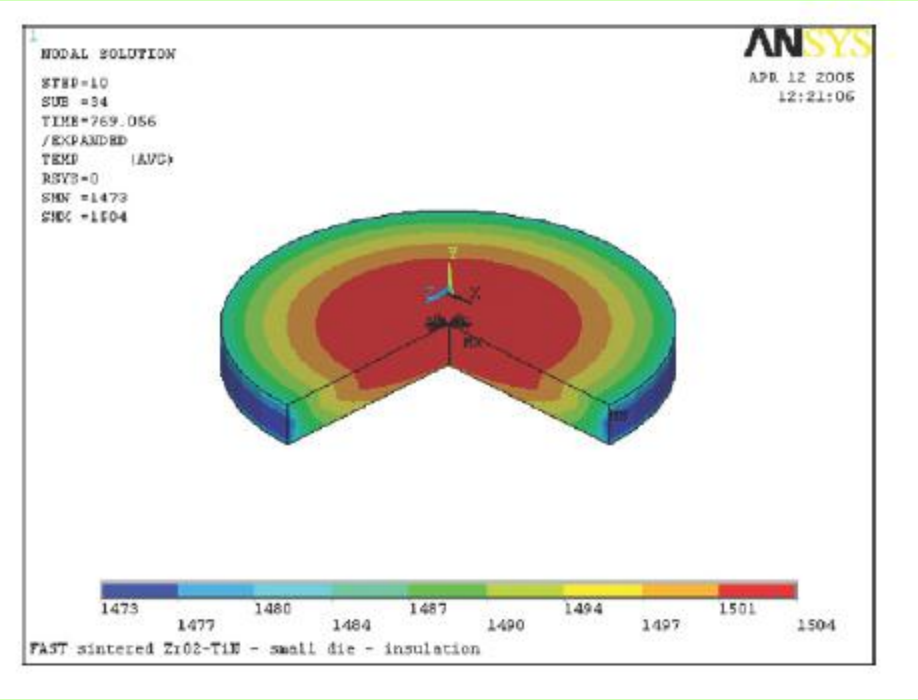
D=40 mm

ΔTmax=142°C

Accuracy of modeling and influence of thermal insulation



Modeling results vs pyrometers reading



Temperature distribution when thermal insulation of die is used. ΔTmax=31°C

Main conclusions (section # 2)

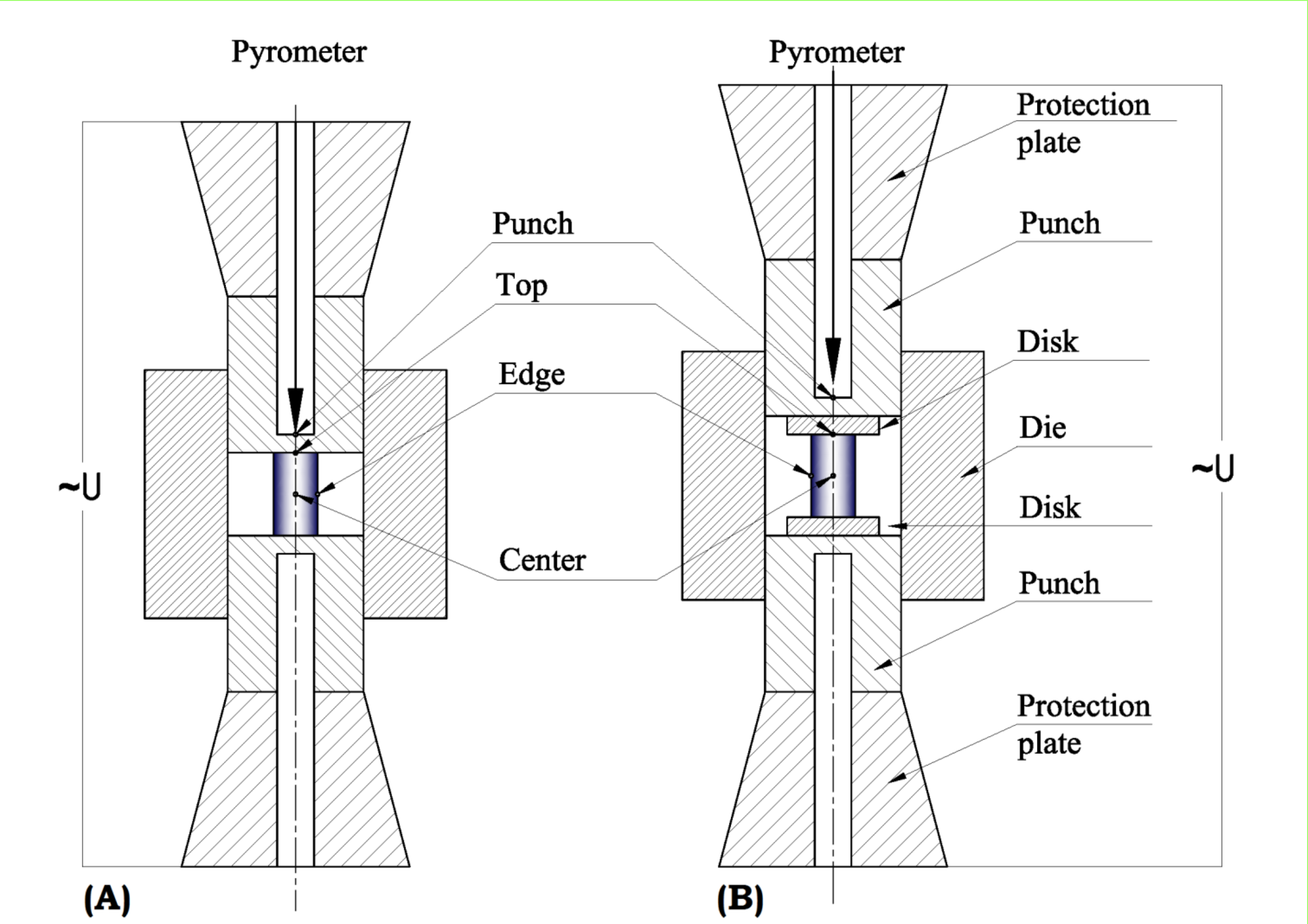
- The application of ceramics powder mixtures containing a conductive phase allows Spark Plasma Sintering of materials suitable for Electrical Discharge Machining.
- During sintering the conductivity of such composite material can be drastically reduced due to decrease of porosity. The Polder – Van Santen and Argento – Bouvard relationships can be used for modeling of conductivity change.
- FEM analysis has shown that application of thermal insulation results in an essential reduction of temperature gradient within a sample.

Plastic deformation of ceramics

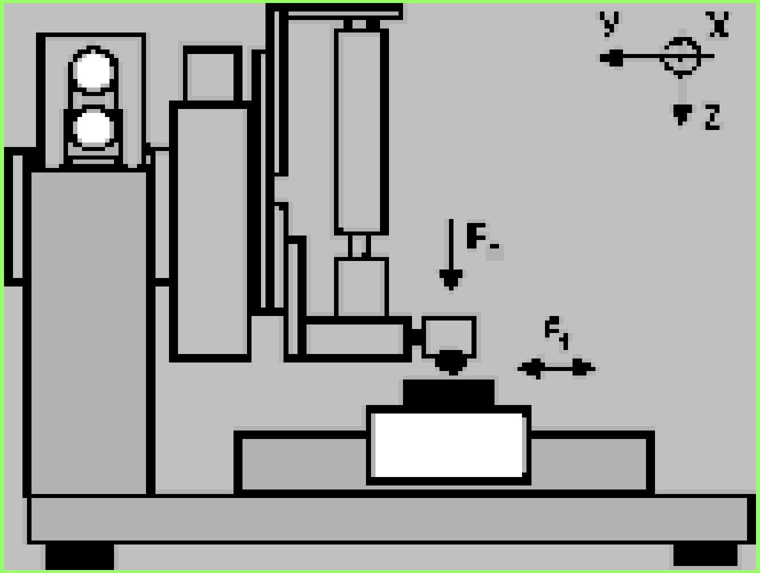
K. Vanmeensel, A. Laptev, H. Sheng, I. Tkachenko,
O. Van der Biest, J. Vleugels

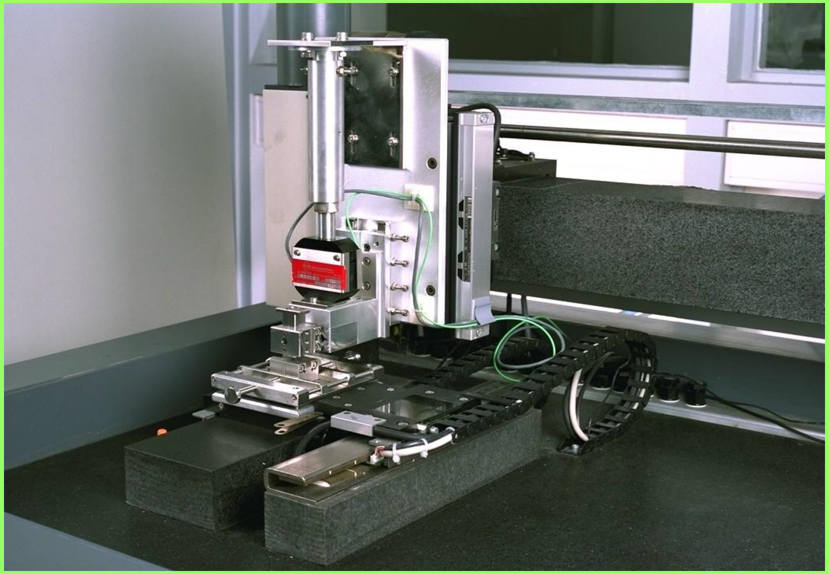
Acta Materialia 61 (2013) 2376–2389

Two tool setups used in experiments



Measurement of friction coefficient



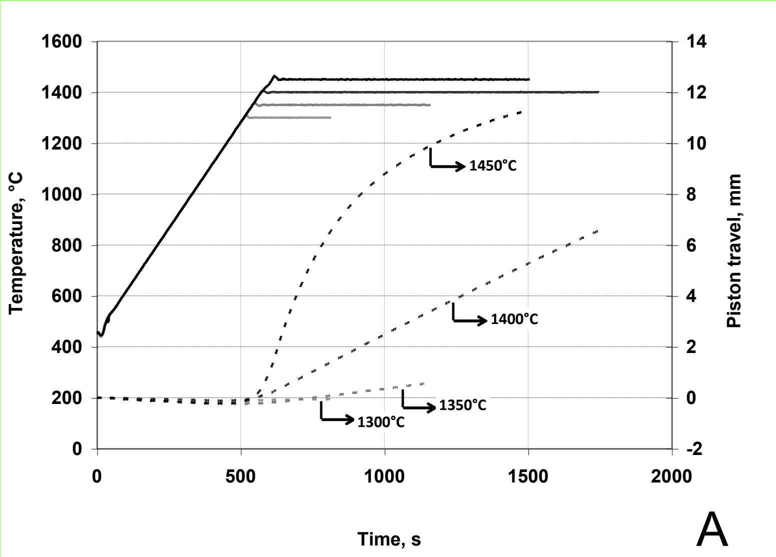


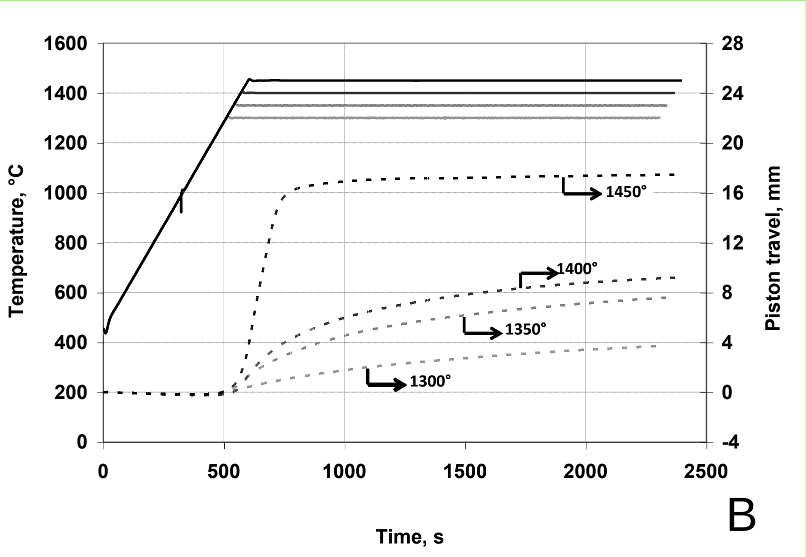
$$f_{av} = \frac{E_{fr}}{2 \cdot F_{fr} \cdot D}$$

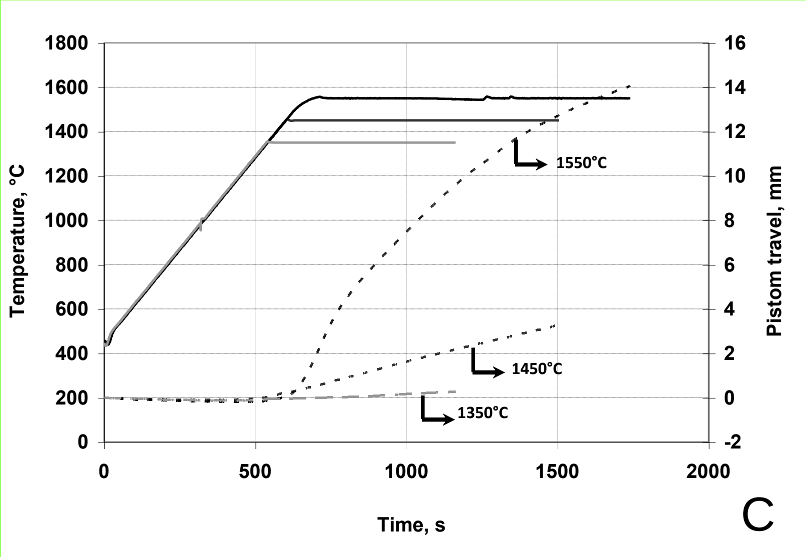
ZrO_2 bal: 10 mm; Load: 1 and 2 N;
Displacement: 1000 µm; Frequency: 1 Hz

E_{fr} – friction work; F_{fr} – friction force; D - displacement

Kinetics of plastic deformation





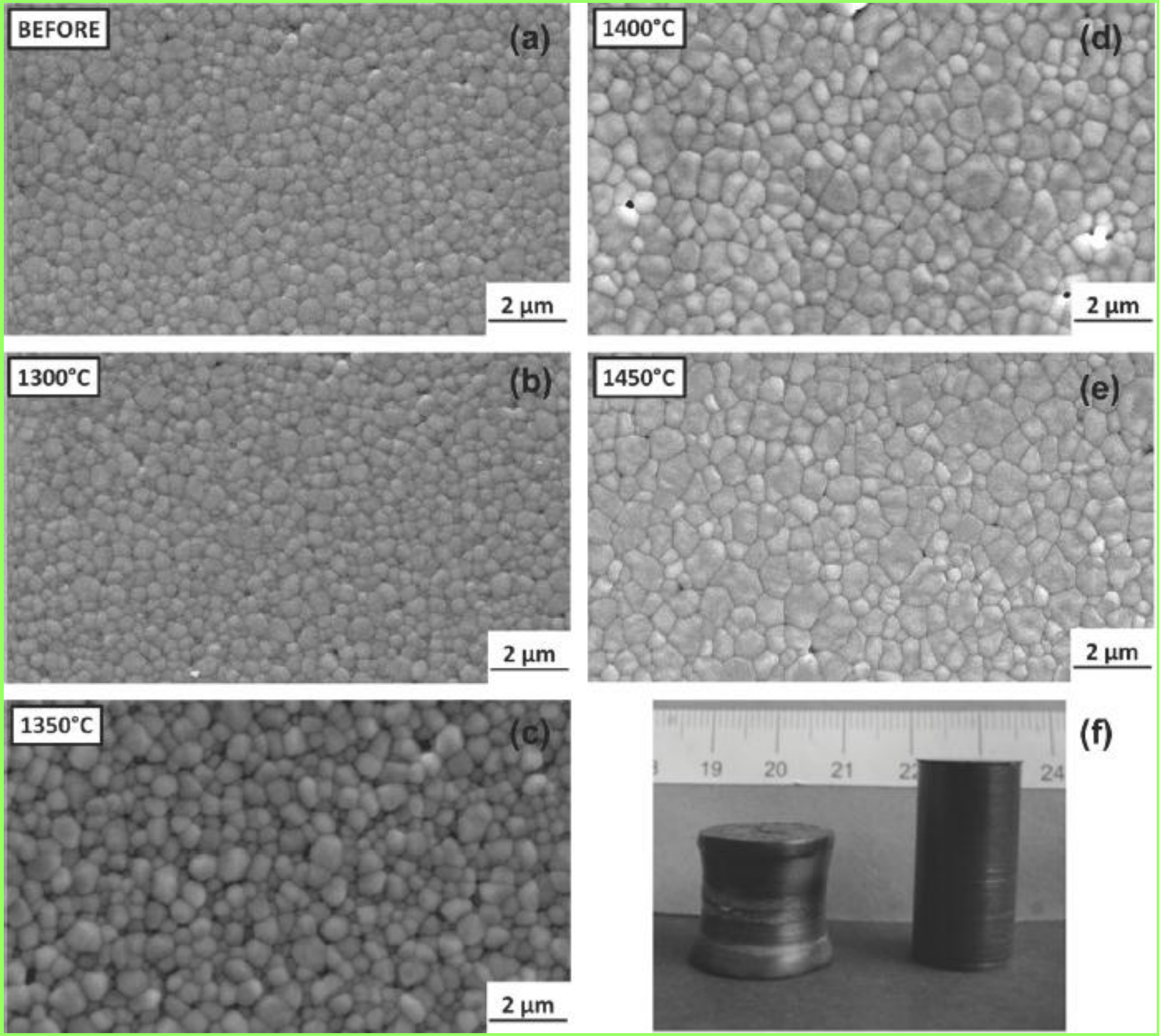


A: 3Y-ZrO_2

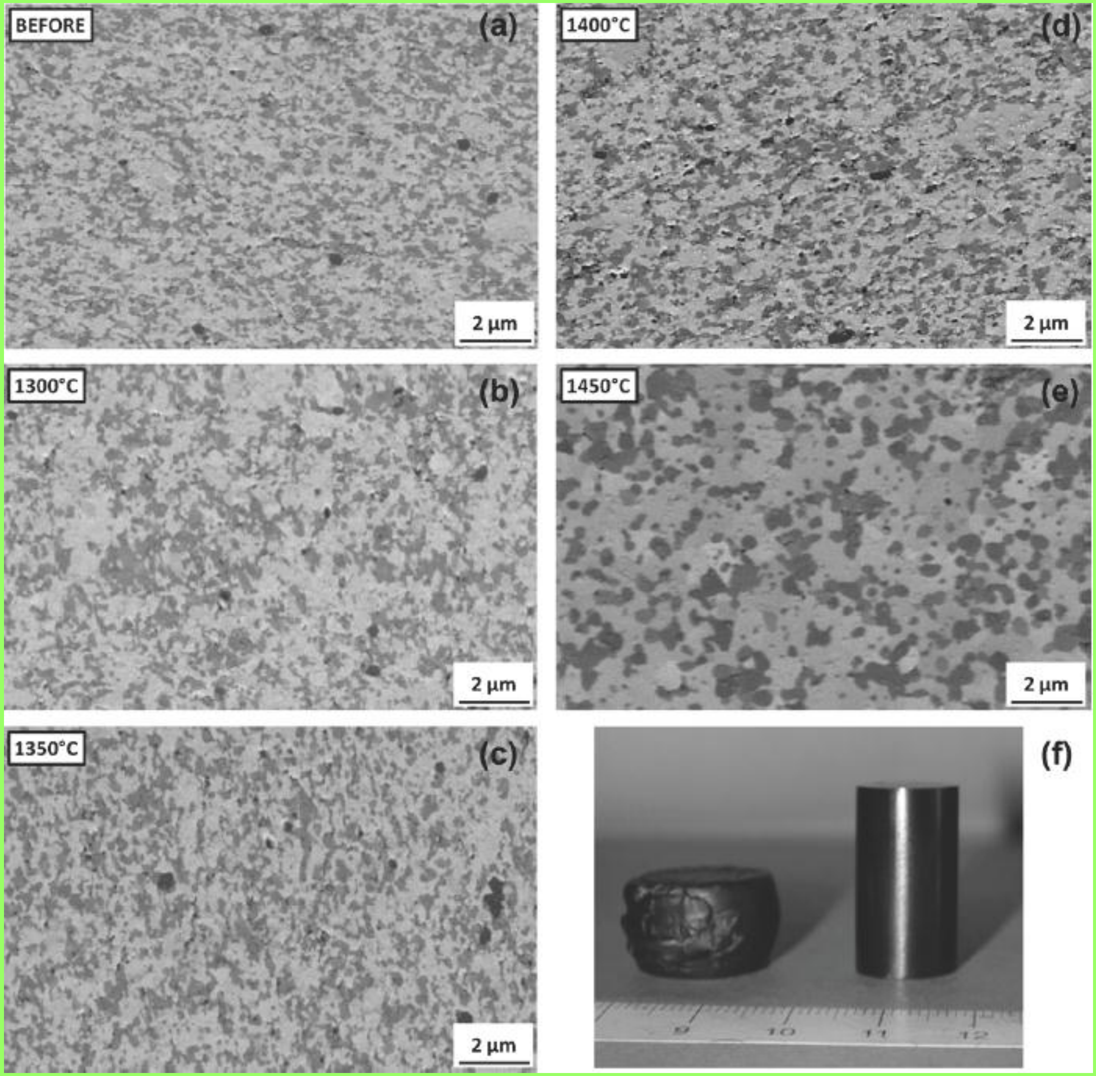
B: ZrO_2-TiCN 60/40 without disc

C: ZrO_2-TiCN 60/40 with disc

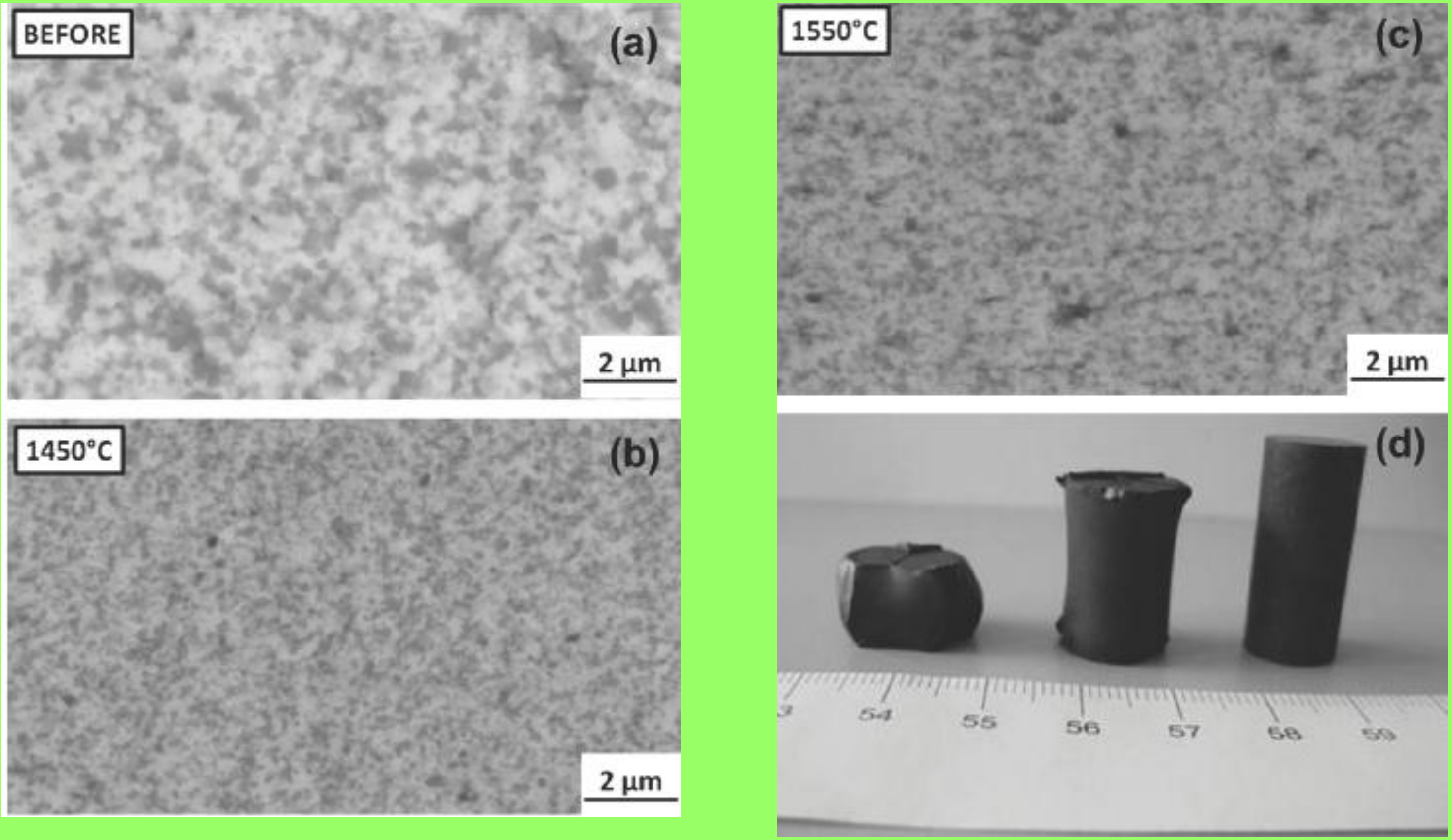
Microstructure and shape of 3Y-ZrO_2 samples before and after deformation



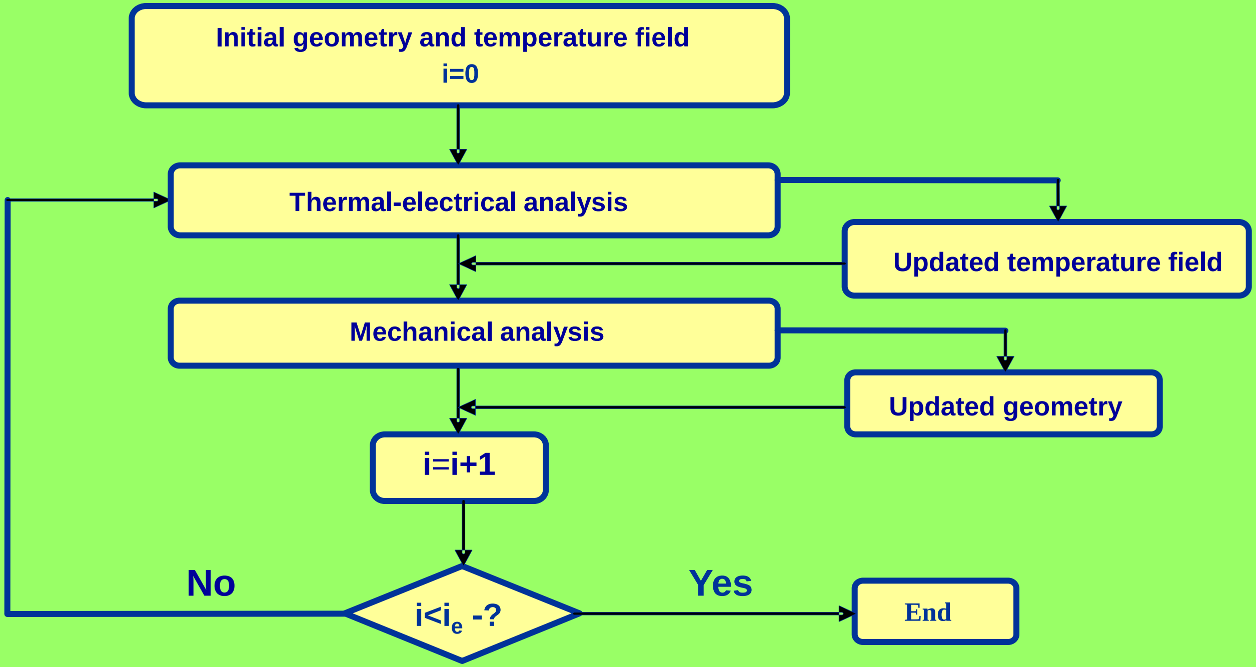
Microstructure and shape of ZrO_2-TiCN samples after deformation without SiC discs



Microstructure and shape of ZrO_2-TiCN samples after deformation with SiC discs

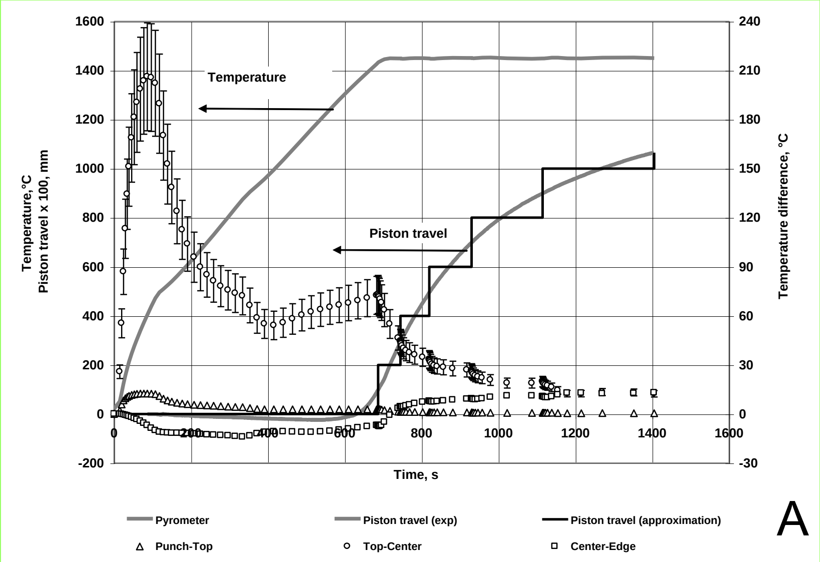


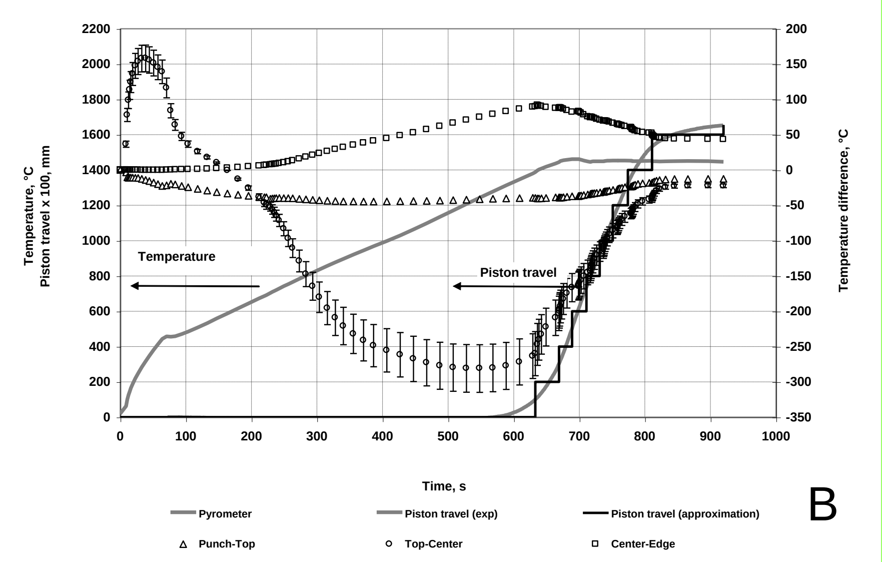
Iteration procedure of FEM analysis

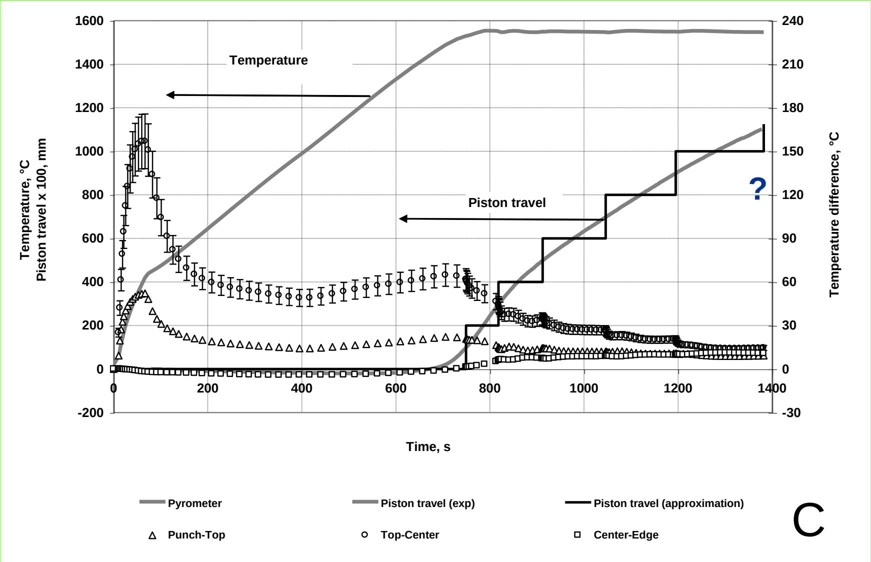


$$\dot{\varepsilon}_{eff} = A_0 \frac{\sigma_{eff}^n}{d^p} \exp\left(-\frac{Q}{RT}\right)$$

Results of FEM modeling





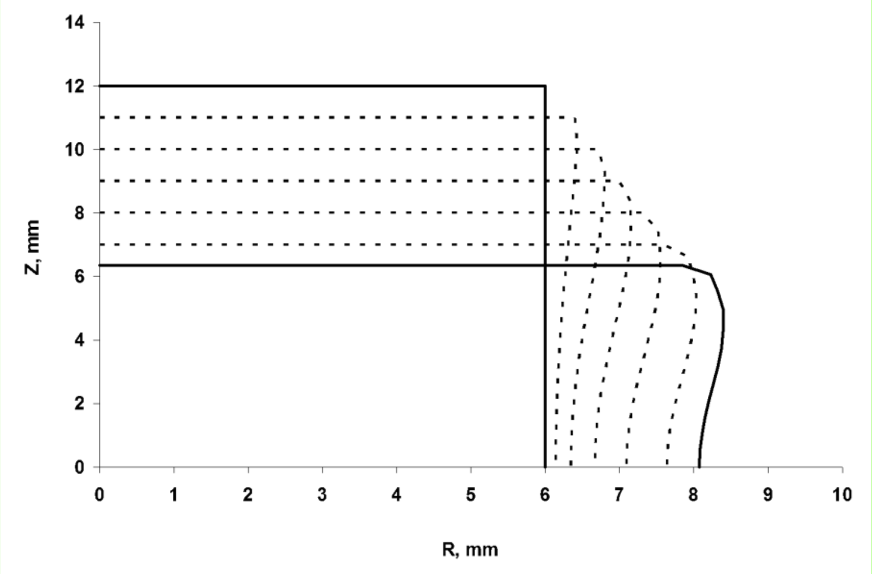


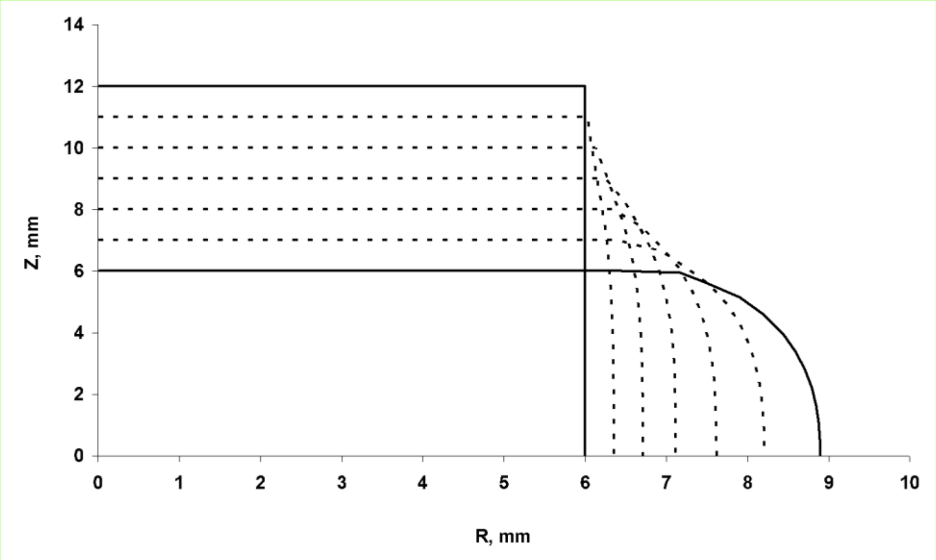
A: 3Y-ZrO_2

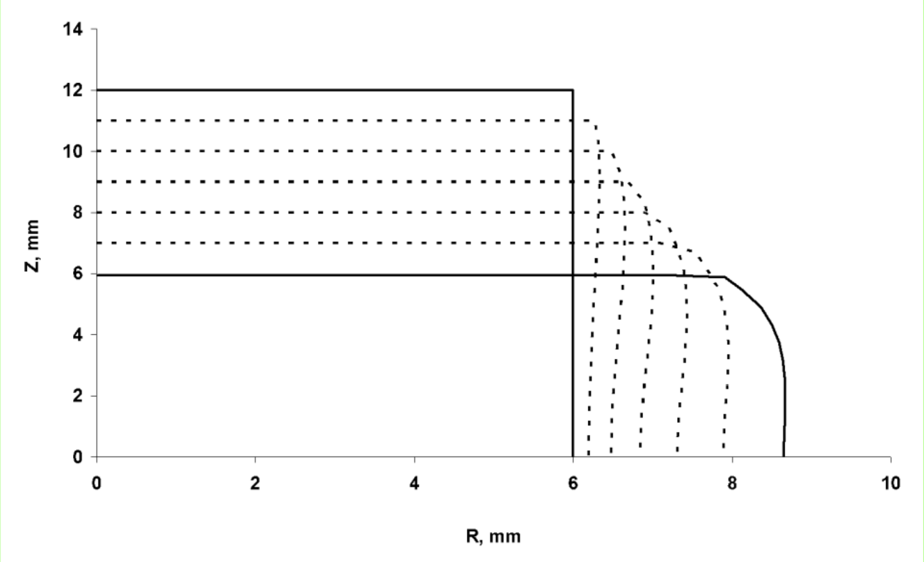
B: ZrO_2-TiCN 60/40 without disc

C: ZrO_2-TiCN 60/40 with disc

Results of FEM modeling





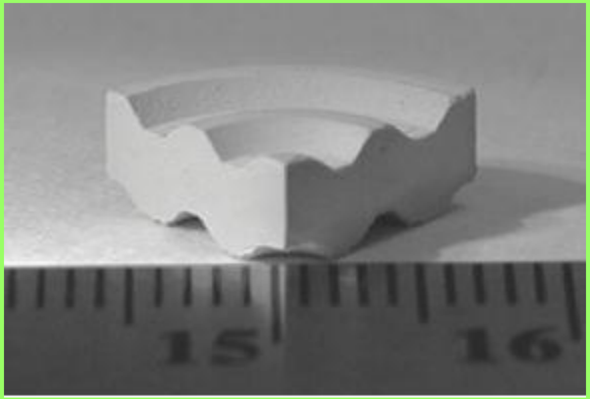


A: 3Y-ZrO_2

B: ZrO_2-TiCN 60/40 without disc

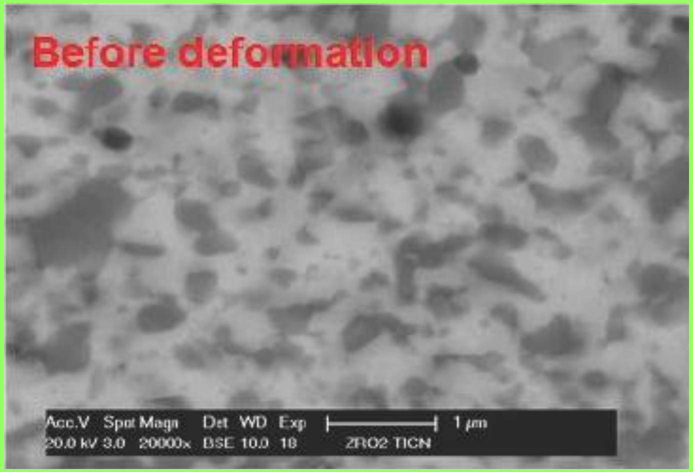
C: ZrO_2-TiCN 60/40 with disc

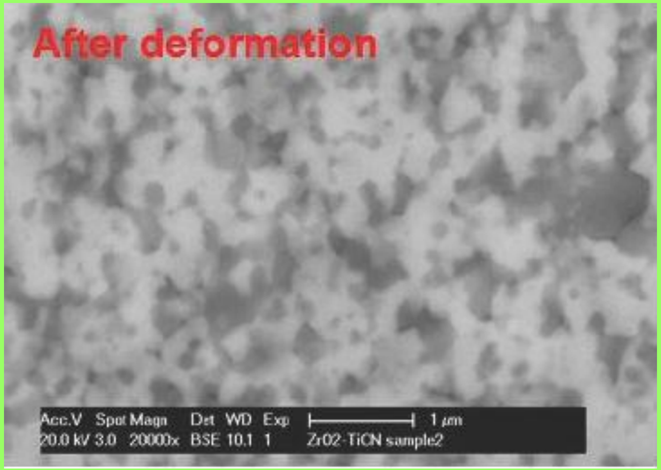
Examples of ceramics forming in SPS apparatus



Al_2O_3-ZrO_2-$MgAl_2O_4$

Jiang D., Hulbert D.M., Kuntz J.D., Anselmi-Tamburini U., Mukherjee A.K. Materials Science and Engineering A 463 (2007) 89–93





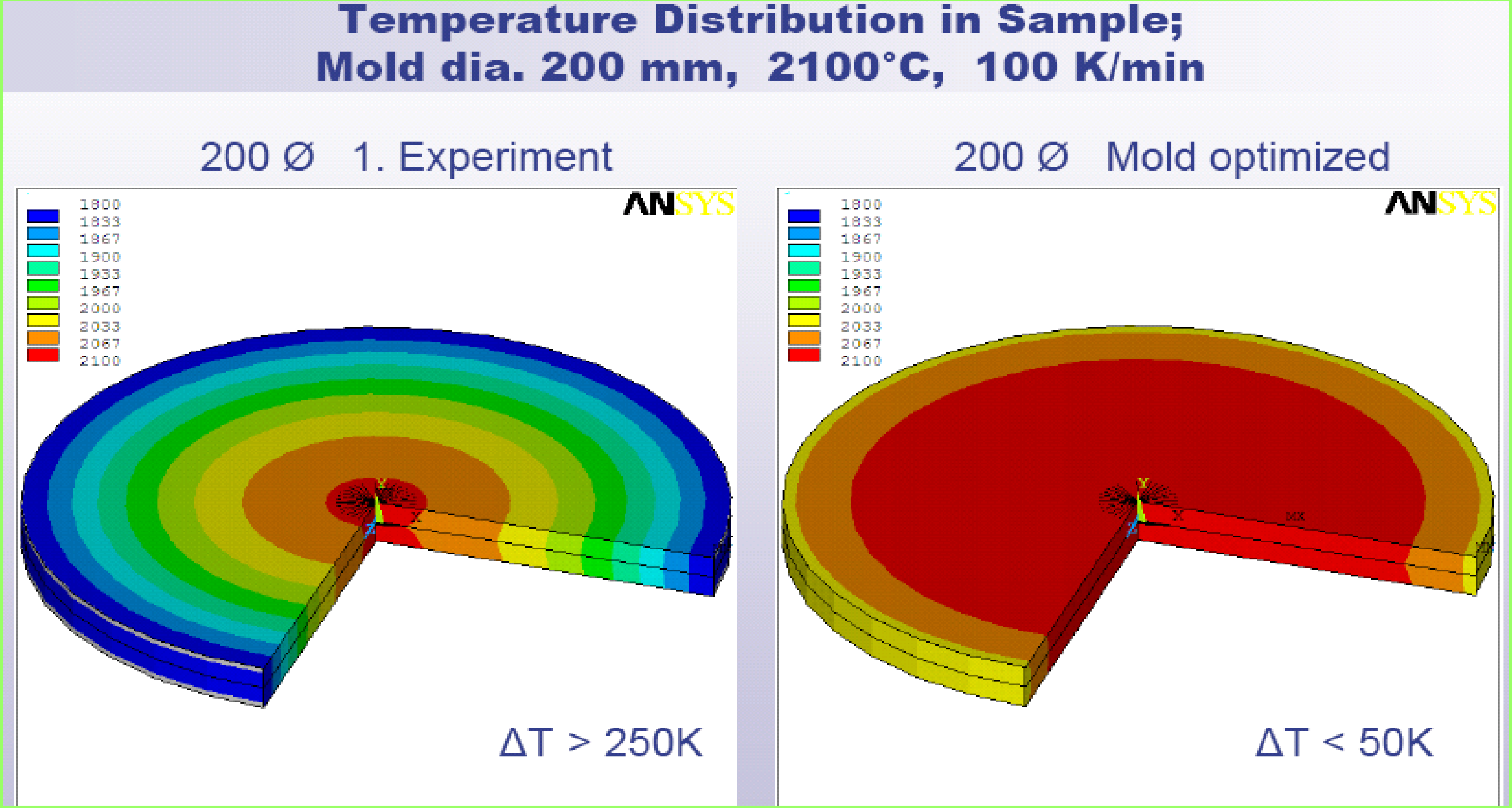
ZrO_2-TiCN

Vanmeensel K. KU Leuven

Main conclusions (section # 3)

- SPS technique can be successfully applied for plastic deformation of both conductive and non-conductive ceramics at deformation rates in the range of 10^{-2}-10^{-3} s^{-1}.
- The presence of secondary TiCN phase retards the grain growth of Zirconia.
- Predominant electrical current paths can essentially influence the temperature distribution in a sample and its shape after deformation. The current paths can be controlled by die design.
- Temperature gradient is especially large at heating and at the beginning of deformation. The gradient diminishes with decrease in sample height and during dwell period at maximal temperature.
- An innovative subsequently coupled thermal–electrical–mechanical FEM procedure was developed to predict temperature, current, voltage as well as stress distributions and geometrical sample change during deformation in a SPS apparatus.

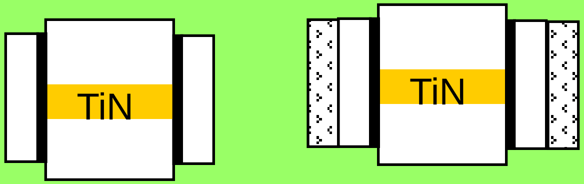
Optimization of die design



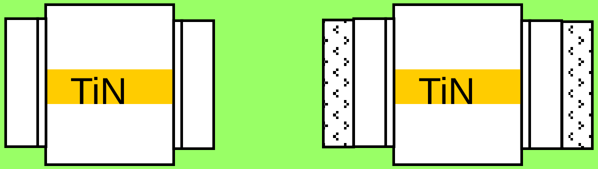
Courtesy: FCT systeme

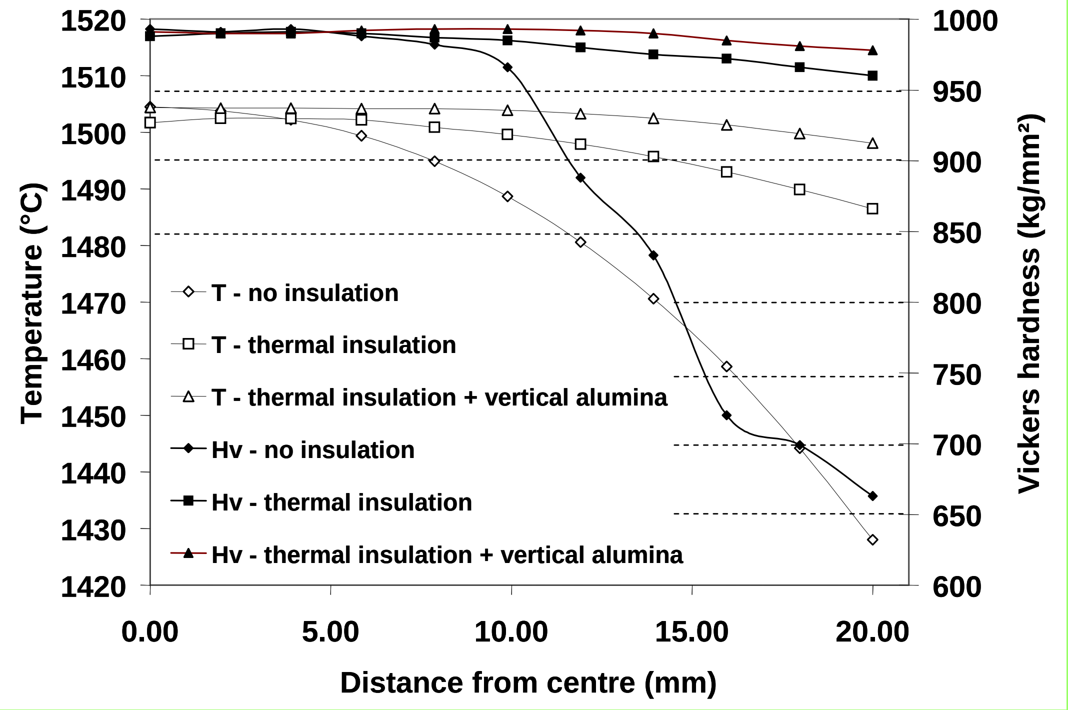
Influence of insulation on temperature and hardness distribution

A. 40 mm diameter die vertical graphite paper



B. 40 mm diameter die vertical alumina paper





Further work

- The change from explanation to prediction of observed at SPS phenomena is necessary.
- The development of a simple method for determination of rheological characteristics of materials needed for SPS modeling is desirable.
- The creation of properties data base for materials processed by SPS technology and for materials used for tool is advisable.
- The special subroutines for modeling of SPS processes taking into account a complex rheology of sintered materials should be written.

Acknowledgment

A. Laptev thanks Research Council of Katholieke Universiteit Leuven for giving possibility of Spark Plasma Sintering investigation in Department of Metallurgy and Materials Engineering by fellowship F/02/096 and Ministry of Education and Science of Ukraine for financial support of research through project No 0107U001301


Thank You for attention !!!