Self-sustained Waves of Exothermic Dissolution in Reactive Multilayer Nanofoils

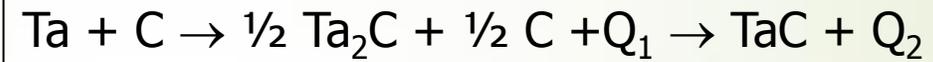# The Phenomenon of Wave Localization for Solid State Self-propagating Reactions

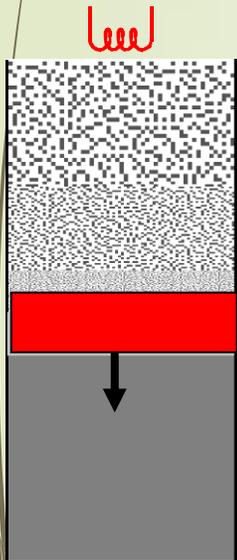**Present-day name:**

Self-propagating High-temperature Synthesis (SHS)

A.G. Merzhanov, V.M. Shkiro, I.P. Borovinskaya, 1967

Combustion products (solid)

Combustion zone (solid)

Initial reagents (solid)

$$Ta + C \rightarrow \tfrac{1}{2}\, Ta_2C + \tfrac{1}{2}\, C + Q_1 \rightarrow TaC + Q_2$$

T_{ad} = 2730 K, T_m = 3100 K
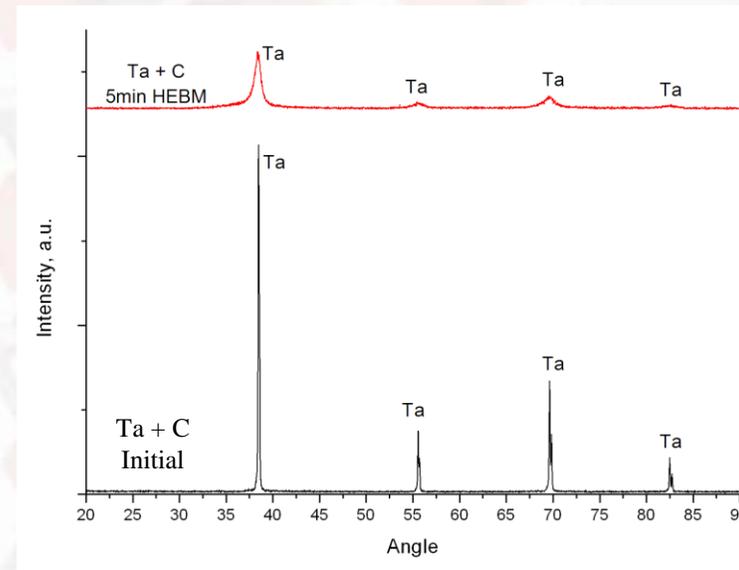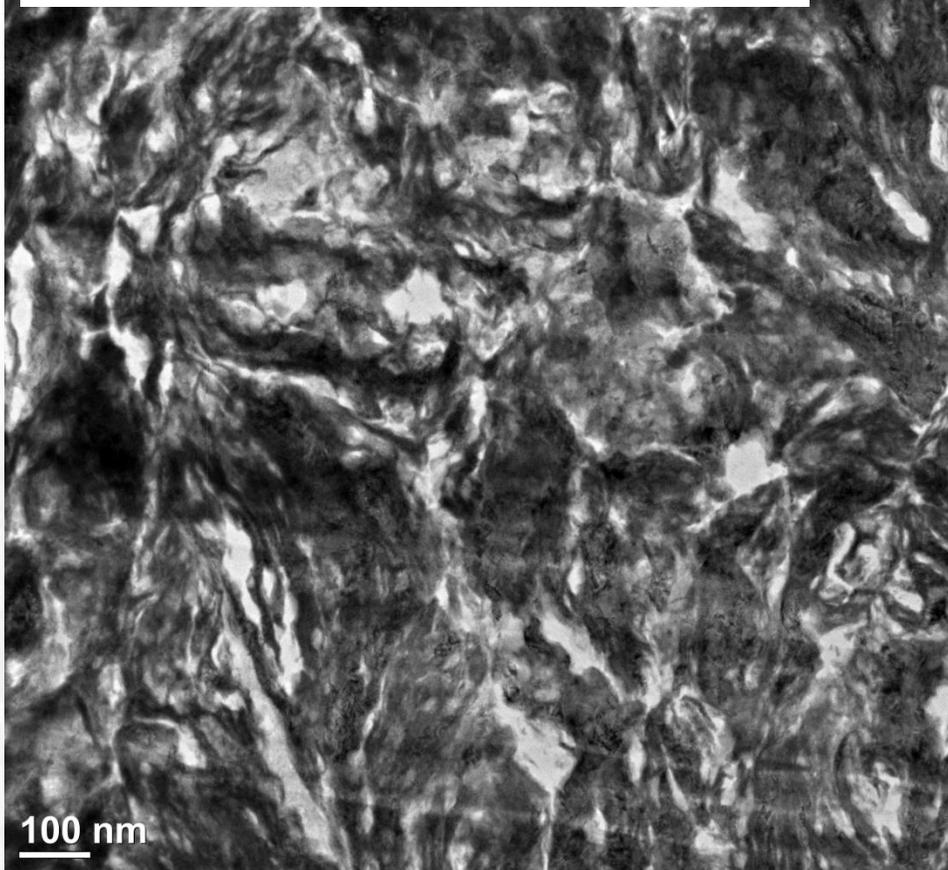

ignition   front propagation   cooling

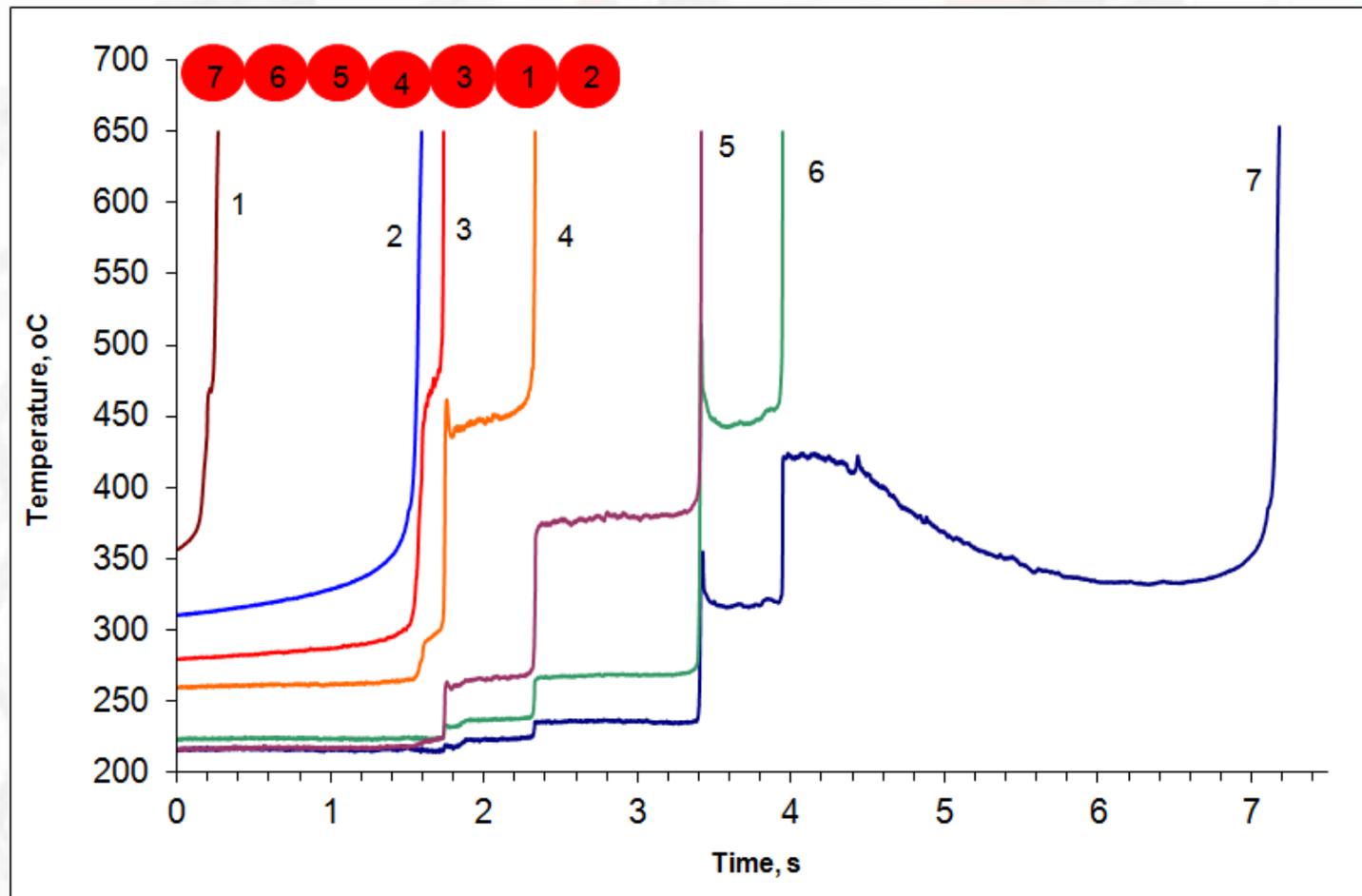# *Solid Flame:*

Recent Advances

Ta + C Reactive Media
after High Energy Ball Milling (HEBM) (2012)

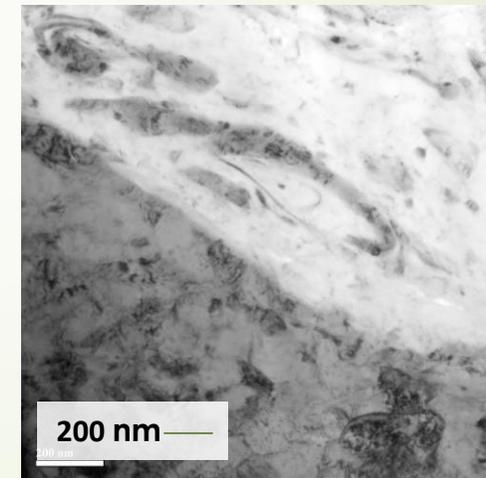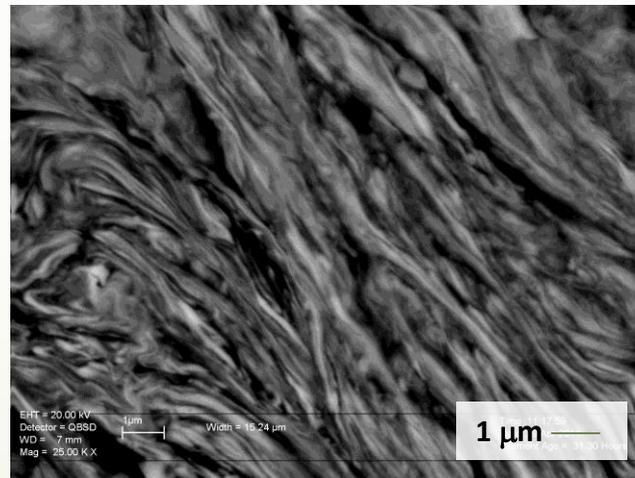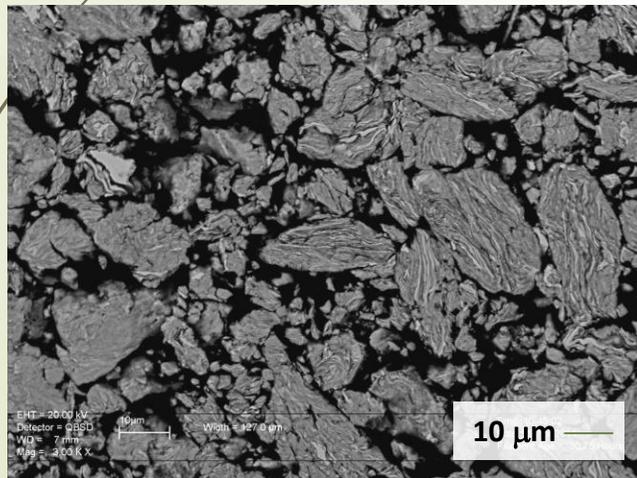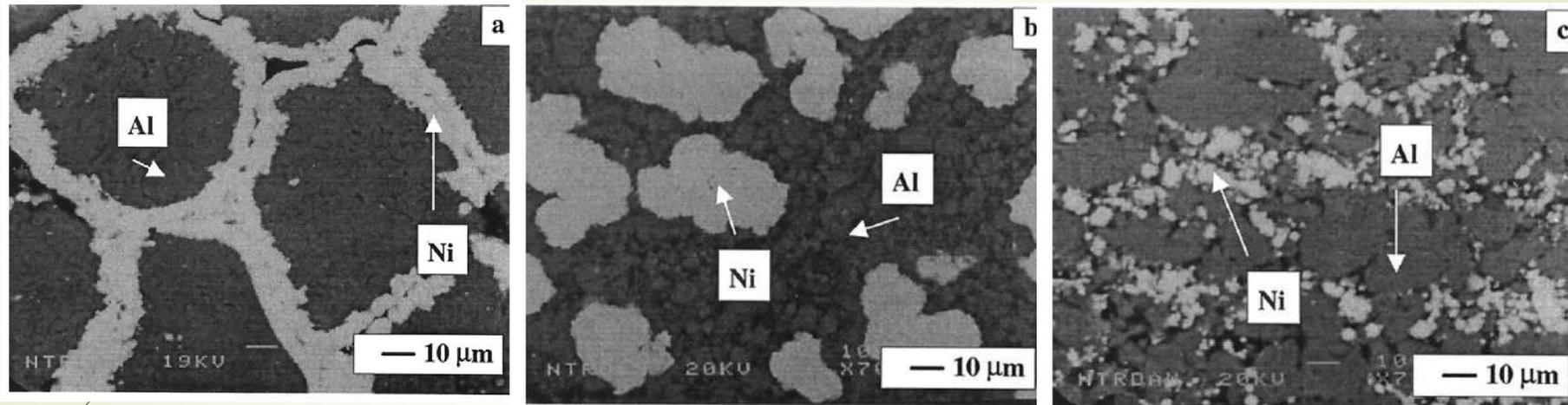Nano-Structure of Composite Ta-C particle

100 nm

Comparison of XRD patterns for Ta-C system before (a) and after (b) HEBM

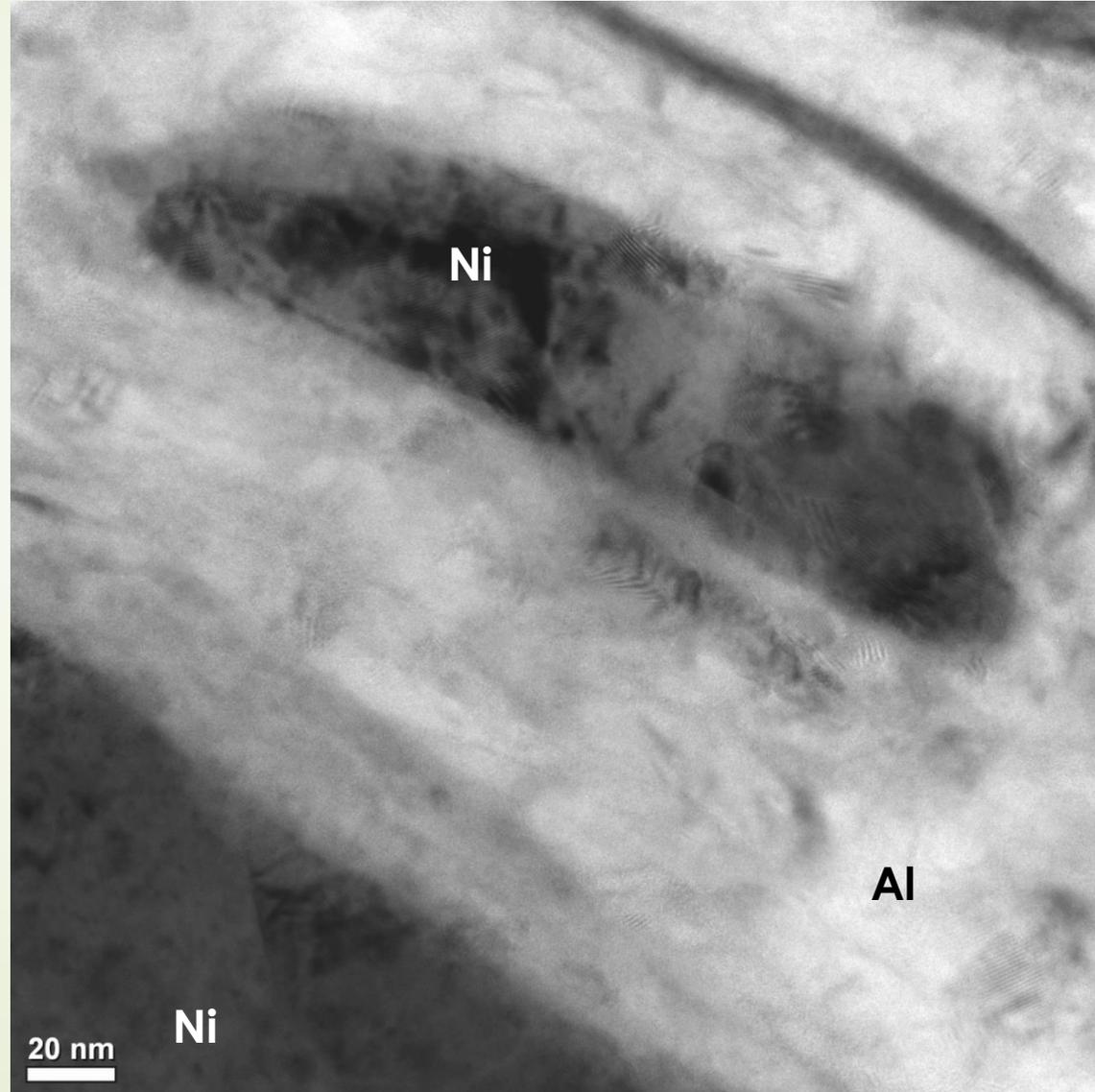# Reaction Front Propagation in Ta-C Nano Heterogeneous System

Ignition temperature of each composite particles ~450◦C !!!

Composite Reactive Nano-particles by HEBM: *Definition*
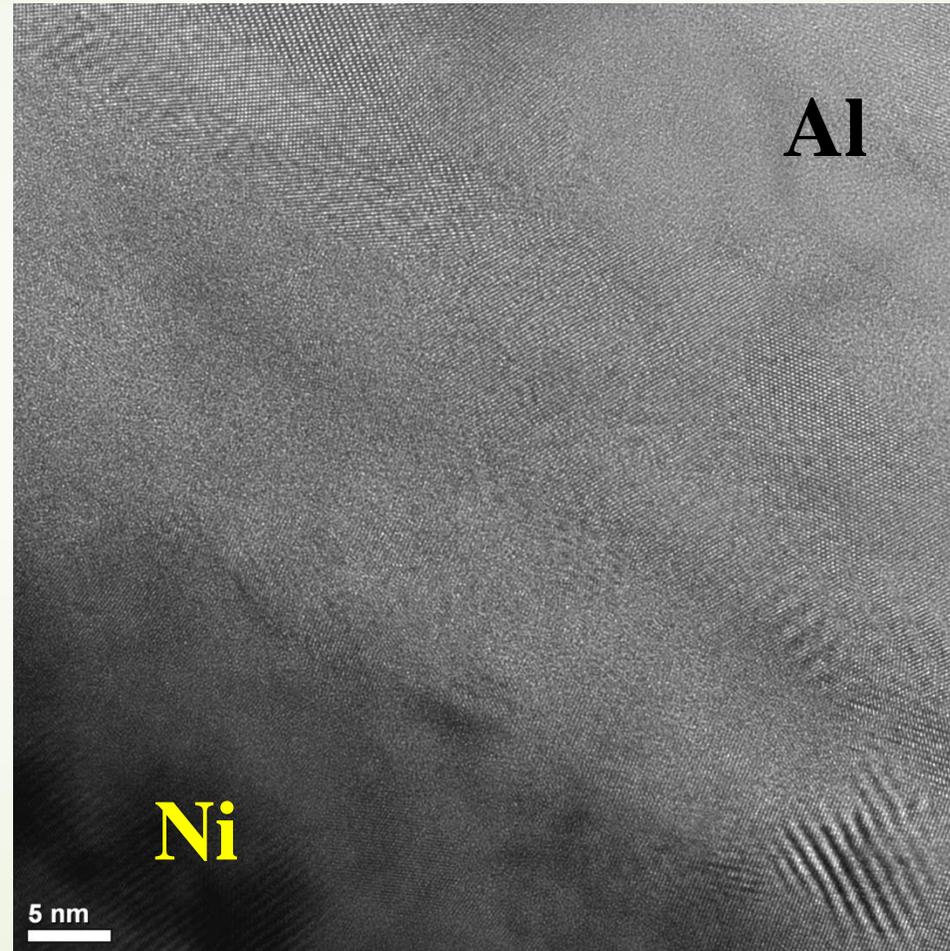

Particles obtained by intensive short term High Energy Ball Milling (HEBM) of reactive powder mixtures.
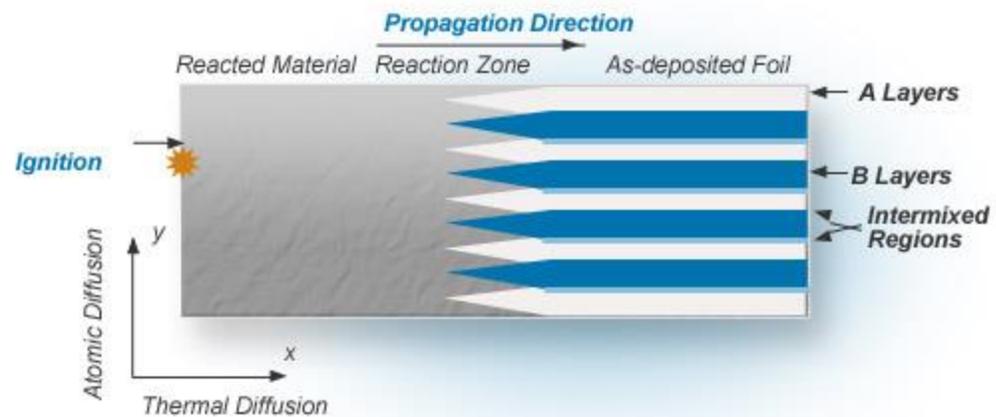
Bright-field TEM image of Ni-Al Media:
Nano Mixing

Bright-field TEM image:

Atomic Mixing

Reactive Nano-Foils
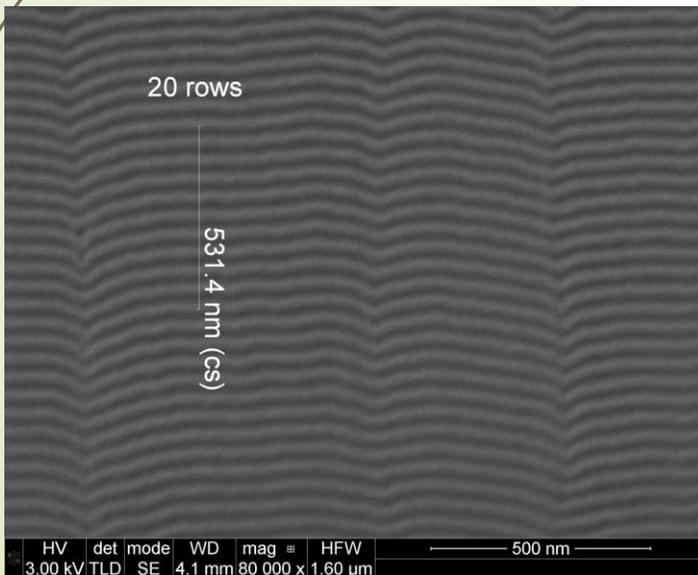
Production of Reactive Multilayer Nanofoils

Simultaneous operation of 2 or more magnetrons

↓

Layer-by layer deposition

↓

Free-standing foil

Source of Al atoms   Source of Ti atoms

100-5000 layers
5-100 nm each

T.Weihs et al. – J.Hopkins Univer., USA, 1996;
P.Tsygankov et al. – Bauman Technical University, Russia , 2000.

Reactive Multilayer Nanofoil
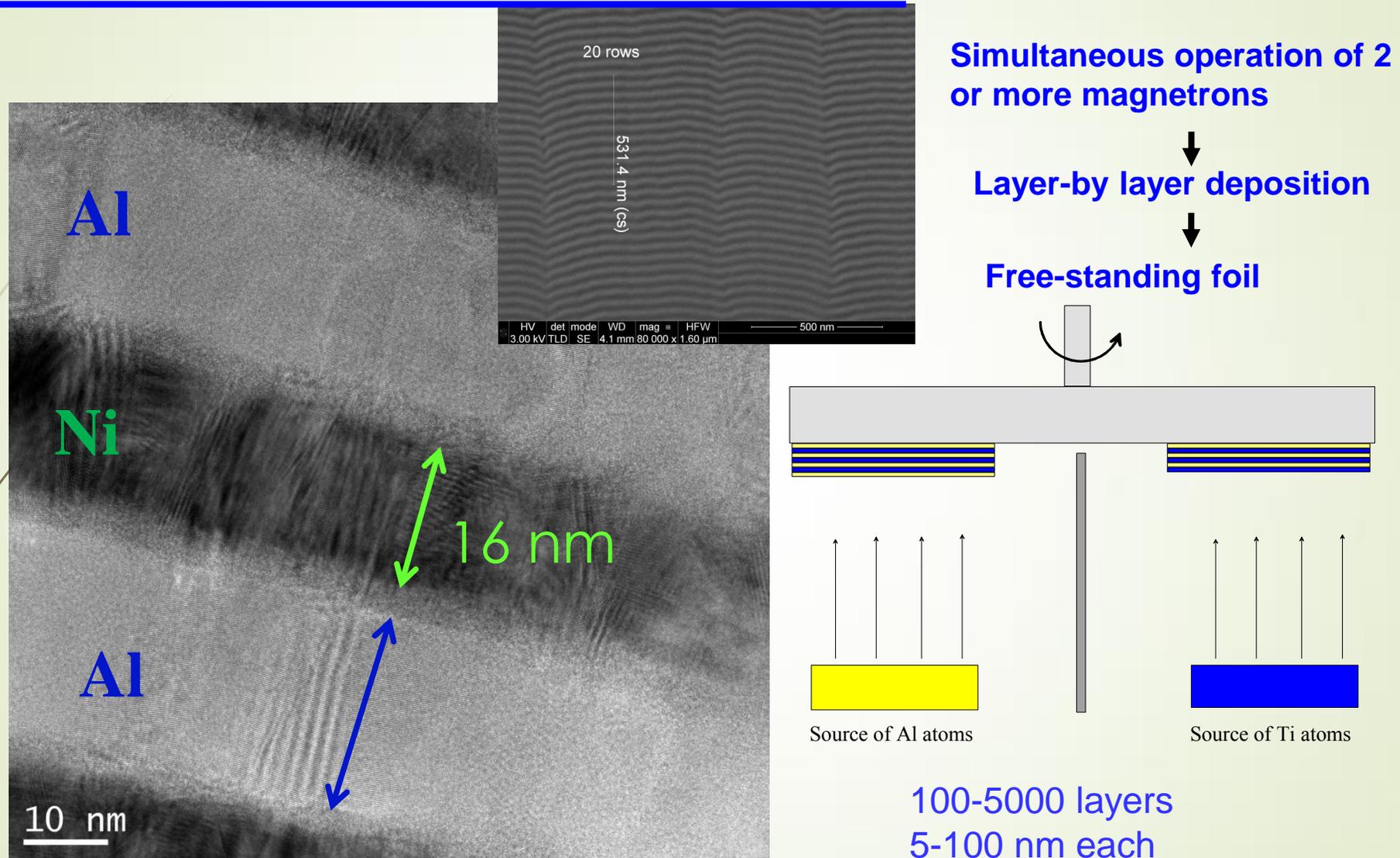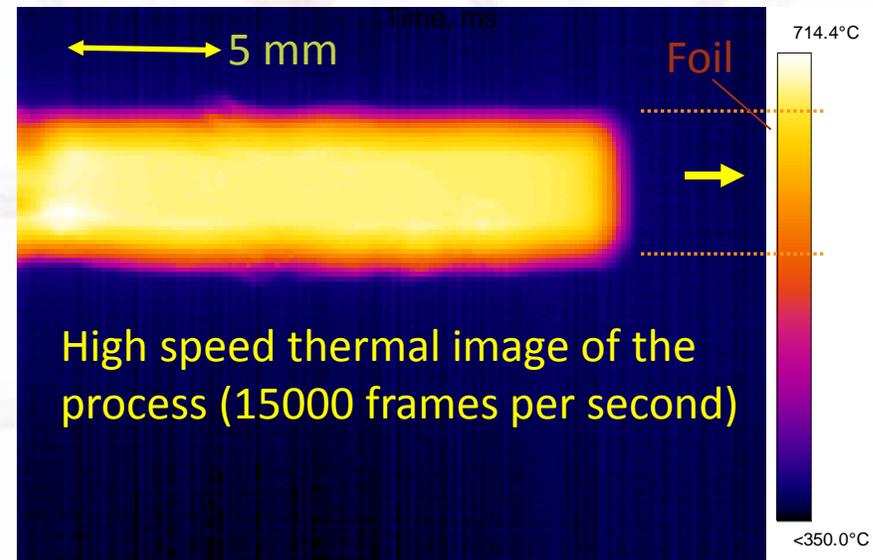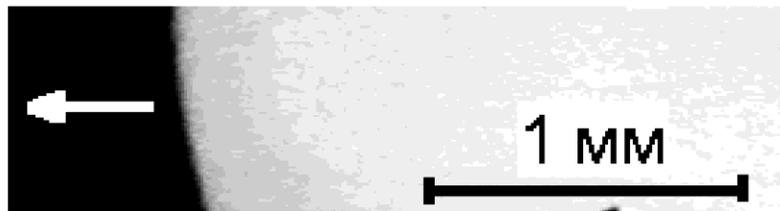

Ni/Al nanofilm (25 nm) by high speed video, 500,000 frames per second

$$U = 8,90 \pm 0,04 \quad m/s$$

б

5 mm

Foil

714.4°C

<350.0°C

High speed thermal image of the process (15000 frames per second)

(a) 4000 kg
Cu block
Foil — Ignition
Cu block

(b) indentation of the foil | line of extinction | point of ignition
10 mm

(c)
5 μm
0 — L
Initial layers | Quenched front of the reaction | Product

Top view (SEM)
10 μm
Cutting ion beam
W-wire
Nano - handler
W-wire
Deposited Pt bridge
TEM sample
Initial lamellar structure | Quenched front of reaction wave | Final products

Microstructure evolution behind the reaction front
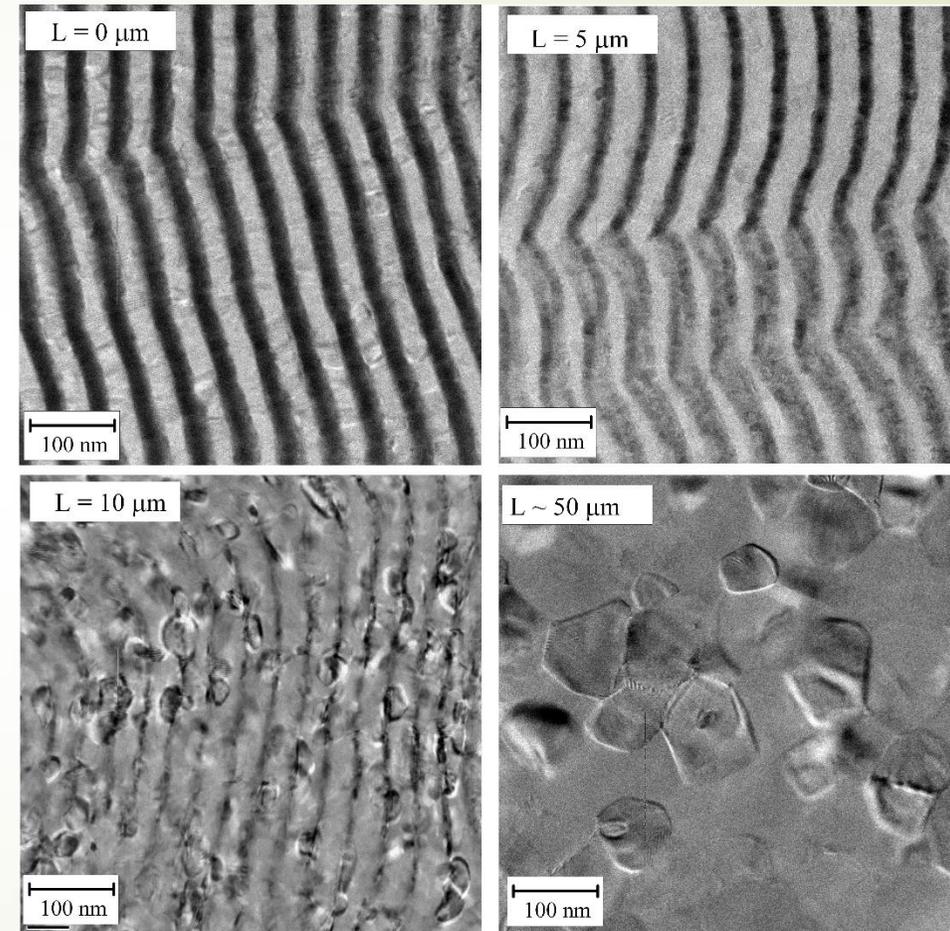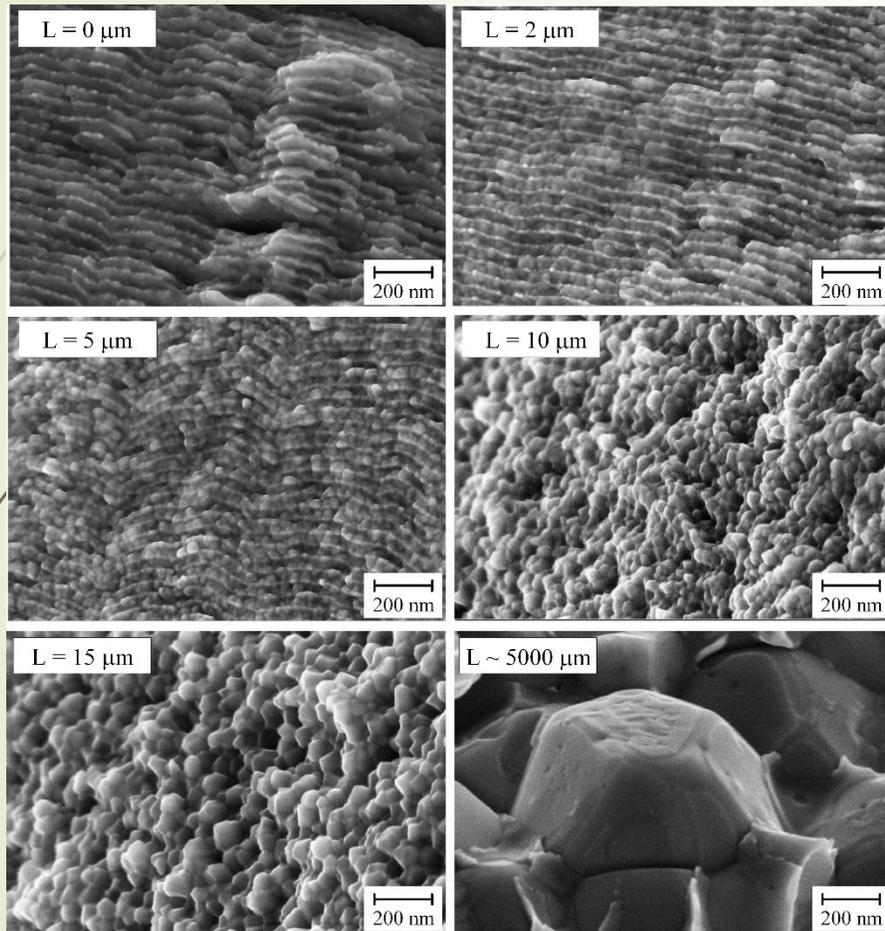
Grain size Growth in Quenched Front
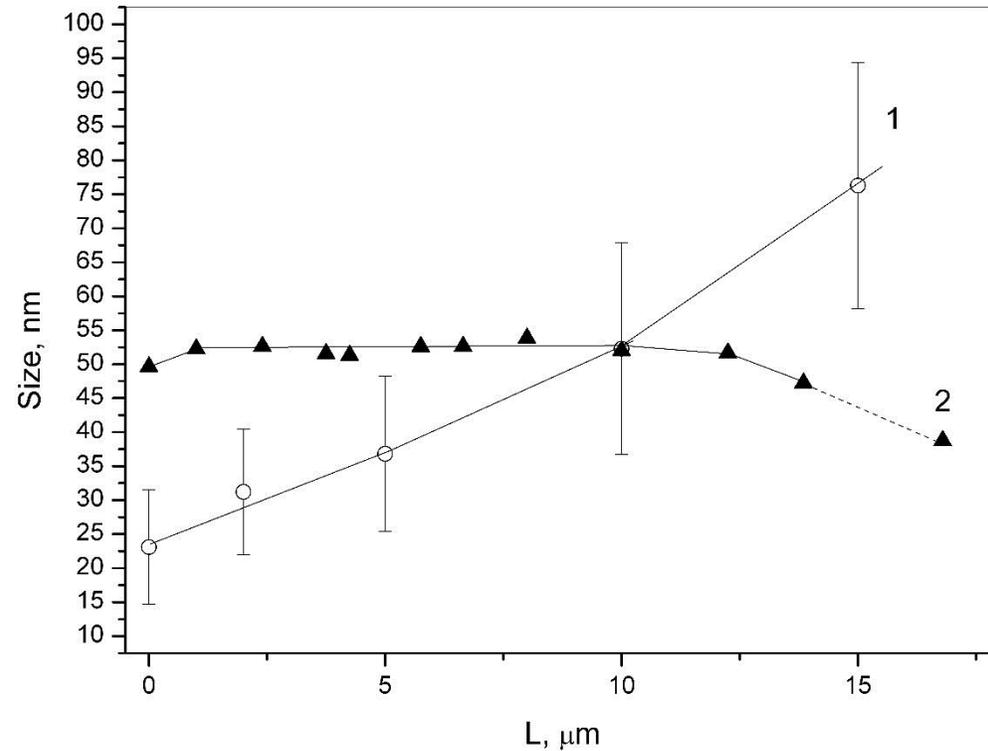
Dynamics of grain size (1) and bilayer thickness (2) behind the reaction front.
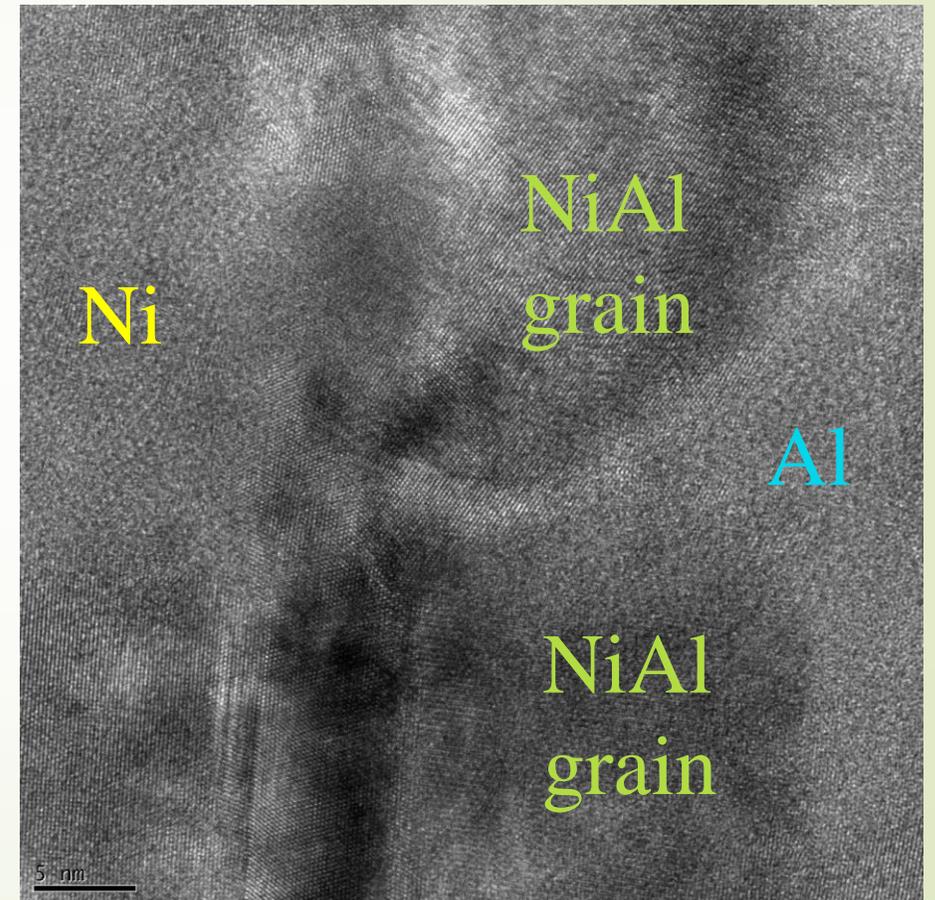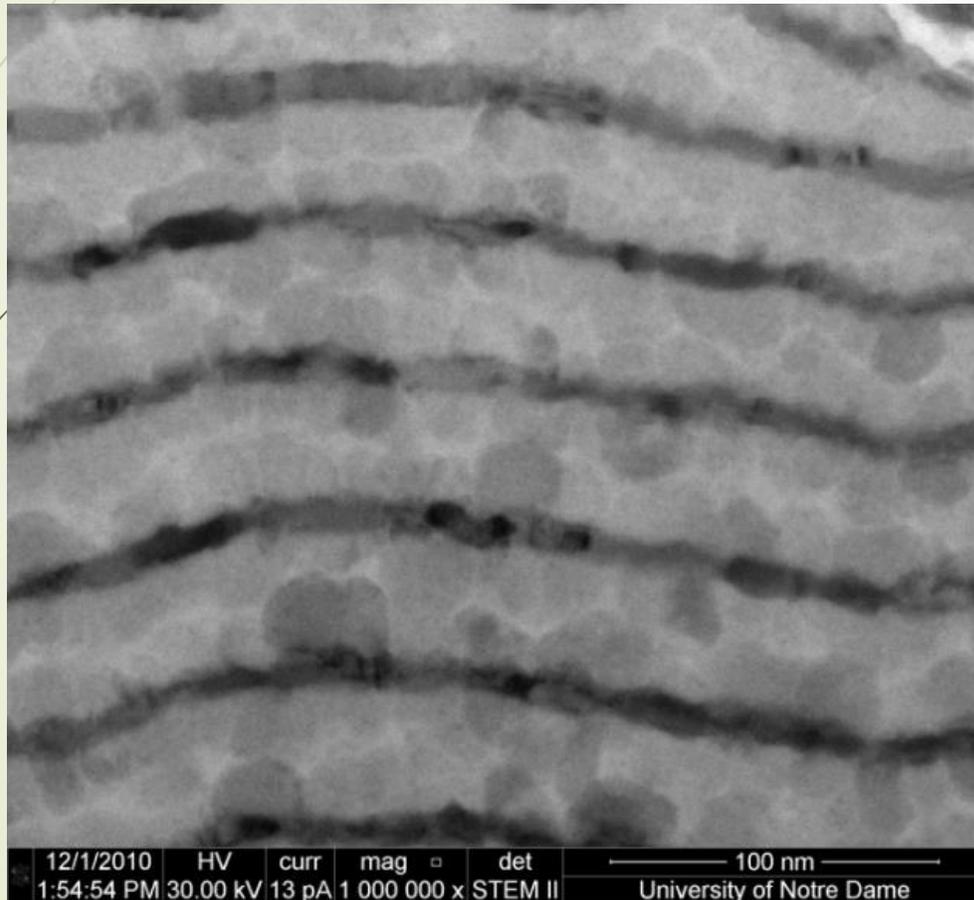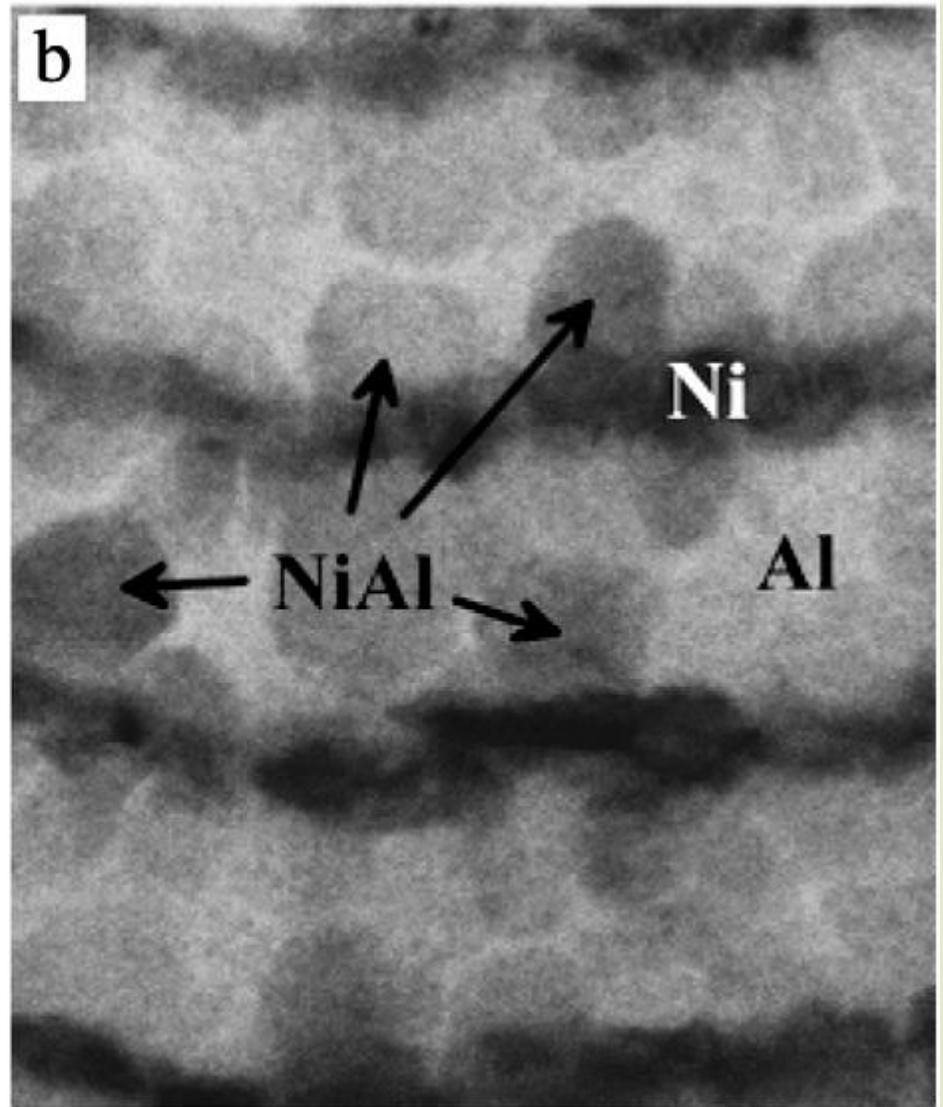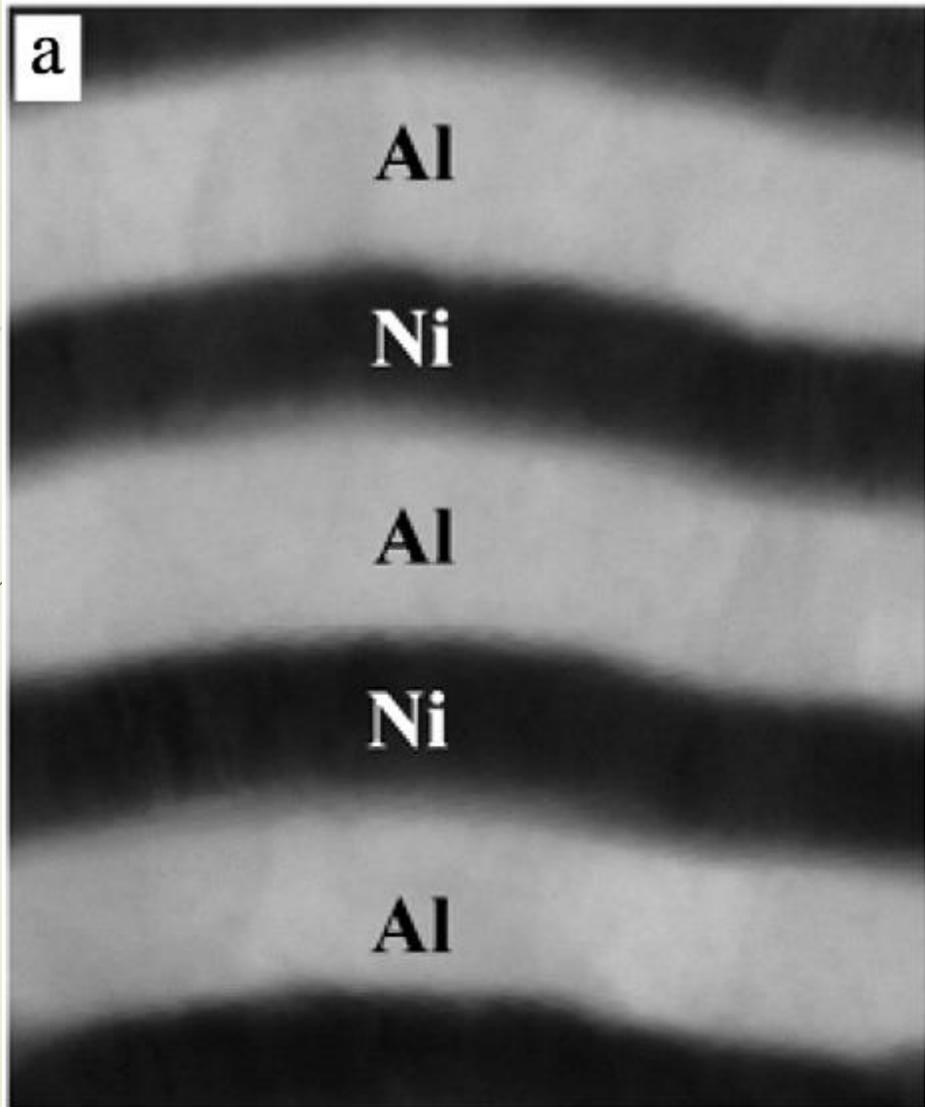
Formation of solid NiAl grains after saturation of the Al-Ni melt

50 nm

Phase Formation in Reaction Front
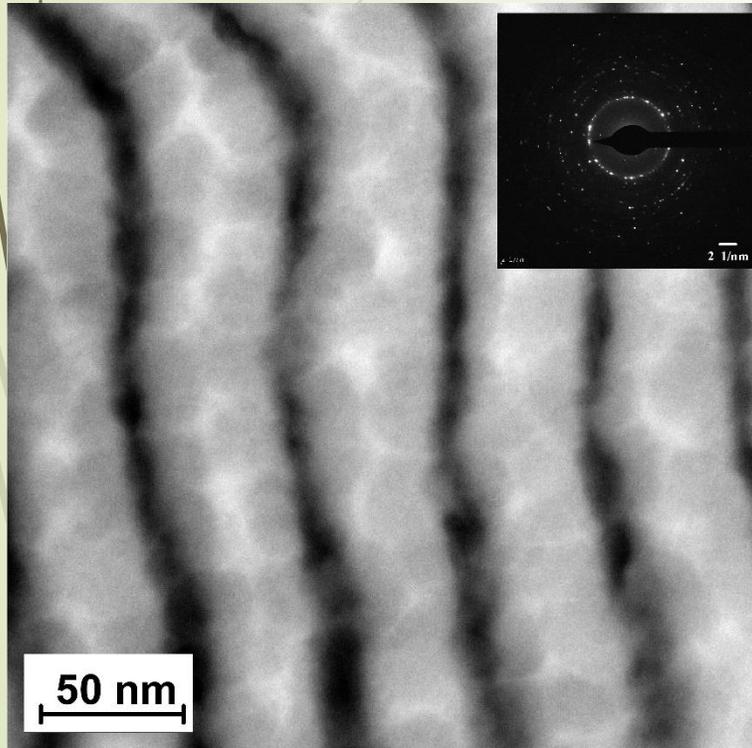
d _exp_ (A)	d _database_ (A)			
	Ni	Al	AlNi	Al3Ni
4.02				4.03 <011>
2.87			2.86 <100>	
2.03	2.03 <111>	2.03 <200>	2.03 <110>	2.01 <022>
1.80	1.76 <200>			1.82 <231>
1.64			1.65 <111>	1.66 <400>
1.43		1.43 <220>	1.43 <200>	1.43 <142>
1.35				1.35 <250>
1.26	1.25 <220>	1.22 <311>	1.28 <210>	1.26 <511>
1.17		1.17 <222>		1.17 <114>
1.08	1.06 <311>			1.08 <053>
1.02	1.02 <222>	1.01 <400>		1.02 <503>

EDS Analyses
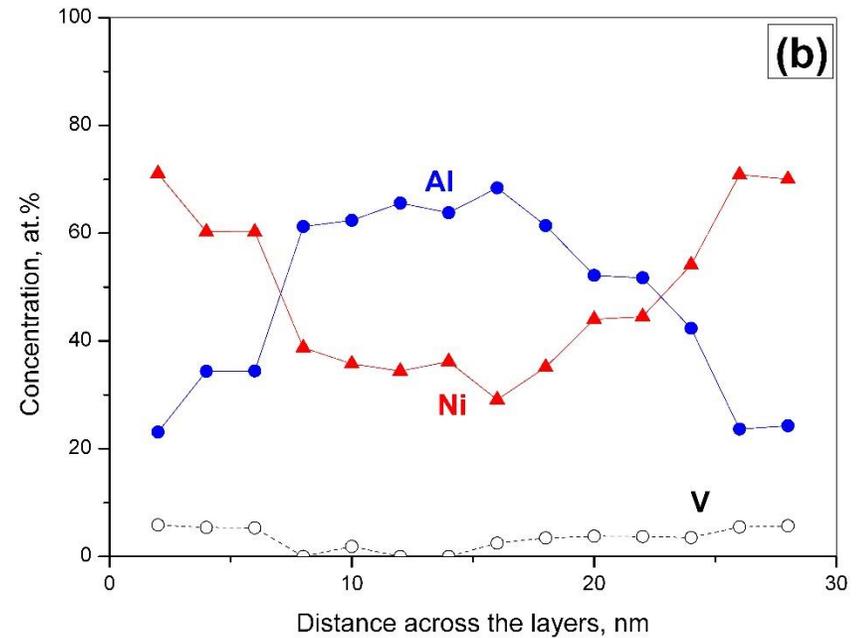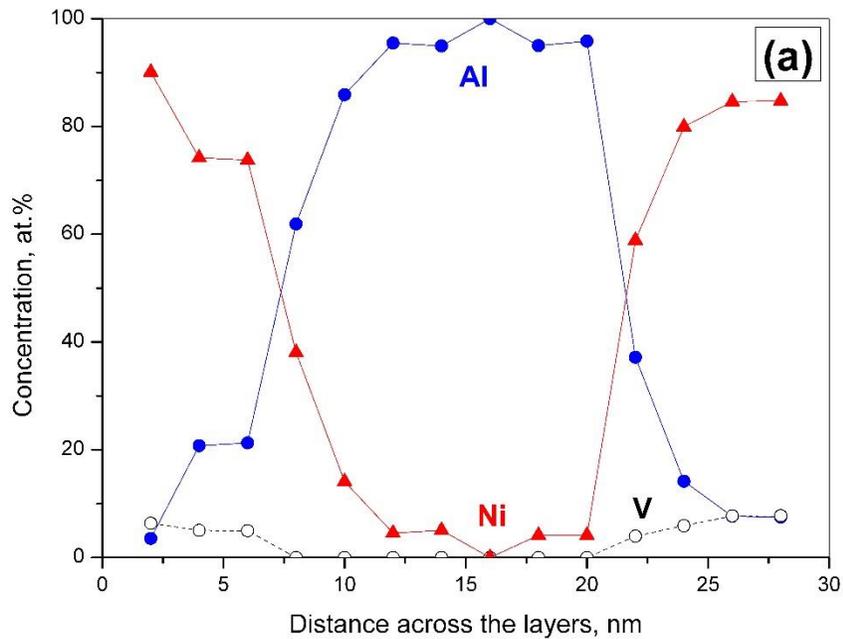
Concentration profiles across the layers in the initial foil (a) and reaction zone (b) in the sample with quenched reaction wave (TEM+EDS at thin cross-sections).

General Reaction Scheme

(A) Photo of the exothermic wave (A);

(B) SEM picture of the quenched reaction front;

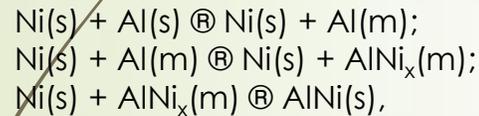(C) TEM pictures of the reaction zone;

(D) dynamics of the thickness of Ni layer (Δ),

Al-rich layer (□), and bilayer period (○),

I-initial structure,

II – Ni dissolution,

III – NiAl precipitation;

(E) schematic draw of the wave structure.

The sequence of the process stages can be represented as follows:

Ni(s) + Al(s) ® Ni(s) + Al(m);

Ni(s) + Al(m) ® Ni(s) + AlNi$_x$(m);

Ni(s) + AlNi$_x$(m) ® AlNi(s),

At the first stage, the aluminum melts just above the reaction front due to heat flow from the reaction zone.

Melting of Al initiates partial dissolution of the Ni layers (stage 2) into Al melt until the saturation concentration (x ~ 0,25 - 0,3) is reached.

At the third stage, solid grains of the NiAl B2-phase nucleate and grow.

The characteristic feature of the process is that the grains have different crystallographic orientations and are separated by liquid/amorphous interlayers. The existence of the liquid between the solid grains allows Ni to dissolve into the melt by means of liquid-state diffusion, which is much faster than solid state diffusion across the intermetallic solid compound.