Foams and Aerogels Materialized, Verified and Being Developed in LPI

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ABSTRACT

LPI proposes the foams of different chemical systems depending on the necessities of laser experiment.

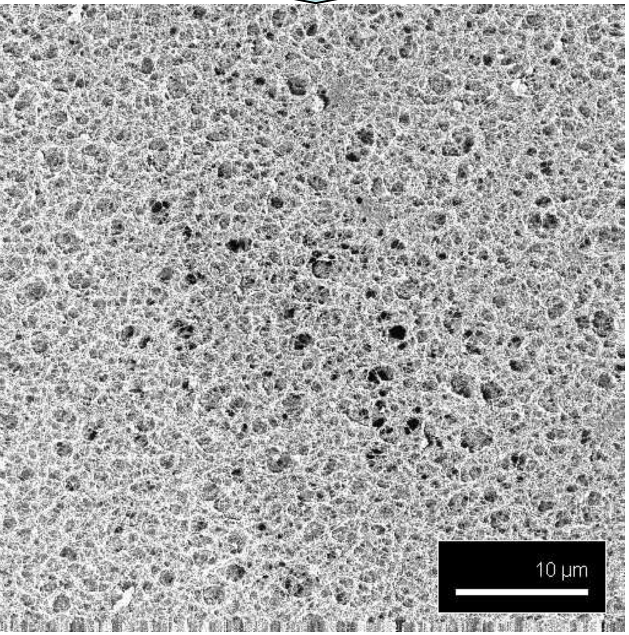
Begun in cooperation with Institute of elemental-organical chemistry and conducted together with Zelinskiy chem.-phys. institute (both belonging to RAS) during the recent years the works with cellulose triacetate resulted in undercritical plastic aerogels used in experiments on different laser facilities for 3ω, 2 ω , and 1ω laser light.

In contrast, the higher densities of foams are fabricated from other chemical materials: deuterated polyetilene, RF resin, beryllium deuteride and others.

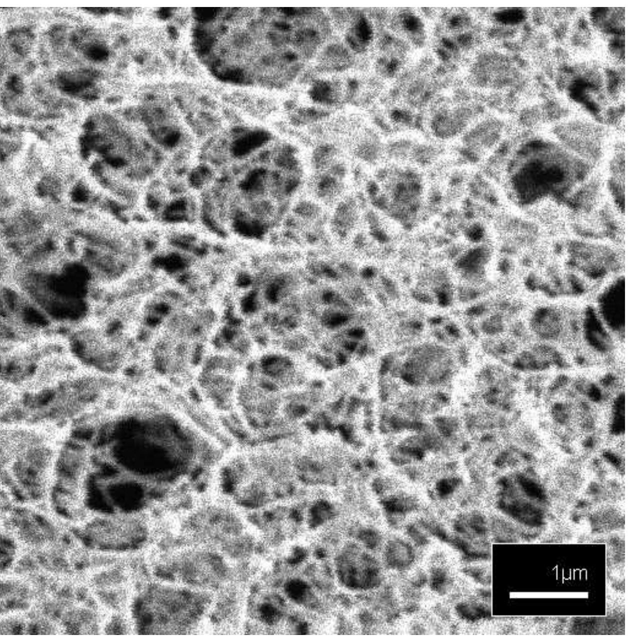
The targets produced in Russia with declared parameters similar to those produced in Scotland by Wigen Nazarov from differing polymers and independent technology routes proved to possess similar structure from the point of view of laser light interacting with them. From time to time such targets met in the one and same laser experiments on PALS, LULI or LIL facilities and produced results supporting each other and thus verifying the measured phenomena. Our foam components also participated in the shots of PHELIX and UNILAC in Germany and KANAL, ISKRA 5, LUCH, MISHEN and NEODIM in Russia.

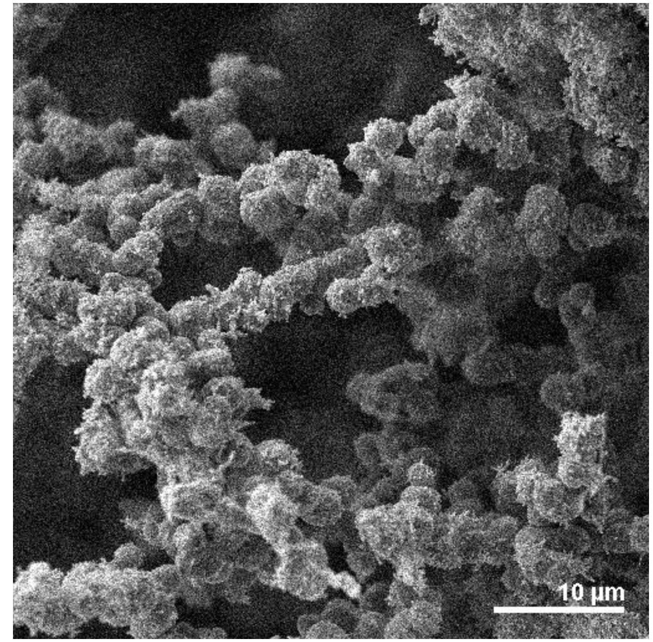
The targets with density gradient are now under development in LPI. These are continuously grown in silicagel in the course of its synthesis. The gradient formation dynamics is measured by X-ray 3-D tomography right in gel substance. The achieved gradient in supercritically dried samples is reported to be **1 $g \cdot cm^{-3}/cm$**, which is still too low for pressure multiplication in laser shots.

TAC and PVF foams

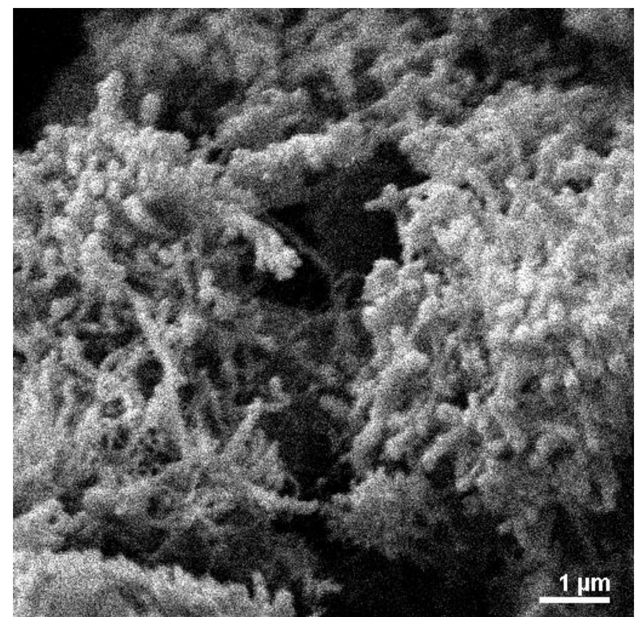


Left: SEM pictures of TAC foams with density 5 mg/cc for heat-and-flow smoothing method

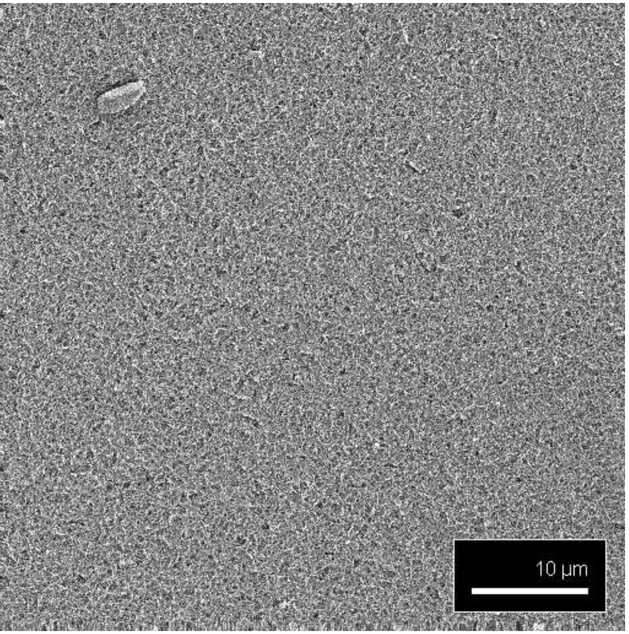




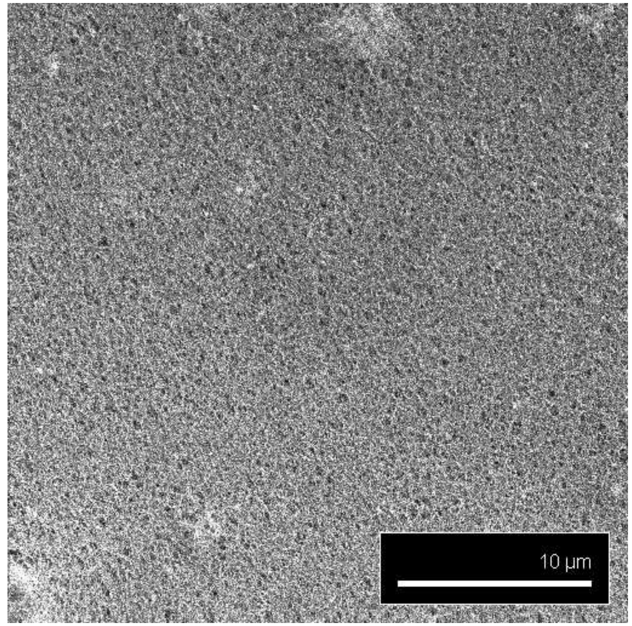
Right: SEM pictures of PVF foams with density 10 mg/cc for dynamic plasma phase plate method



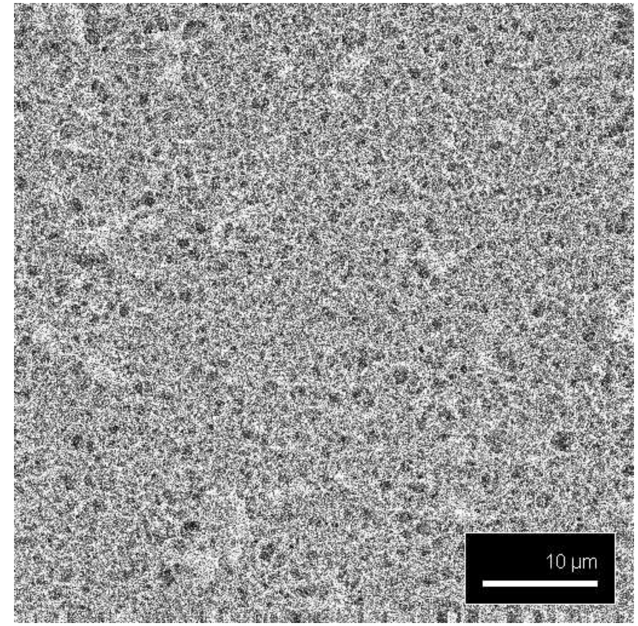
Synthesis of 01/19/2005: Polymer TAC 3D-nets of 10 mg/cc (SEM)



TAC without Cu



TAC with Cu nanoparticles of 10% by weight

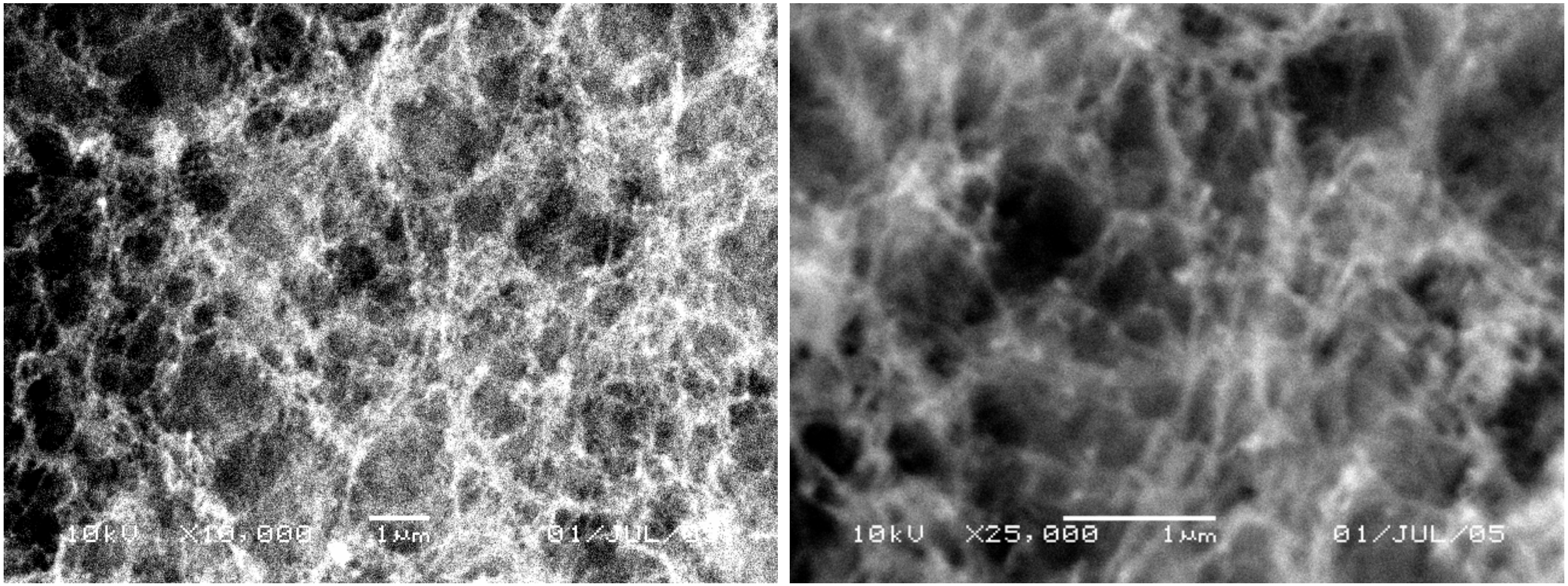


TAC with Cu nanoparticles of 20% by weight

References:

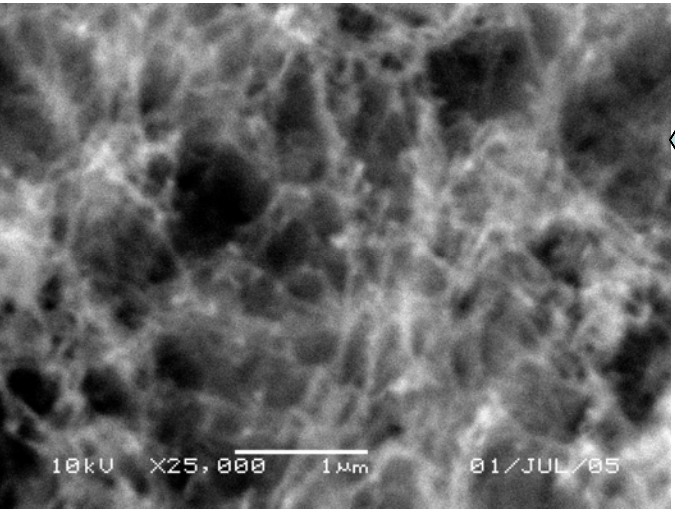
- *A*.M. Khalenkov, N.G. Borisenko, et al. Experience of microheterogeneous target fabrication to study energy transport in plasma near critical density. // Laser & Particle Beams, 2006, Vol. 24, pp. 283-290.
- N.G. Borisenko, et al. Regular 3-D networks with clusters for controlled energy transport studies in laser plasma near critical density. // Fusion Sciences and Technology, 2006, V. 49, #4, pp. 676-685.
- N.G. Borisenko, et al. Intensive (up to 10^{15} W/cm^2) Laser Light Absorption and Energy Transfer in Subcritical Media with or without High-Z Dopants. AIP Conference Proceedings, 2006, Vol. 849, pp. 242-246.

SEM photos of TAC 3D-networks dependent on coating.



SEM images of TAC 5 mg/cc with carbon 20 nm coating. Scale bar is 1 µm. Gold visualizes rougher structure with larger inter fiber distances, thin details are lost

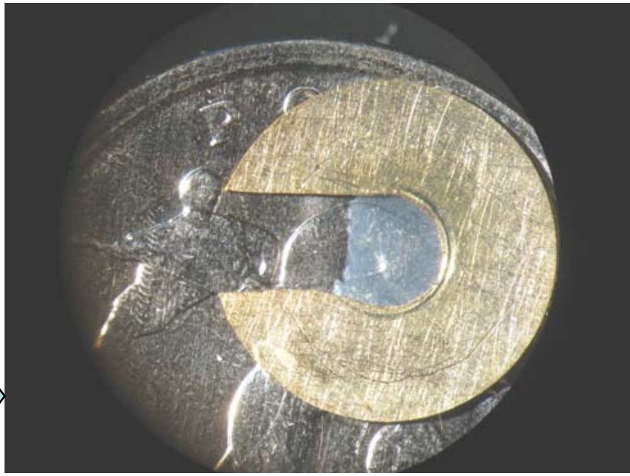
Targets of plastic aerogel TAC for experiments with undercritical plasma density

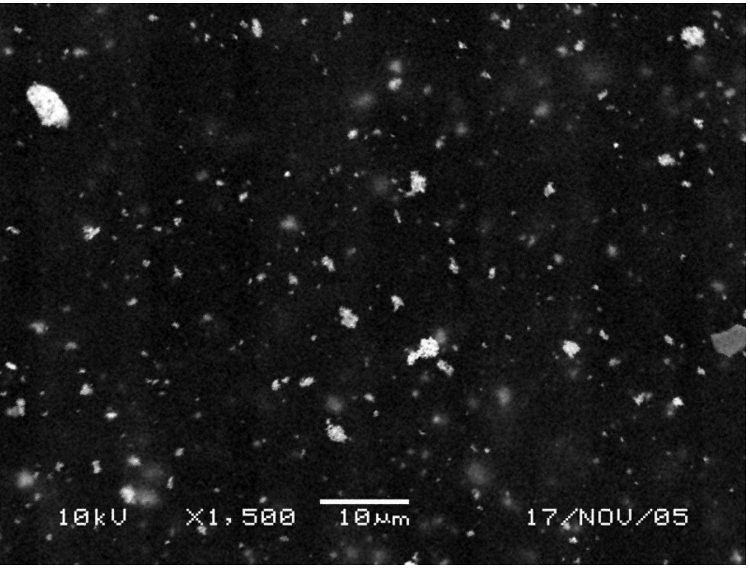


Density fluctuations <1% in the focal area Ø 300 µm

TAC 10 mg/cc

300 µm in the holder with a slit on the kopeik





Similar repeatable submicron structure

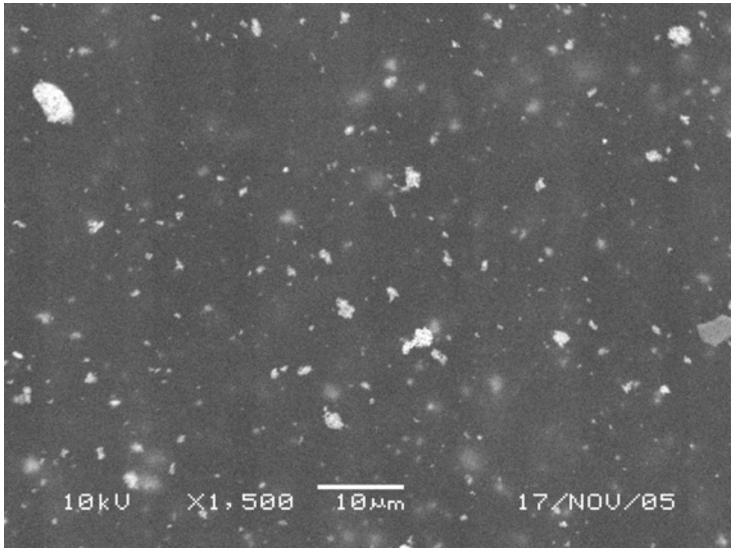
(in the range 50-to-1 mg/cc, inter fiber distance 0.7- 1.6 µm, 30-to-50nm diam., fiber density 0.2 g/cc)

Doping with Cu or Cl (15% mass)

10 mg/cc, Cu particles 40 nm, concentration $5 \cdot 10^{12}$ cm^{-3},

3% nanoparticles agglomerated

Nanoparticles agglomeration and size distribution



TAC, 10 mg/cc, 15 % (wt) Cu,
Lebedev Physical Institute, 2005

At low gel concentration the UDP high-Z admixtures tend to agglomerate. So the additional effort is necessary to achieve uniform heavy particles distribution

Centrifugal separation

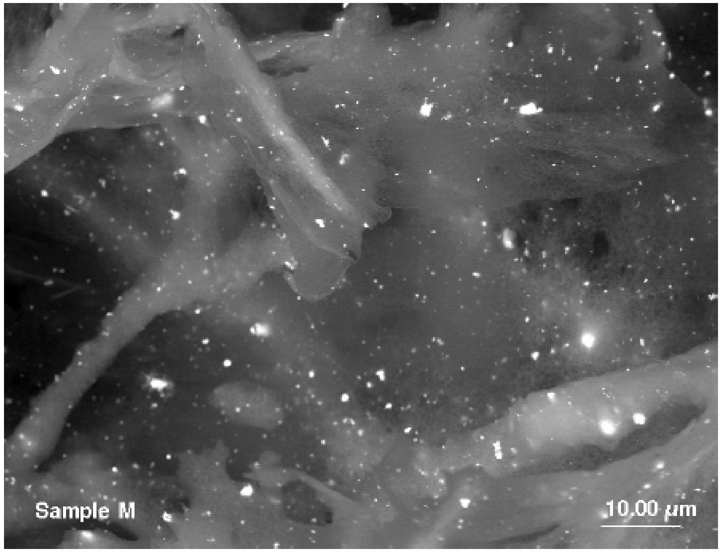
→ Eliminate large particles

Dispersion

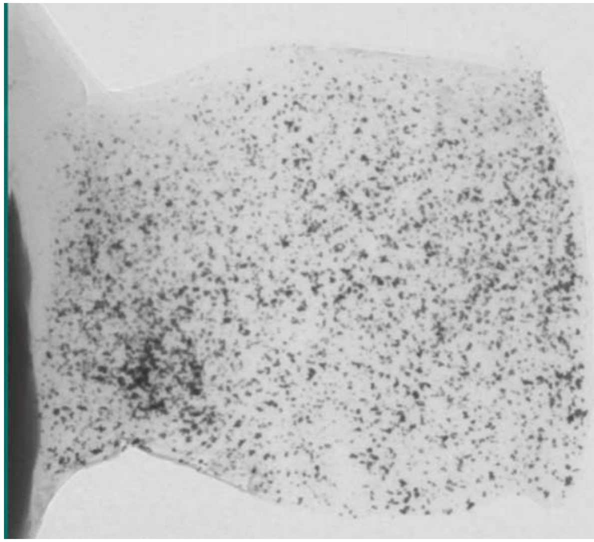
→ High-power ultrasonic dispergators

Appretes

→ UDP particles treated with different hydro-oleophobic chemicals

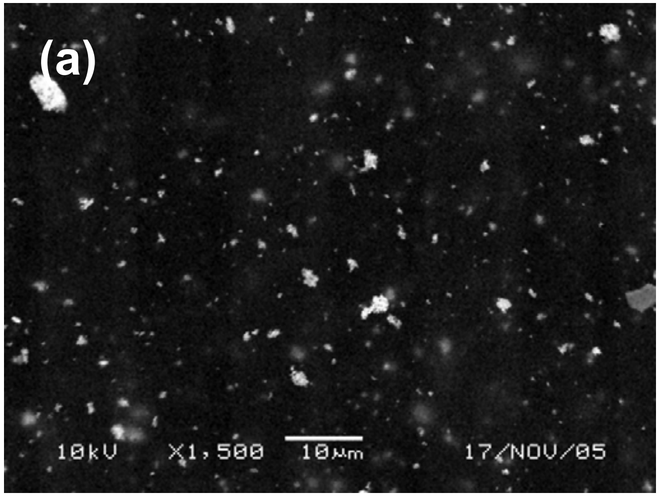


K. Youngblood, (Schafer, Sandia, USA)
15 TFM report, 2003; foam TPX 14 mg/ccm

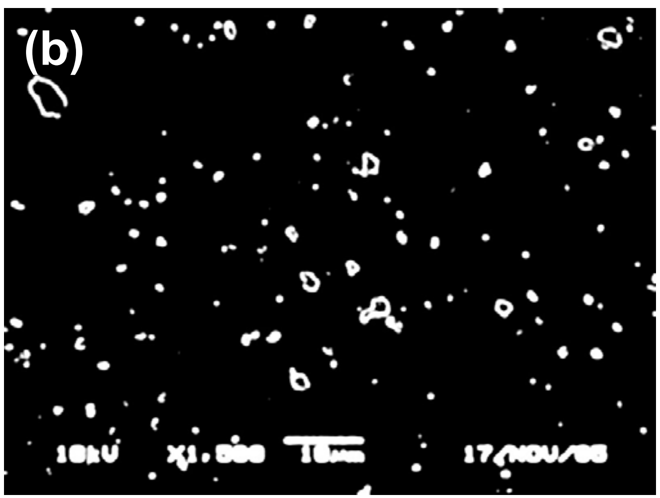


F. Lewis,AWE UK, 15 TFM report, 2003,
Au clusters in foam of 50 mg/cc

Verifying chemical composition and size of nanoparticles



Low density TAC 10 mg/cм^3 with embedded, 15% Cu particles; the image prepared for counting



The same frame processed and counter-ready

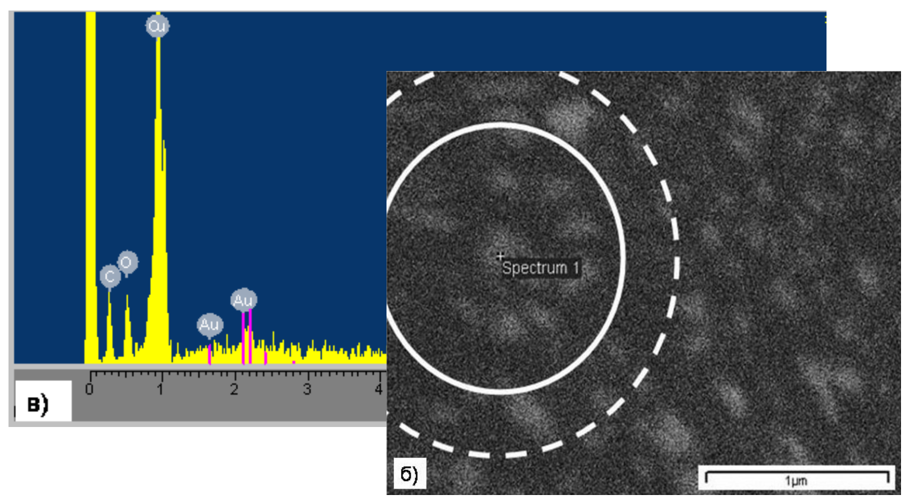
(a)-(b)

Material contrast SEM in low vacuum (~1 Pa):

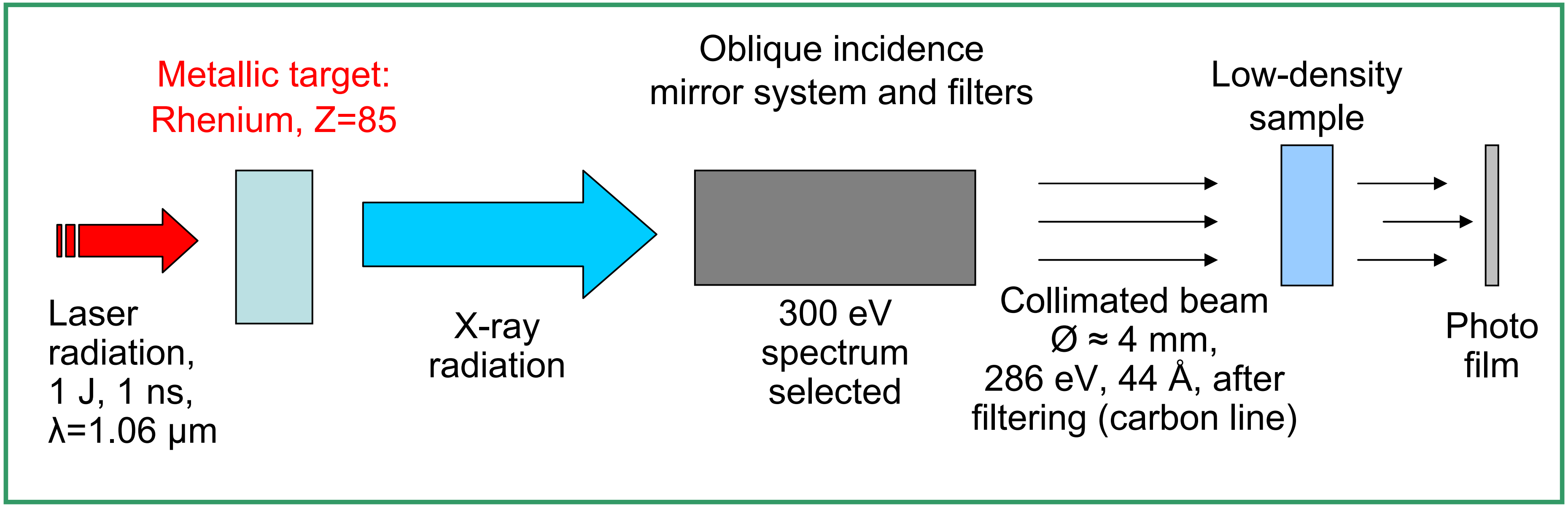
<u>no conducting layer deposited</u>

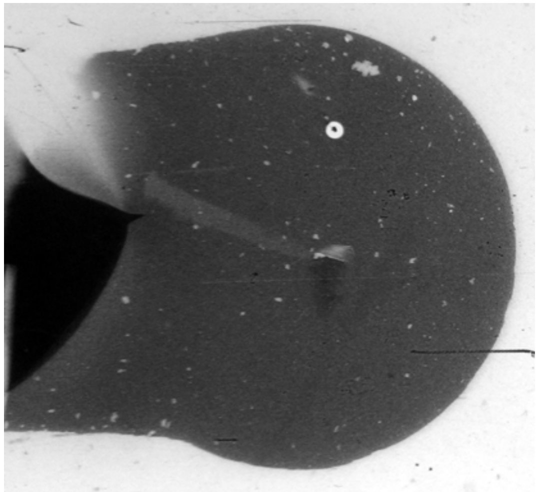
+ software processing for automatic agglomerates counting

(c) *High-Z additive in TAC were analyzed with Au deposition, Cu 50 nм is detected*

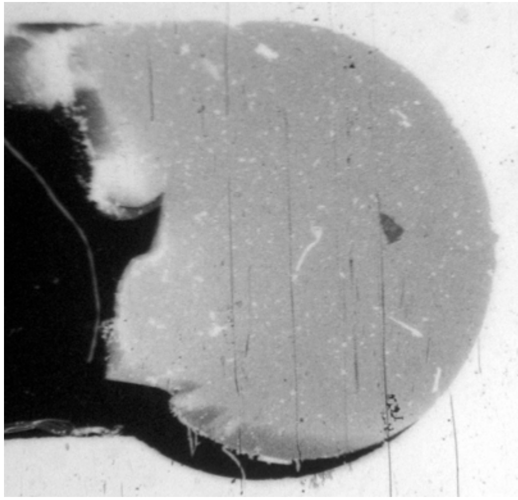


4.4 nm x-ray source for 3D low-density target monitoring

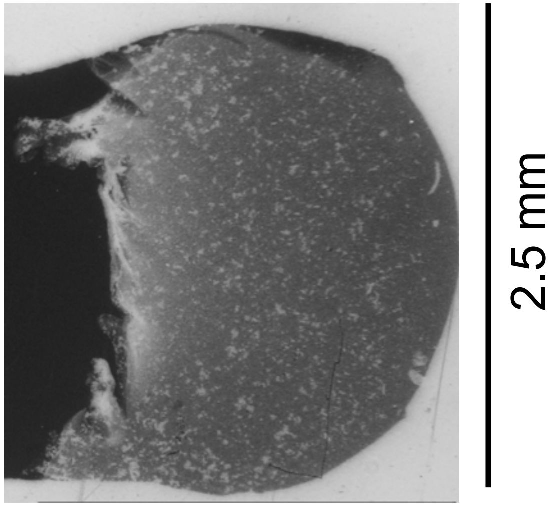




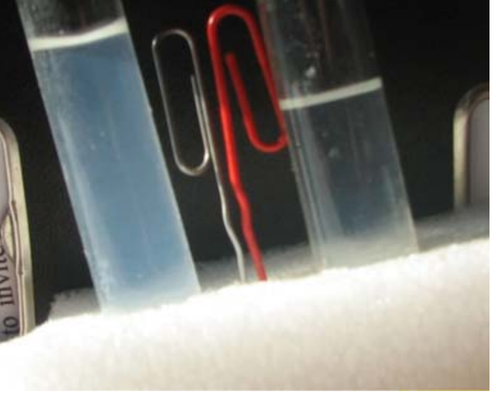
TAC 4.5 mg/cc



TAC10 mg/cc, 10% Cu
300 μm thick layers



TAC 10 mg/cc, 20% Cu



Uniform silicagels

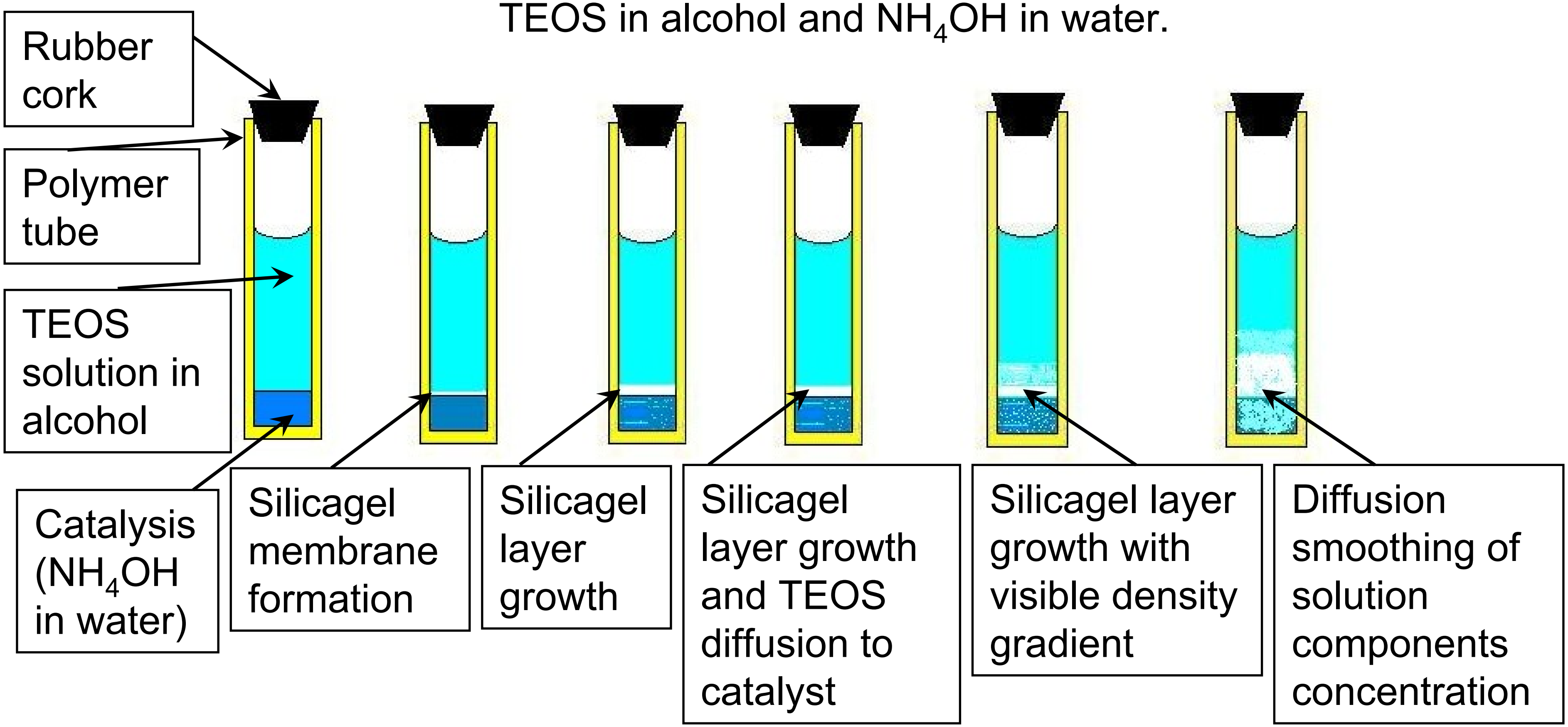
Flat layer of growing silicagel from catalyst boundary

- We try to work out the novel fabrication technology for the density-gradient targets.
- Method of density-gradient gel formation is based on the 1D catalyst diffusion into gel growing from the boundary of polymer solution with frozen catalyst.
- The alcohol-soluble silicate monomer $Si(OC_2H_5)_4$ - **tetraethylorthosilicate** (TEOS) - was polymerized to create branched polymers or dense colloidal particles at hydrolysis of TEOS with catalyst - 20% NH_4OH-in-water solution).
- There could be a doubt about the density variations in the gel via height. One can suspect the constant density that is accompanied by differing structure to give the visible optical density change. The direct proof could be the X-ray images of the realized density gradient in the layer.

Ref: N.G. Borisenko, A.A. Akunets, Yu. A. Merkuliev. Polymer aerogel with density gradient for ICF targets. // Book of Abstracts. 3rd Moscow Workshop on Targets and Applications, 15-19 October, 2007, Moscow, Russia, p. 52

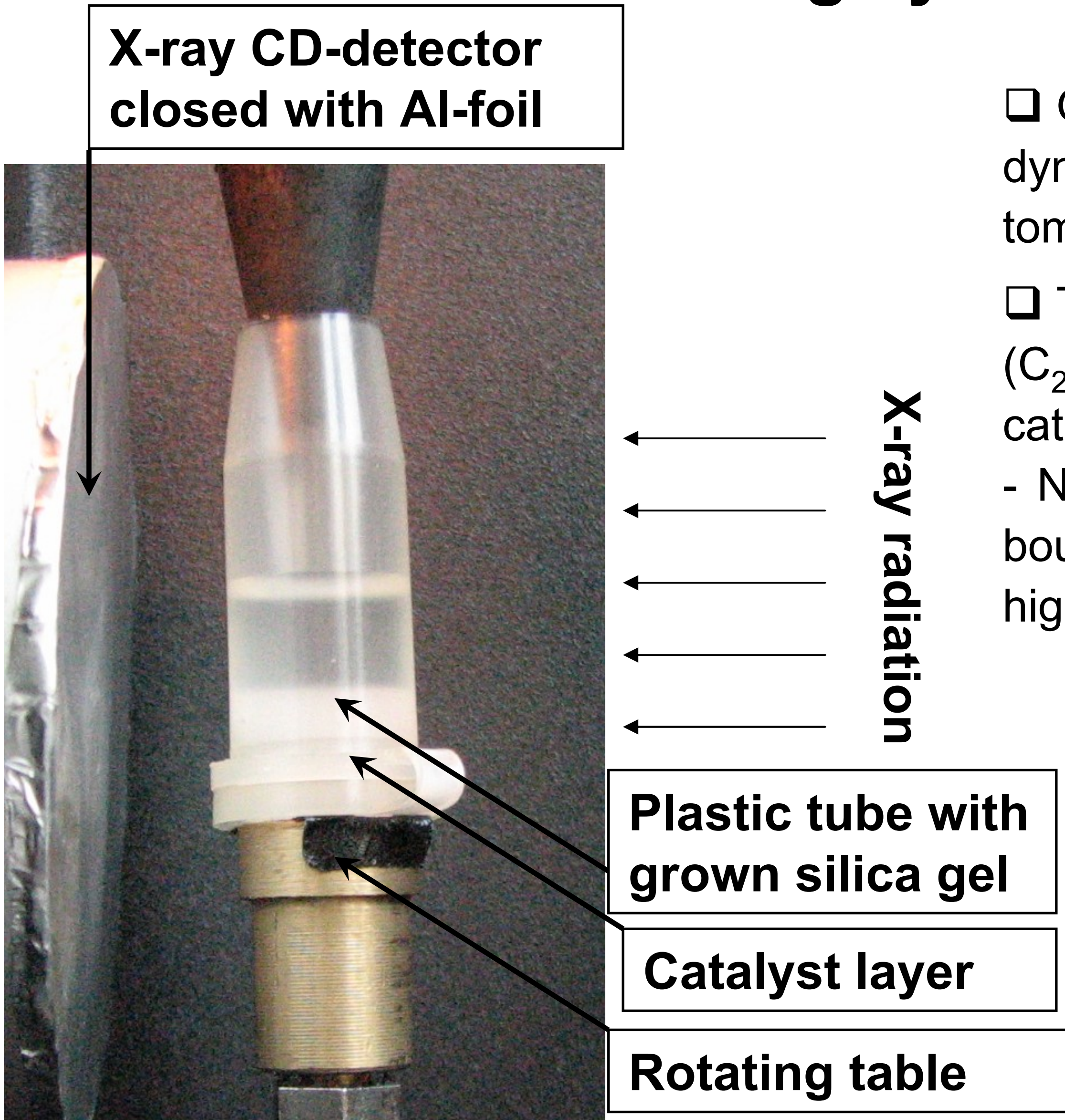
Growth scheme for gel with density gradient

We first organize a flat surface between the two solutions:
TEOS in alcohol and NH_4OH in water.



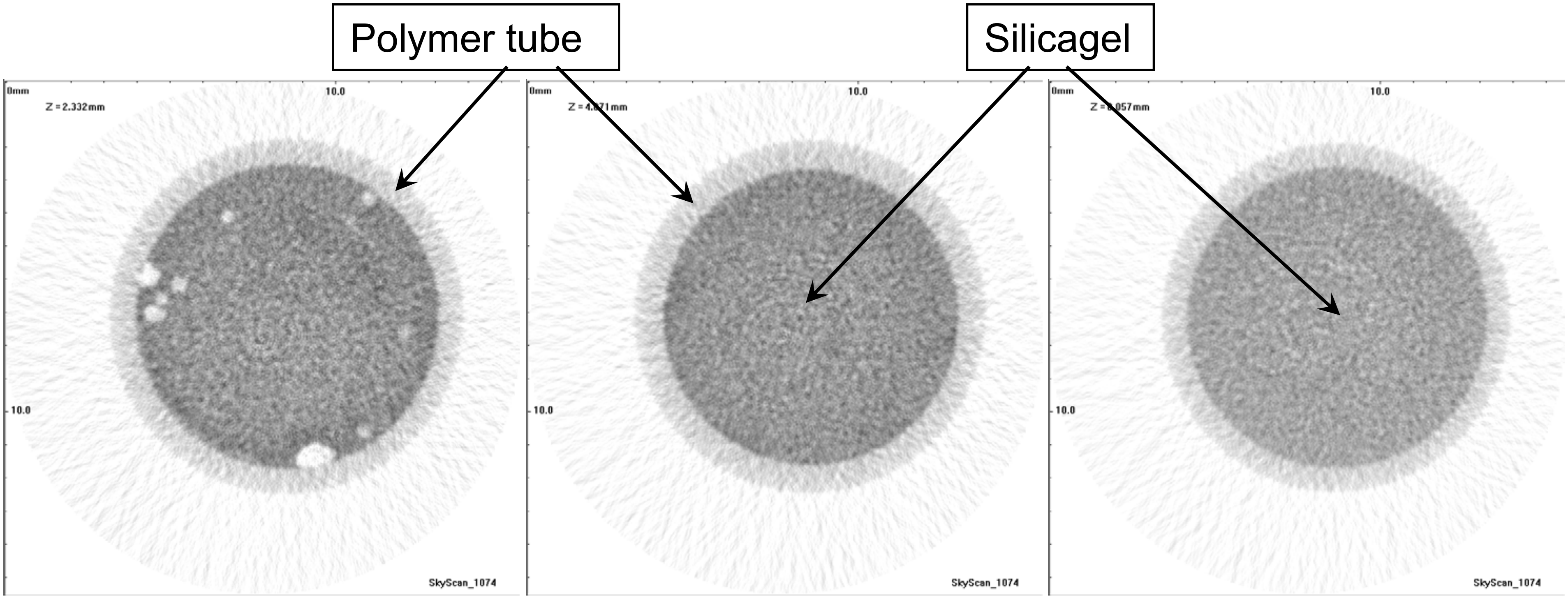
Silicagel starts growing intensively for several minutes thus forming a solid white membrane on the boundary between the two solutions. Soon the growth rate diminishes, the formed gel layer becoming more and more transparent with the height. The white color means a relatively coarse structure is formed to scatter the light diffusively.

Apparatus for characterization of gel growth during synthesis



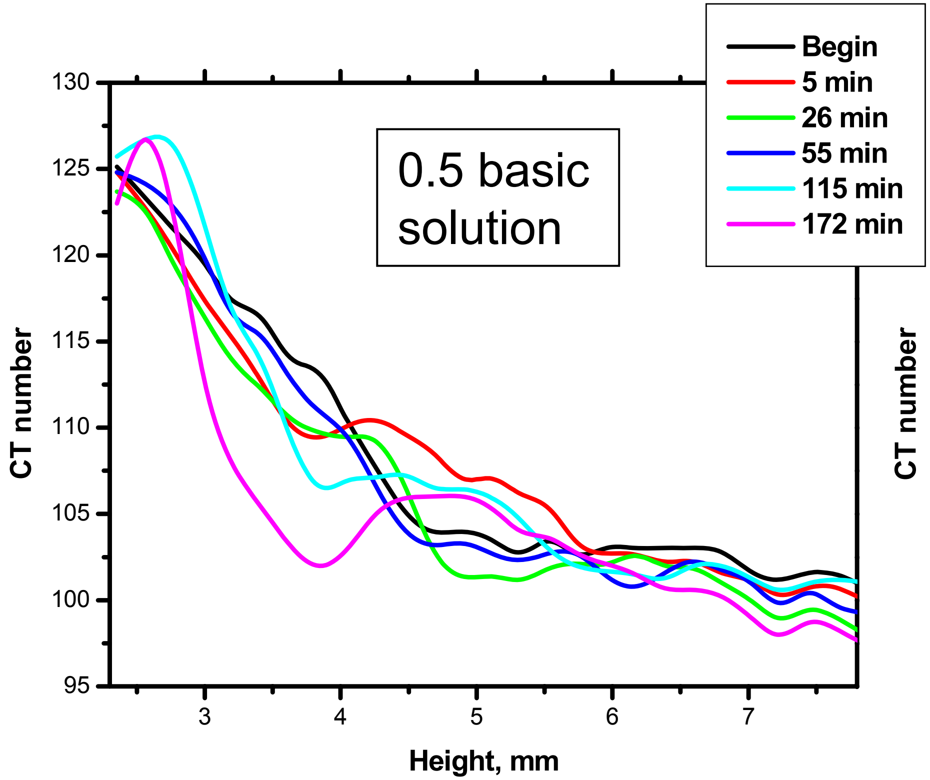
- Gradient density gel formation dynamics is studied by 3D X-ray tomography images.
- Tetraethylorthosilicate (TEOS) – $(C_2H_5O)_4Si$ - solution in alcohol with catalyst (20% ammonium hydroxide - NH_4OH - in water) on the contact boundary are exposed to create high X-ray contrast images.

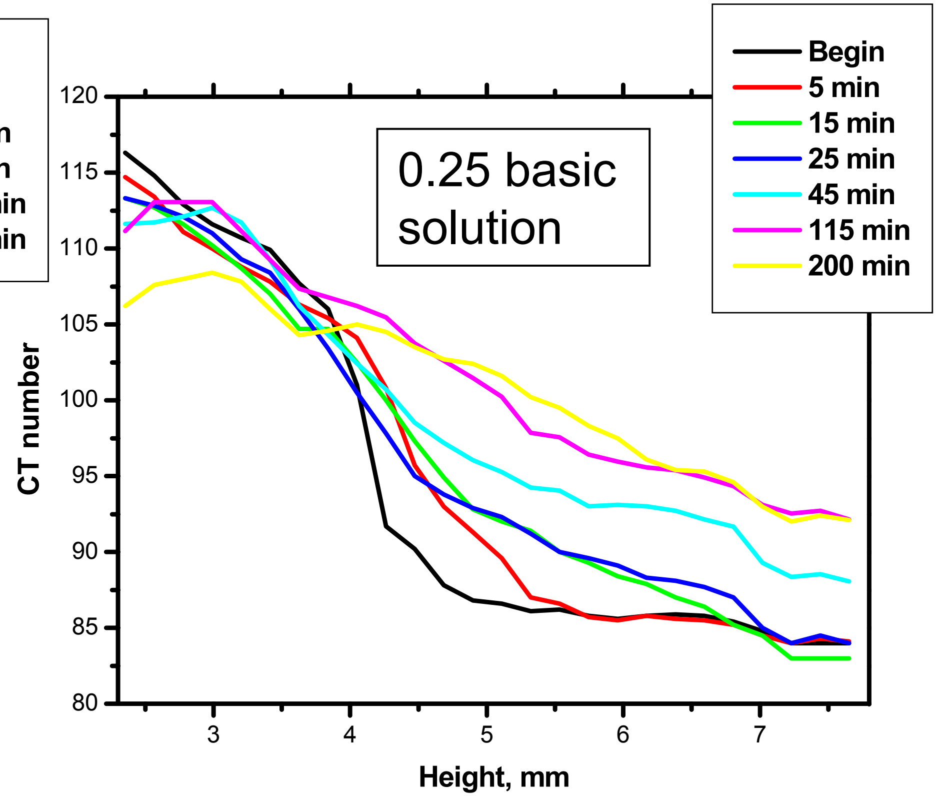
Horizontal cross section of silica gel with density gradient



- The pictures show horizontal cross sections silica gel during gel growth (from left to right) on the levels of 2.33 mm, 4.07 mm and 8.06 mm
- TEOS solution boundary level for different samples changes from 2.5 mm to 3.3 mm.
- This sample boundary level is 2.7 mm from the bottom, left cross section has gas bubbles under silica gel membrane.
- Lower cross section has darker color, then upper cross section and gel has higher SiO_2 concentration
- Middle and right cross sections have identical color. **Gel growth front is stable (flat)**

Dependence of SiO_2 concentration via height





Dependences of count tomography (CT) numbers via height of TEOS solution (tube) during gel growth at various time after TEOS solution is placed on catalyst for two start concentrations (left – 0.5 basic solution, right – 0.25 basic solution).

SiO_2 concentration can be found from comparison of CT numbers with calibrating SiO_2 concentration.

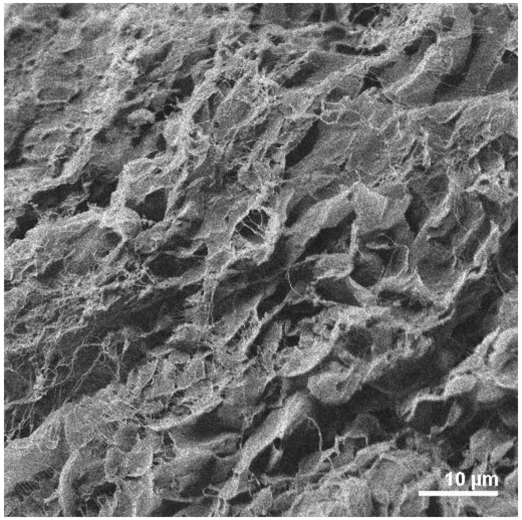
The resultant gradient is **1g/cc per 1cm height** on the left and half of that on the right variant

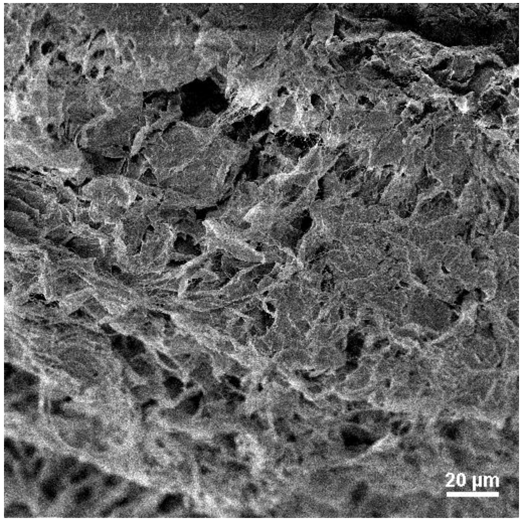
Measuring gradient while gel growth means 1000-fold quicker found technology regime

The X-ray images of acceptable contrast by Skyscan X-ray tomography revealed:

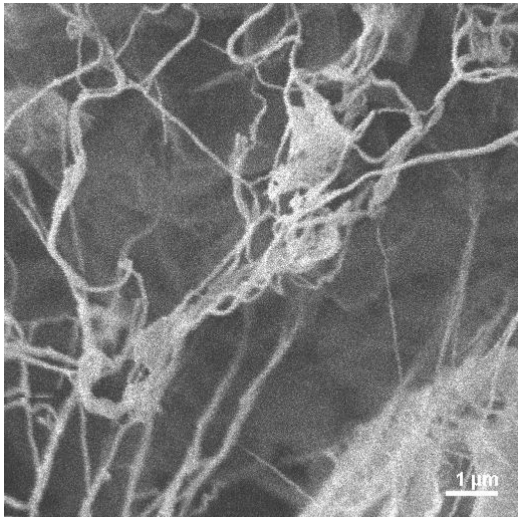
1. Layers of silicagel having density gradient are really produced.
2. Density profiles are measured and density gradients estimated in experiments at different TEOS concentrations.
3. Gel growth front is proved to be plane with the fabrication route chosen. Thus silicagel growth is stable.
4. The obtained density gradients of about 1g/cc per 1 cm length are an order of magnitude less compared to what is necessary for EOS study (10g/cc per 1 cm length).
5. The processes producing defects (such as gas bubbles, discontinuity inside the gel layer) are found.
6. The phenomena were observed that were not taken into account while gel growth modeling. Thus gel restructuring after long time could be connected with silica molecules transport in presence of water and catalyst
7. Simulation of density gradient gel growing at catalyst diffusion is possible, but it is necessary to select the diffusion coefficients of solution components and of silica clusters. Only large massive of such data can give the correct diffusion coefficients and allow quantitative simulations of the gel formation for comparison with experimental results.

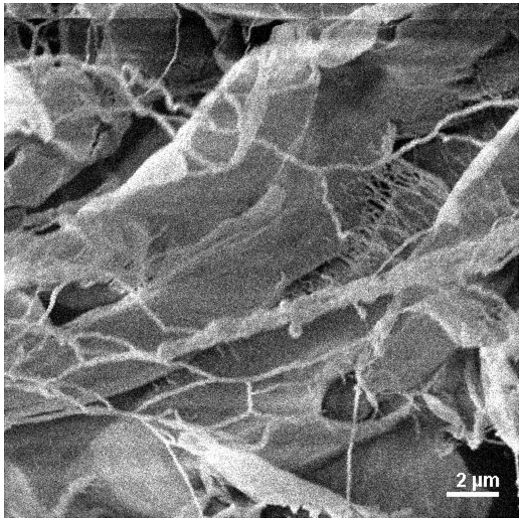
Deuterated polyethylene foams



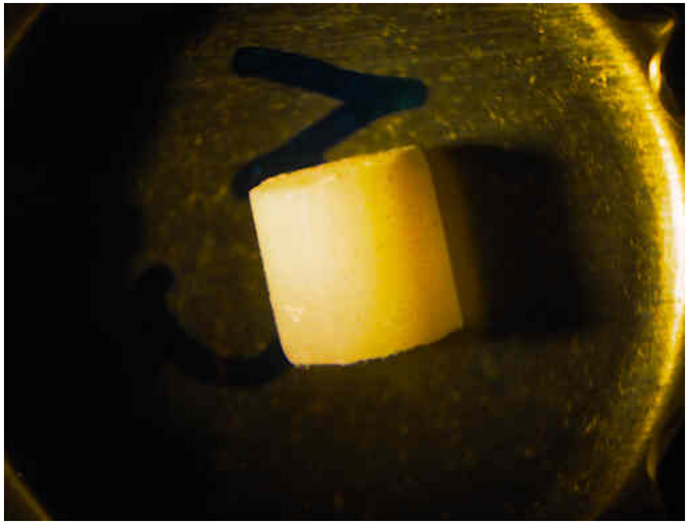


$(CD_2)_n$ foams, density 20 mg/cm^3 and 40 mg/cm^3





The targets for experimental demonstration of increasing of neutron yield when using the foam targets with cells 30-50 µm



Photography of $(CD_2)_n$ foams with 10 mg/cc density for ps-laser targets and liners of Z-pinch.

1. Belyaev V.S., et al. Composition, Density and Structure Dependent Neutron Yields from Deuterated Targets in High-Intensity Laser Shot. // Journal de Physique IV (France), June 2006, Vol. 133, pp. 507-509.
2. Belyaev V.S., et al. High-Intensity Laser Pulse Interaction with Solid of Variable Density. AIP Conference Proceedings, 2006, Vol. 849, pp. 237-241.

Verification experiments

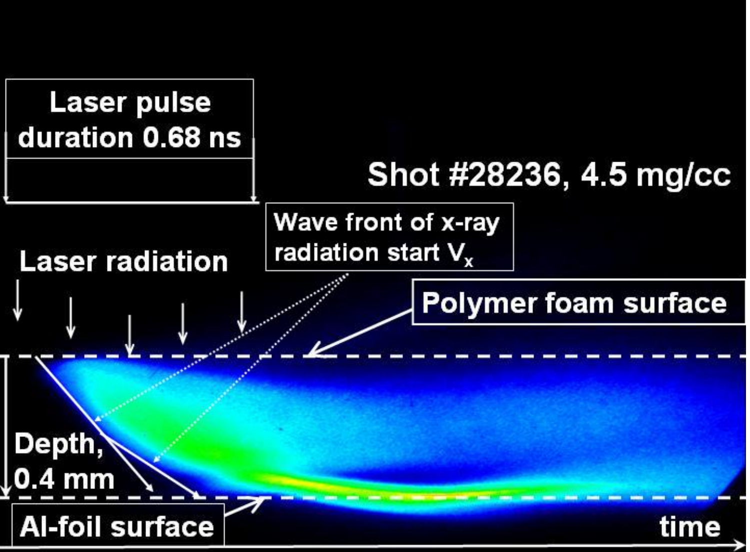
- Similar plastic aerogels (regular open-cell foams, 3-D networks) of TMPTA (Nazarov) & TAC (Borisenko) used to smooth the radiation nonuniformities from different powerful lasers perform other physical processes in microheterogeneous plasma as well, which are essential for the energy transfer models.
- Large dynamic range of registration characterized both optical and X-ray streak-camera images. Their processing shows the weak heating of metal-foil on the rear of the aerogel to appear long before the main heat arrives by thermal conductivity and hydrodynamic waves.
- Measured light transmission through the microheterogeneous plasma and the non-linear optical effects help to explain how the solid foil is heated through the aerogel.
- The analysis of already published experiments on PALS and LIL is done in order to extract the additional information from signals only several-fold higher than the noise.
- The processes important for the energy balance were studied in these experiments:

  1- laser light diffusion through microturbulent plasma in the vicinity of the critical plasma density and light transmittance via target density and thickness;

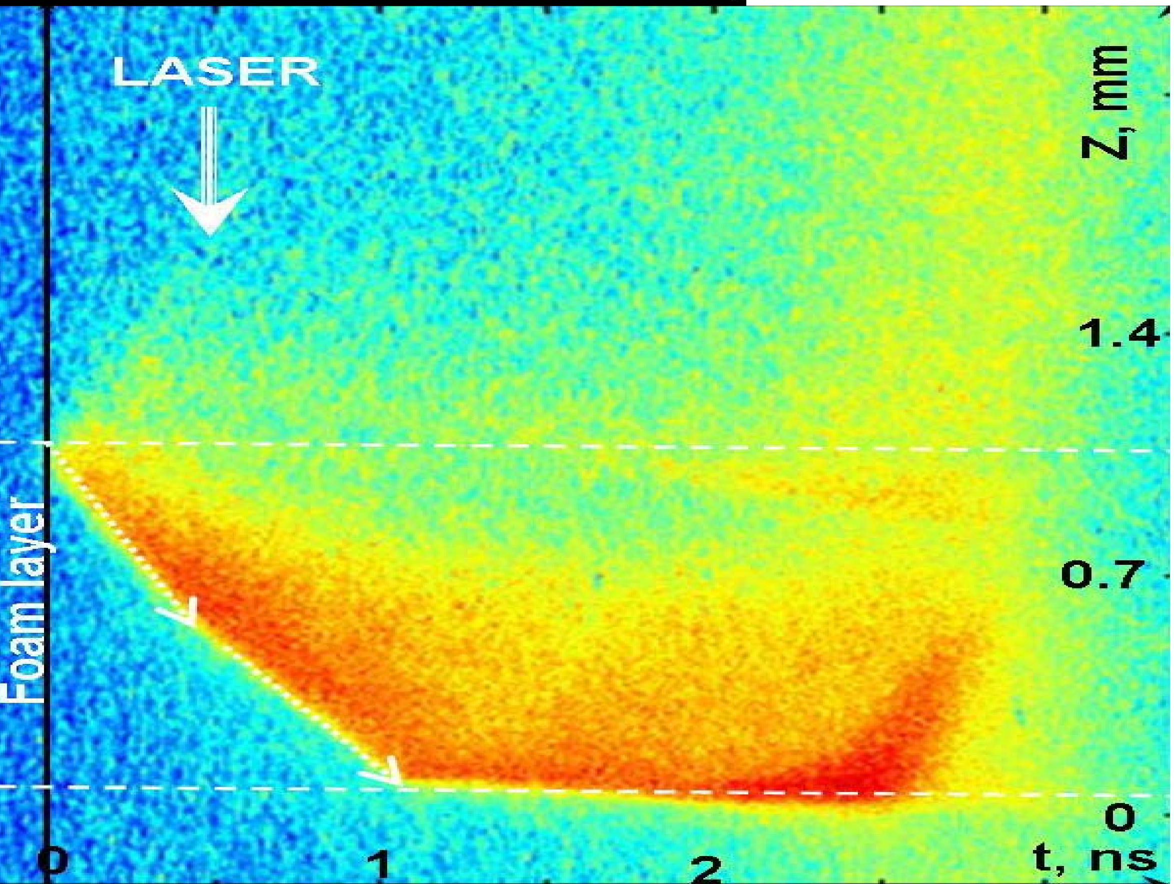
  2 – part of laser pulse energy transferred into SRS, SBS, harmonics and so on (see also oral talks by Fronya TH-0201 and Starodub TH-1001 on ECLIM 2008);

  3 – heating of Al (Cu)-foil (shell) by passed and by converted radiation, which result in the material flux meeting and slowing down the main heat-and-material wave in the low-density matter.

PALS x-ray streak-image in comparison with LIL data



TAC +Al foil (Borisenko)

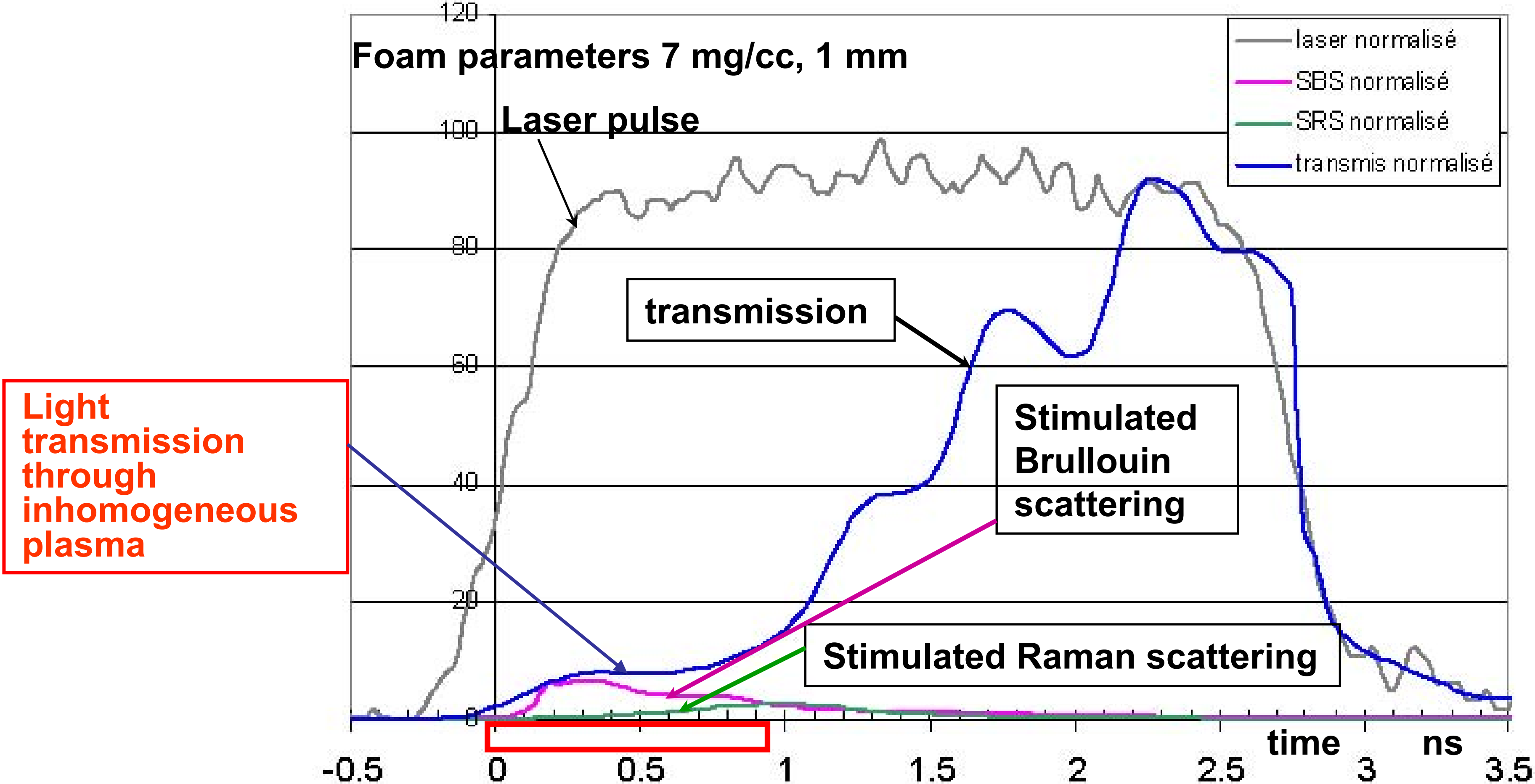


TMPTA +Cu foil (NAZAROV)

both scales match

Similar fluxes give close ionization front velocities

Laser radiation transmission through undercritical aerogel.

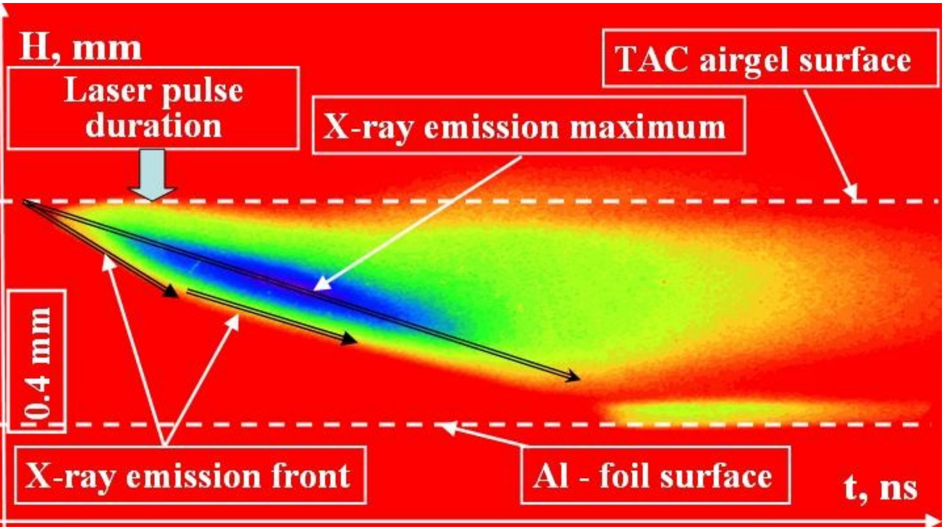


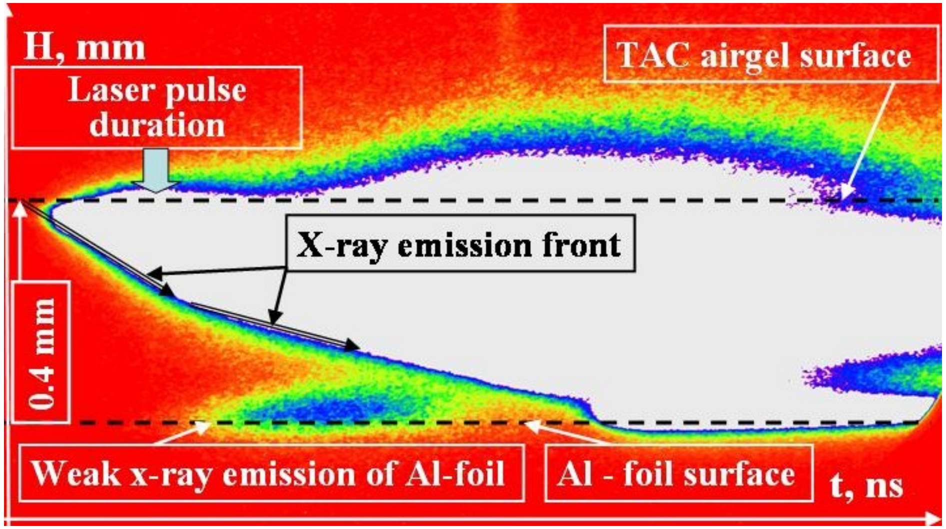
Normalized signals from laser, transmission of laser radiation, stimulated Bruillouin scattering, stimulated Raman scattering. (LIL 2007 experiment)

Laser energy transmitted through the target relative to the energy delivered to the aerogel

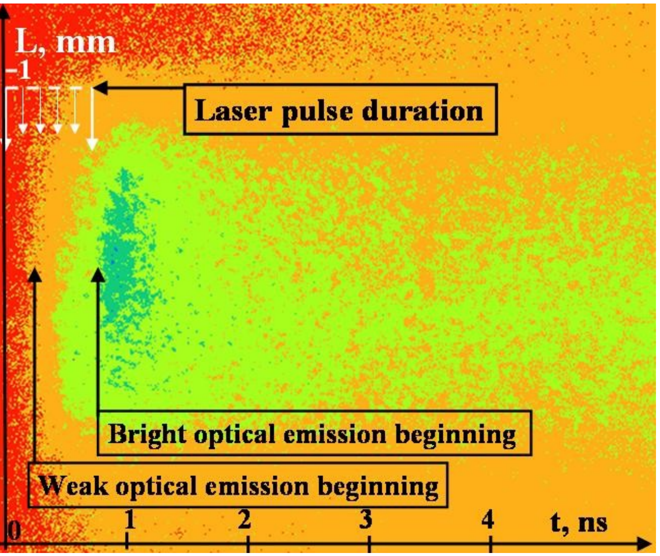
↓ density \ thickness →	400 μm	200 μm	100 μm	Optical loss distance
$(6\text{-}8)\cdot10^{14}$ W/cm^2				
1/2 N_{cr} 9 mg/cc	<0.3%	3,2±0.6 %	15±3,5 %	55 μm
1/4 N_{cr} 4.5 mg/cc	5.5±1%	21±4.5%	50±12%	135 μm
1/8 N_{cr} 2.25 mg/cc	28±5%	55±10%		320 μm
$(3\text{-}4)\cdot10^{14}$ W/cm^2				
1/4 N_{cr} 4.5 mg/cc	5±1%	17±4%	42±9%	120 μm
1/8 N_{cr} 2.25 mg/cc	19±3%	34±5%	67±12%	240 μm

Weak signals of Al-foil emittance prior main heat arrival

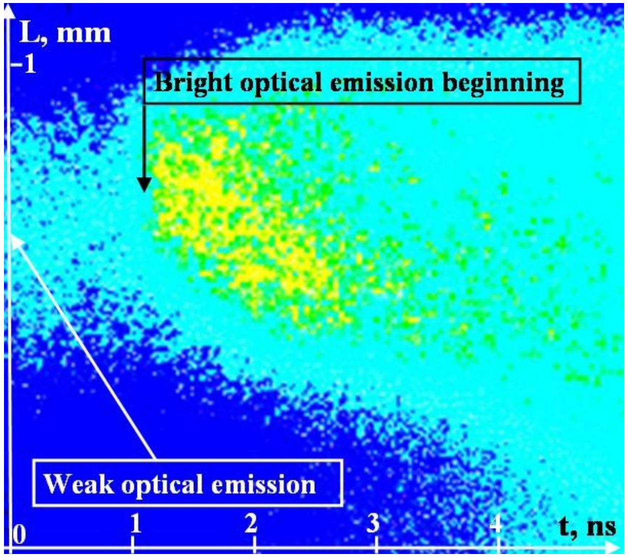




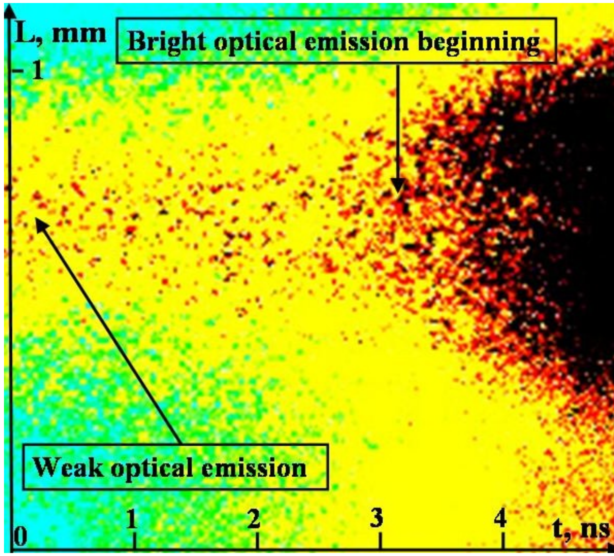
28204, 4.5 mg/cc, 400 μm



28204, 4.5 mg/cc, 400 μm



28232, 9.1 mg/cc, 400 μm



28231, 9.1 (TAC&Cu) mg/cc, 400 μm

Story and results

The experiments at PALS facility in 2005 (third iodine laser harmonic λ= 0.438 μm, 0.17 kJ, τ =0.35 ns) revealed unexpected specifics of the energy transport in plasmas born from the solid targets with initial density corresponding to half and to quarter of critical plasma density. The phenomena of the velocity change during x-ray emission front propagation inside the low-density matter, delay of the shock wave appeared on the Al-foil from the rear side of the foam (1D model of a sphere), and others [1-3] needed theoretical interpretation.

In 2006 for the first time the laser light transmission through the submicron polymer network changing into plasma was measured at PALS facility in details. Partial laser radiation transmission was shown to be essential from the very beginning of target irradiation.

Further in 2006 the experiments at the facility LULI 2000 (up to 2.0 kJ, λ= 0.52 μm, τ=2 ns) and in 2007 the experiments at LIL facility (1 quard, 10 kJ, λ= 0.351 μm τ=2.7 ns) confirmed the deceleration of x-ray front propagation velocity, SRS and SBS input into energy balance. Along with foam smoothing effect the temporal dependence of laser radiation transmission through plasma of the target density equal to quarter of the critical plasma density was also measured.

The experimental data from different lasers witness about the similar nature of the foam effects in spite of the different basic wavelengths and pulse duration.

Small signals are to be taken into account for the sake of future target optimization

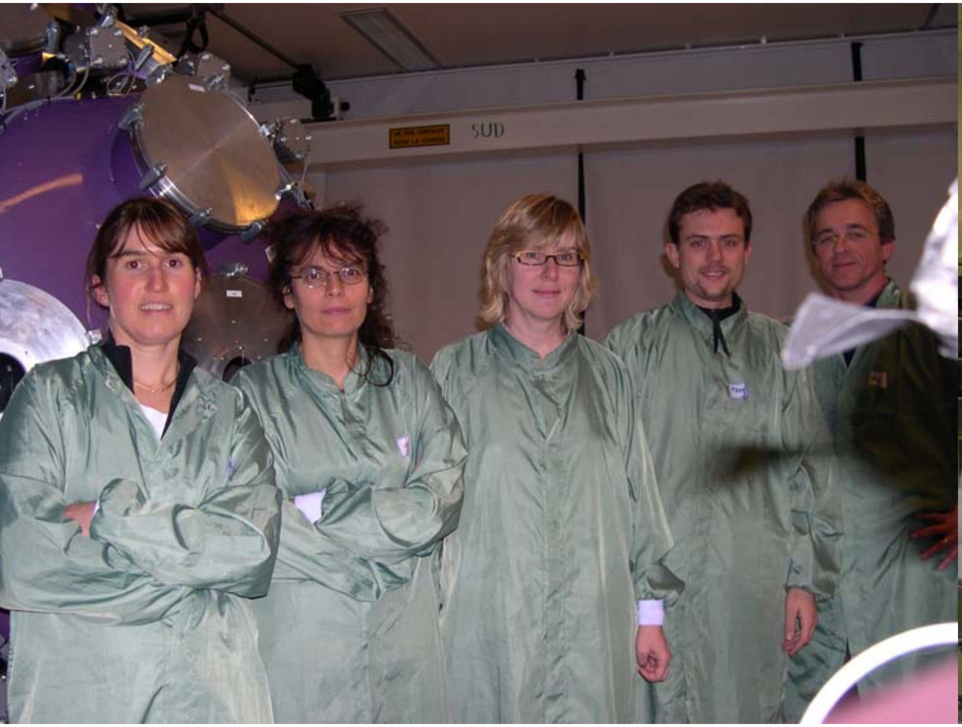
Complex target characterization is demanded!

Acknowledgements

PALS: D. Klir, V. Kmetik, E. Krousky, J. Limpouch, K. Masek, M. Pfeifer, J. Ullschmied et al.

LULI: S. Depierreux, C. Labaune, D.T. Michel, C. Stenz, V. Tassin, et al.

LIL: S. Depierreux, C. Labaune, D.T. Michel, C. Stenz, M. Grech, S Huller, P. Nicolai, D. Pesme, W. Rozmus, C. Meyer, P. Di-Nicola, R. Wrobel, E. Alozy, P. Romary, G. Thiell, G. Soullie, C. Reverdin, B. Villette.



We also acknowledge the partial support of RFBR (#06-02-17526 and # 07-02-01148), LULI, LIL and PALS staffs, Lidia Borisenko technical assistance