UV radiation induced processes in AlN and its potential application for solid state dosimetry

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Layout

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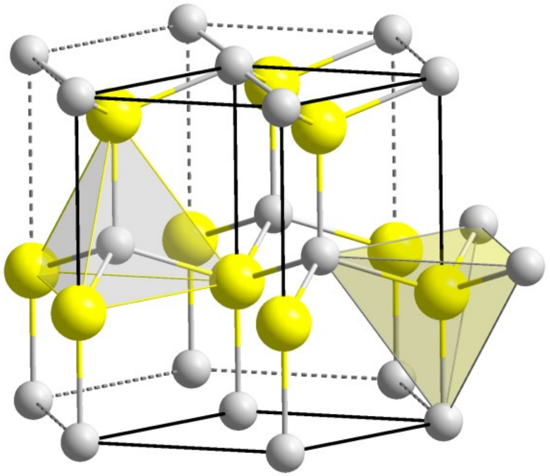
- 2. Experimental details. AlN samples. Methods of photoluminescence (PL), thermoluminescence (TL) and optically stimulated luminescence (OSL). Equipment.
- 3. AlN ceramics. PL at RT. AlN ceramics as a potential dosemeter of ionising radiation. AlN ceramics as a potential dosemeter of UV light. Use of 480 nm emission band for UV induced TL and OSL.
- 4. AlN nanostructures. PL. TL. OSL. Summary of main features.
- 5. Conclusions. Estimation of AlN applicability for solid state dosimetry

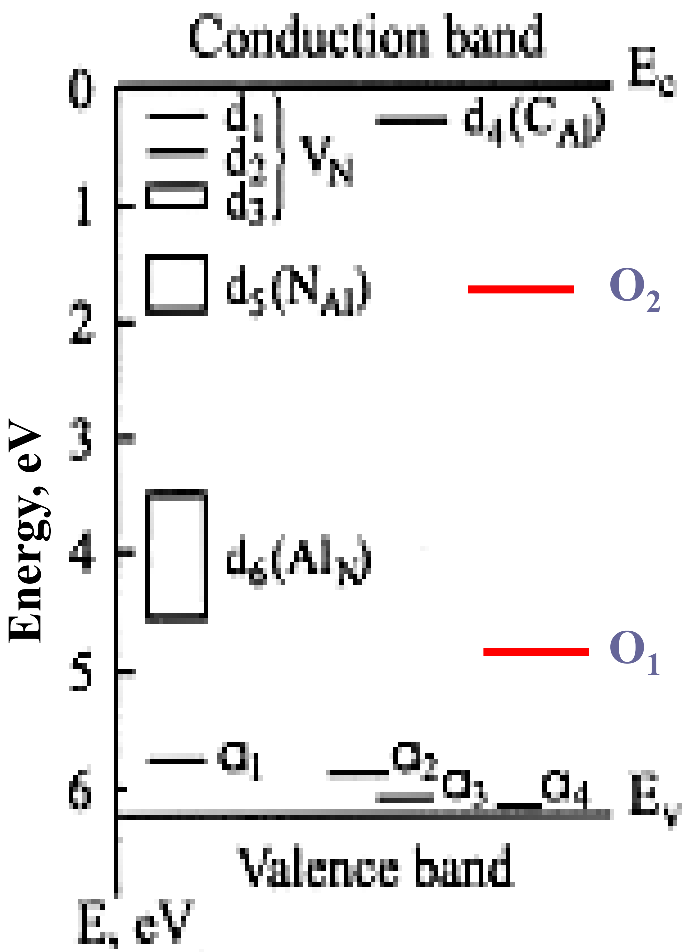
Introduction. Characterization and application of AlN



AlN - wide and direct bandgap material, III-V compound; covalently bounded material with hexagonal crystal structure;

- Wide bandgap $\mathbf{E_g = 6{,}2\ eV}$ (200 nm) ***(Jamashita et al. (1979),*** emitting 210 nm light ***(Yoshitaka Taniyasu et al. (2006); -*** UV-LED devices, deep UV optoelectronics, dielectric layers in optical storage media ;
- High melting point (2800 ^{0}C), good chemical and thermal stability (decomposes at 1800 ^{0}C) – applications in extreme environmental conditions;
- High thermal conductivity (~275 $W \cdot m^{-1} \cdot K^{-1}$) and high electrical insulation ***(Wikipedia)*** – electronic substrates, chip carriers where high thermal conductivity is essential;
- **AlN** – prospective material for radiation detection; light detector of **B** (315 nm – 280 nm) and **C** (280 nm – 200 nm) UV light regions.

Introduction. Defects and their levels in energy band diagram



Tansley & Egan (1992)

Donor levels

d_1, d_2, d_3 – N vacancies (V_N)
d4 – C_{Al}
d5 – N_{Al}
d6 - Al_N

Tansley & Egan (1992); *Chu et al. (1967)*; *Francis and Worell (1976)*; *Jenkins and Dow (1989)*; *Mohammad et al. (1995)*; *Boguslawski et al. (1996)*; *Gorczyca et al. (1997)*

Acceptor levels

α_1 – Al vacancy (V_{Al})
α_2 – C_N
α_3 – Zn_{Al}
α_4 - Mg_{Al}

Francis and Worell (1976); *Jenkins and Dow (1989)*; *Mohammad et al. (1995)*; *Boguslawski et al. (1996)*; *Gorczyca et al. (1997)*

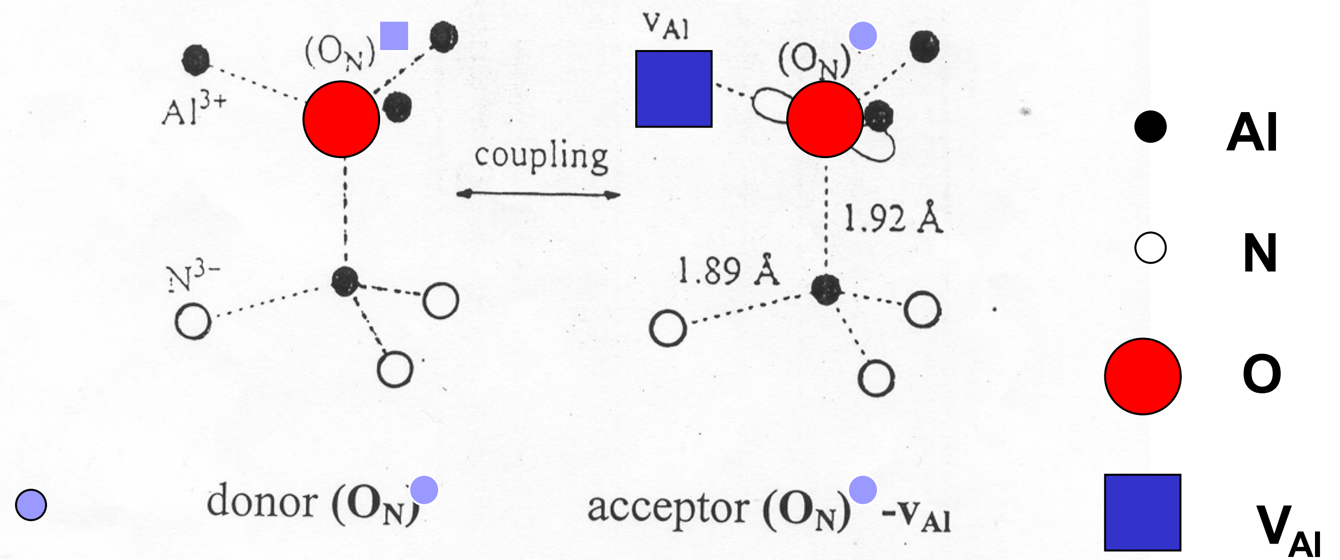
Oxygen-related defects
O_1 – O_N-V_{Al}
O_2 - O_N

G.A.Slack, T.F. McNelly, (1976)
R.A.Youngman, J.H.Harris, (1990)
B.Berzina, L.Trinkler, et al. (2002)

http://www.ioffe.rssi.ru/SVA/NSM/Semicond/AlN/bandstr.html

Introduction. Structure of oxygen-related defects detected by ODMR method

a) Defect structure



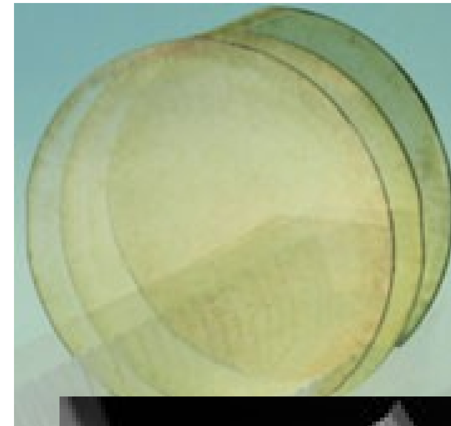
S.Schweizer, U.Rogulis, J,M.Spaeth, L.Trinkler and B.Berzina, *Phys. Stat. Solidi (b)*, **219** (2000) 171.

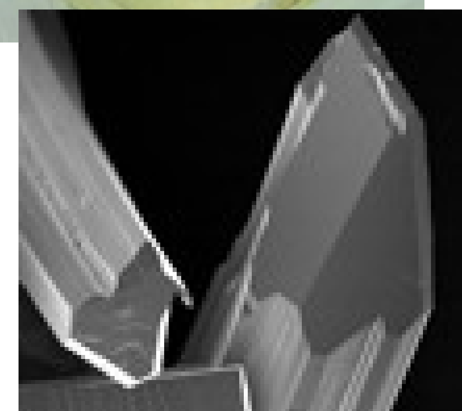
G.A.Slack et al. (1976)

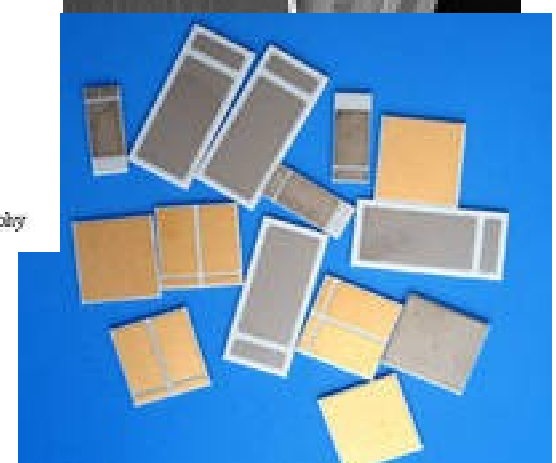
R.A.Youngman, J.H.Harris, (1990)

Introduction. Different forms of AlN

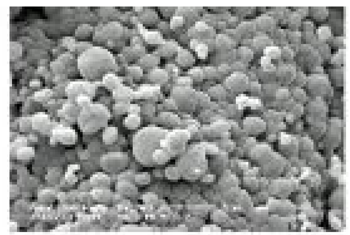
- Single crystals
- Thin films
- Powders of different grain size
- Ceramics
- Nanostructures



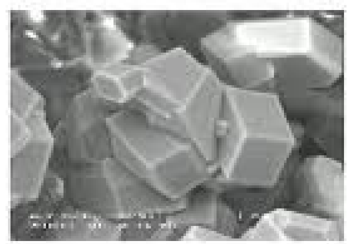


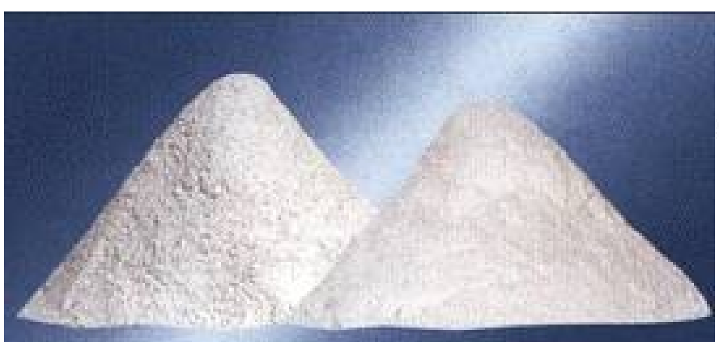


Morphology of the ultra fine AlN powders with spherical shape



Morphology of the AlN powders with perfect crystallography





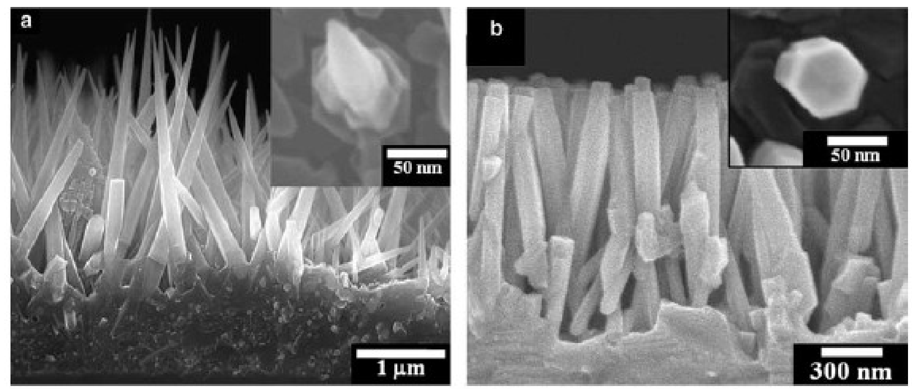
Experimental details. AlN samples

- AlN ceramics

Samples of high-density AlN ceramics were manufactured in Institute of Inorganic Chemistry, Riga Technical University, Latvia (*Palcevskis et al., 1997; 1999*) by sintering at temperature 1600-1800 ºC during 300-900 min of AlN and Y_2O_3 powders (2-9%) produced by plasma synthesis. Grain size 2-5 µm. The produced polycrystalline ceramic cylinders were sliced into 1 mm thick tablets.

- AlN nanostructures

Nanotips Nanorods

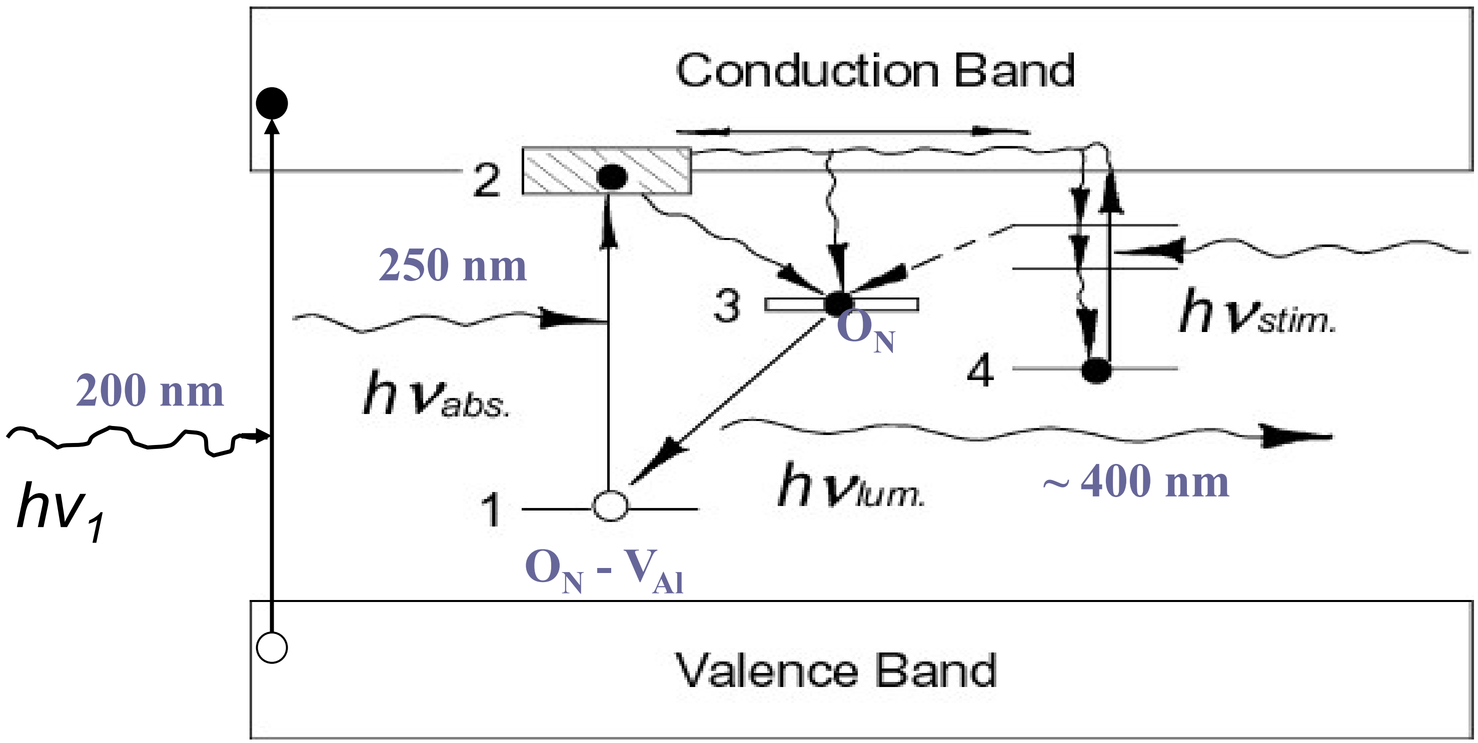


Produced in Center for Condensed Matter Sciences, National Taiwan University, Taiwan (*Shih-Chen Shi, 2005*) by thermal CVD in NH_3 at 900 – 1100ºC during 30 min. Length 1200 nm, diameter 10-100 nm.

Experimental details. Methods of photoluminescence (PL), thermoluminescence (TL) and optically stimulated luminescence (OSL)

- PL is luminescence revealed under continuous irradiation with ultraviolet (UV) or visible light. PL characteristics used: emission spectra and excitation spectra.
- Irradiation of wide band gap materials with UV light and ionizing radiation results not only in immediate response in the form of luminescence but also in ionization – release of previously bound charge carriers. After relaxation charge carriers can be trapped on trapping centers and stay there until supply of additional stimulation energy in the form of heat of light (visible or infrared) releases them, allowing their participation in recombination processes with light emission - TL or OSL, correspondingly.
- OSL characteristics used: OSL emission spectra, OSL excitation spectra, OSL stimulation spectra.
- TL characteristics used: TL glow curves, TL emission spectra, TL excitation spectra.

Participation of oxygen-related defects in PL, TL and OSL mechanisms



Pairs of O_N-V_{Al} and O_N defects are responsible for all type emissions: PL, OSL and TL

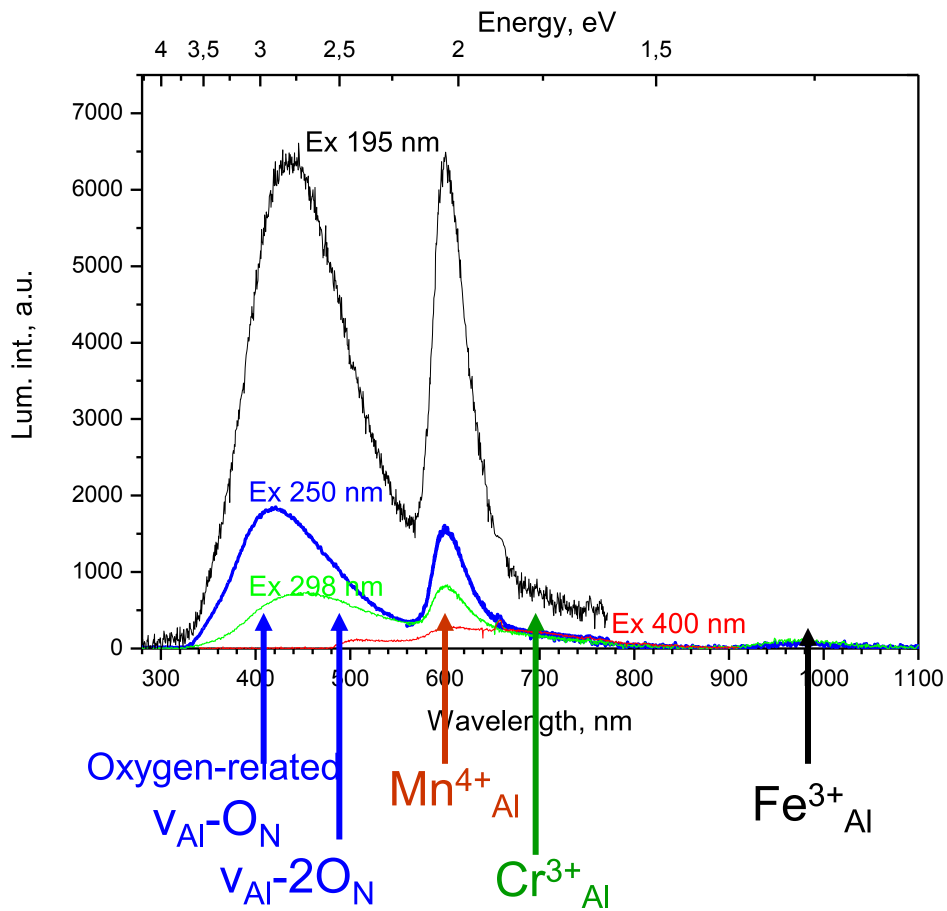
B.Berzina, L.Trinkler, et al. Rad. Eff. & Def. in Solids, 157 (2002) 1089-1092.

Experimental details. Equipment

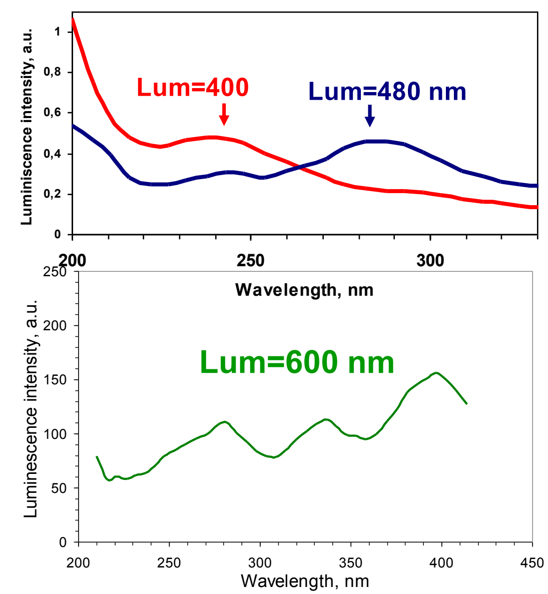
- Experiments on PL, OSL and TL(0-300 °C) were carried out in Institute of Solid State Physics, University of Latvia, Latvia. The setup was equipped with a deuterium lamp (400 W) as a source of UV light and a grating monochromator in the excitation channel. The luminescence signal was analyzed either with a prism monochromator and detected with photo multiplier tube, or with a grating monochromator equipped with a CCD camera. TL measurements were done using a small home-made oven with linear heating up to 300 °C.
- Ionizing radiation induced TL and partly OSL measurements were fulfilled in Riso National Laboratory, Denmark, using the available equipment: Riso model TL/OSL readers with linear planchet heating and Alnor Dosacus TLD reader, operating with hot nitrogen heating. For most of irradiations a 90Y/90Sr beta source build in the Riso TL reader and a 60Co standard gamma calibration facility were used.
- Some of TL measurements were done in University of Nice-Sophia Antipolis, LPES-CRESA, France, using a home-made TL reader with linear heating and an optical multichannel analyzer. UV irradiation was carried out with a deuterium lamp (50 W) with attached interference filters.

AlN ceramics. PL at RT

PL emission



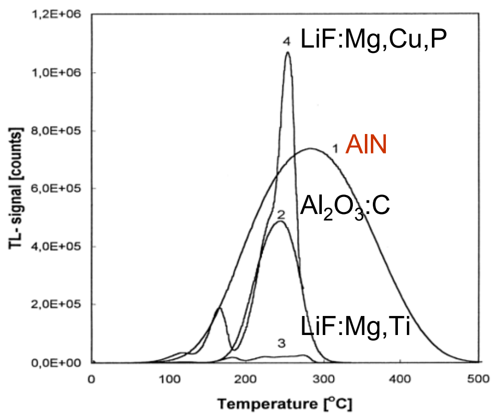
PL excitation



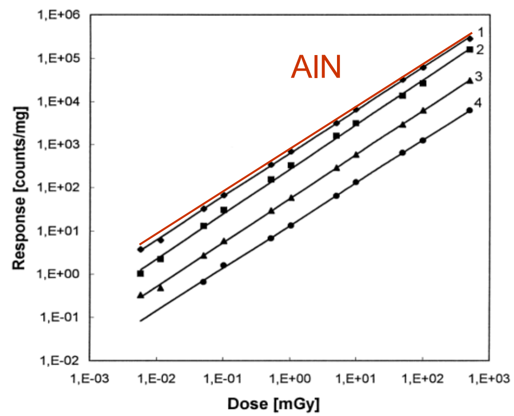
Schweizer, S. et al.,(2000)
Nappe, J.C. et al., (2011).
Karel, F. et al.,(1966).
Benabdesselam, M. et al.,(1995).
Baur, J. et al., (1995).

PL of AlN ceramics: 2 types of oxygen-related defects, manganese, chromium and iron centres. These centres take part also in TL and OSL processes.

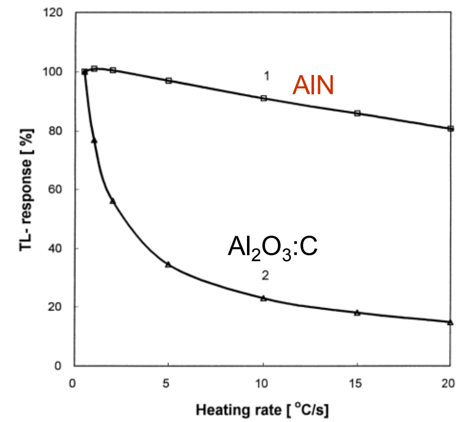
AlN ceramics as a potential dosemeter of ionising radiation



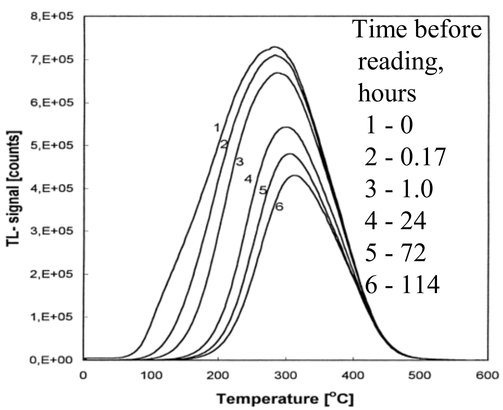
Glow curves of TLD, beta irradiation Ratio of the TL lightsum constitutes 54:27:7:1 for AlN ceramics, LiF:Mg,Cu,P, Al_2O_3:C and LiF:Mg,Ti.



Gamma ray (60Co) dose response of AlN ceramics (1), LiF:Mg,Cu,P (2), Al2O3:C (3) and LiF:Mg,Ti (4)



Influence of heating rate on the TL response of AlN ceramics (1) and Al2O3:C (2) irradiated beta radiation.



Fading of TL signal during storage at RT

TL

Advantageous features of AlN ceramics for application for TL dosimetry:

- high TL sensitivity to exposure to ionising radiation,
- large linear dynamic range of dose response;
- good repeatability and re-usability without requirement of any further annealing in addition to the readout;
- small influence of heating rate on the integrated TL signal;
- mechanical and chemical hardness.

Disadvantage of AlN ceramics for dosimetry application is the high fading of the TL signal on storage at RT.

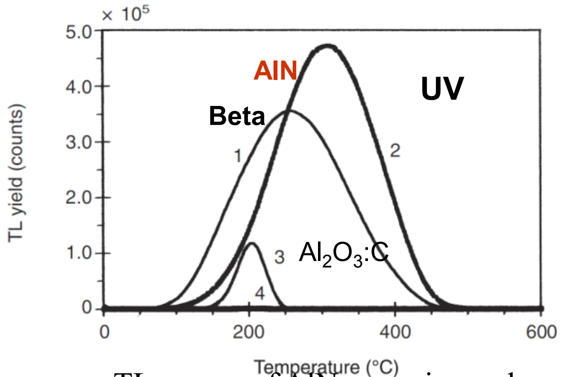
OSL

OSL signal is lower and fading rate higher than TL signal after the same irradiation conditions.

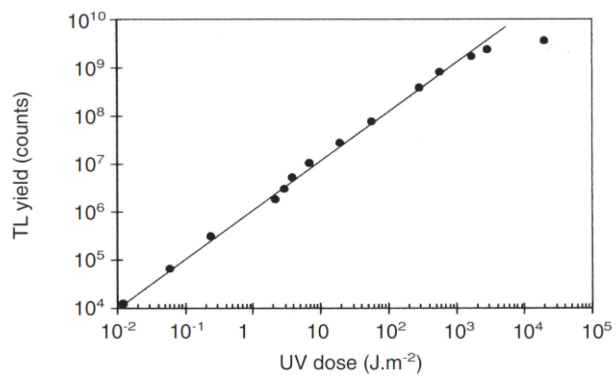
Trinkler et al, (1998)

AlN ceramics as a potential dosemeter of UV light

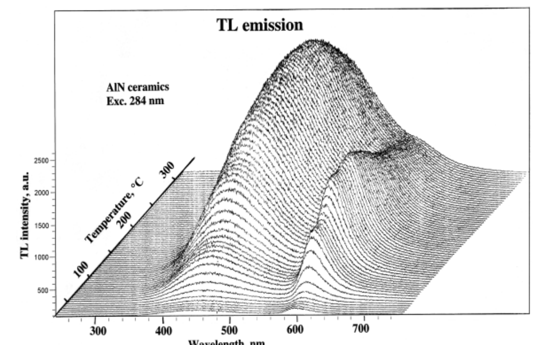
TL



TL curves of AlN ceramics and Al_2O_3:C after irradiation with beta dose and UV light

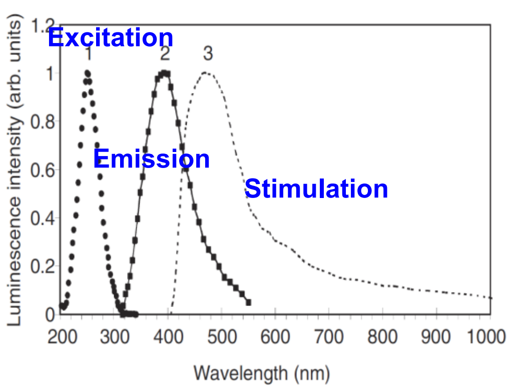


UV light dose dependence of TL from AlN.

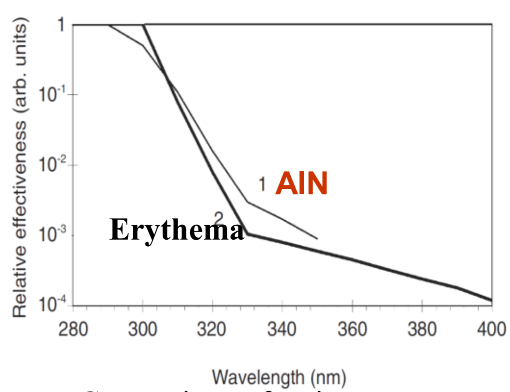


3D image of UV light induced TL emission from AlN ceramics

OSL



Spectral characteristics of UV light induced OSL of AlN ceramics



Comparison of action spectrum of human erythema (*CIE Research Note, 1987*) with OSL excitation spectrum of AlN ceramics.

TL and OSL

Advantageous features of AlN ceramics for application for UV light dosimetry:

- high TL and OSL sensitivity to UV light within 200-350 nm, covering UV-C and UV-B regions,

-OSL excitation spectrum is similar to spectral sensitivity of human skin – potential personal UV dosemeter,

- large linear dynamic range of UV dose response of TL;

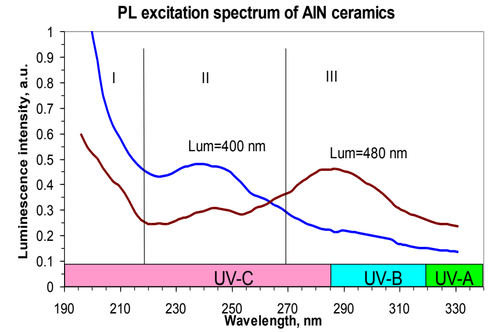
Disadvantage of AlN ceramics

-High fading rate of TL and OSL signal.

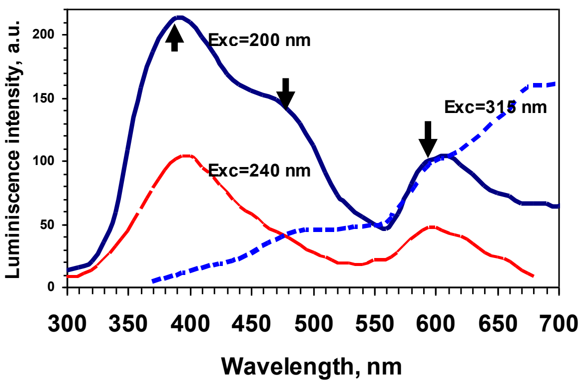
- ***Trinkler et al., , 2000, 2001, 2002***

AlN ceramics. Use of 480 nm emission band for UV induced TL and OSL

PL excitation

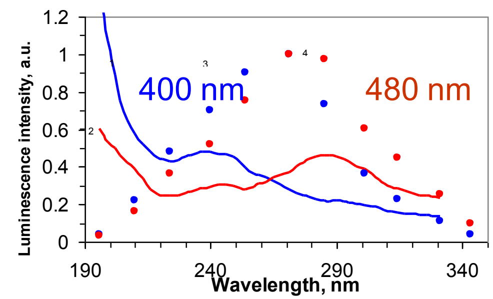


PL emission

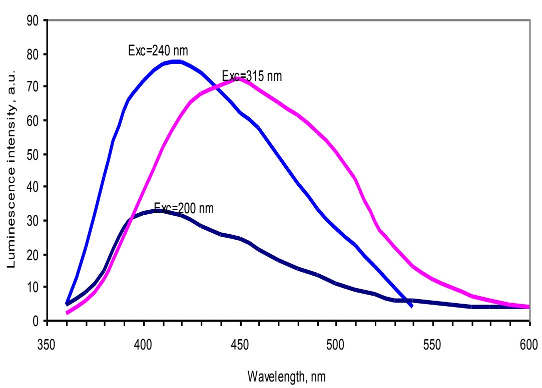


Oxygen related bands 400 and 480 nm are revealed in PL, TL and OSL emission spectra. Each of them has its own band in PL, TL, OSL excitation spectrum.

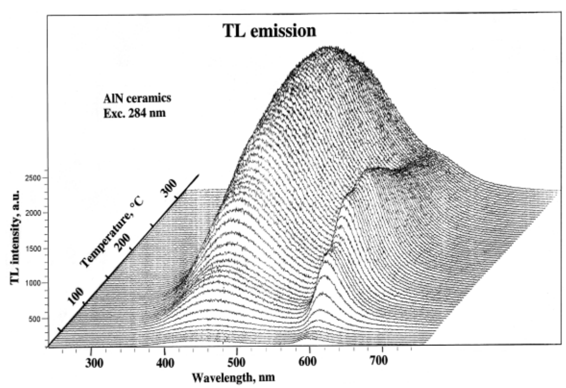
TL excitation



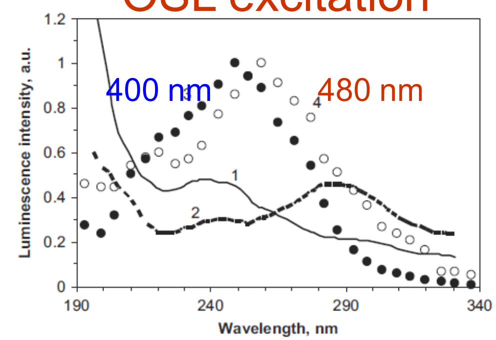
OSL emission



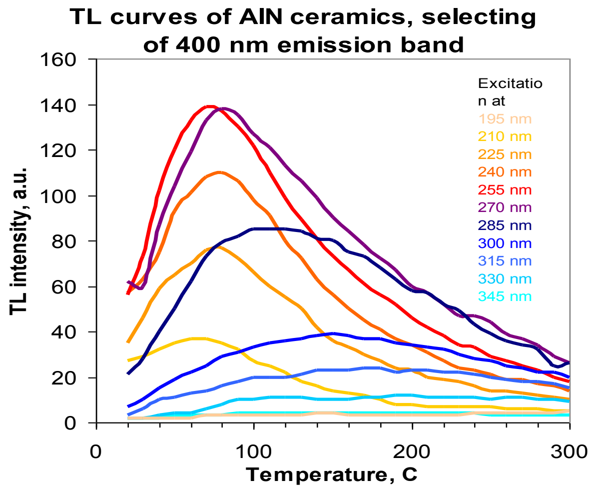
TL emission

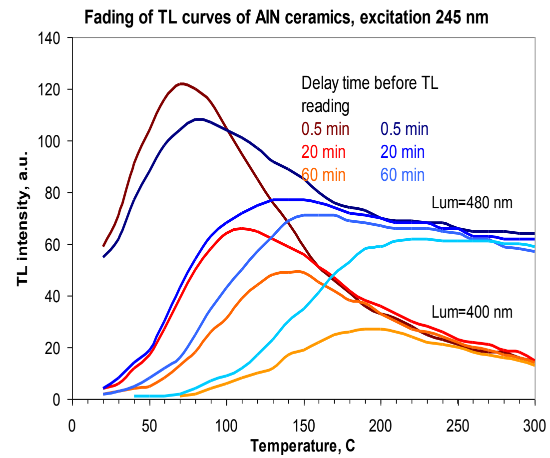


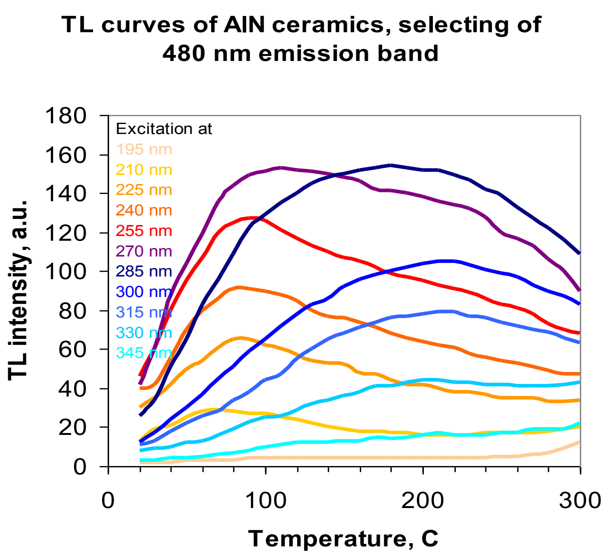
OSL excitation

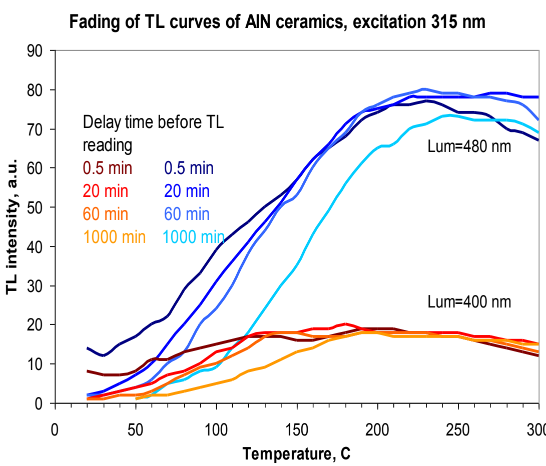


AIN ceramics. Use of 480 nm emission band for UV induced TL and OSL









Mutual relation is found between:

a) excitation 245 nm, emission 400 nm and peak 80 ºC;

b) excitation 280 nm, emission 480 nm and peak 220 ºC.

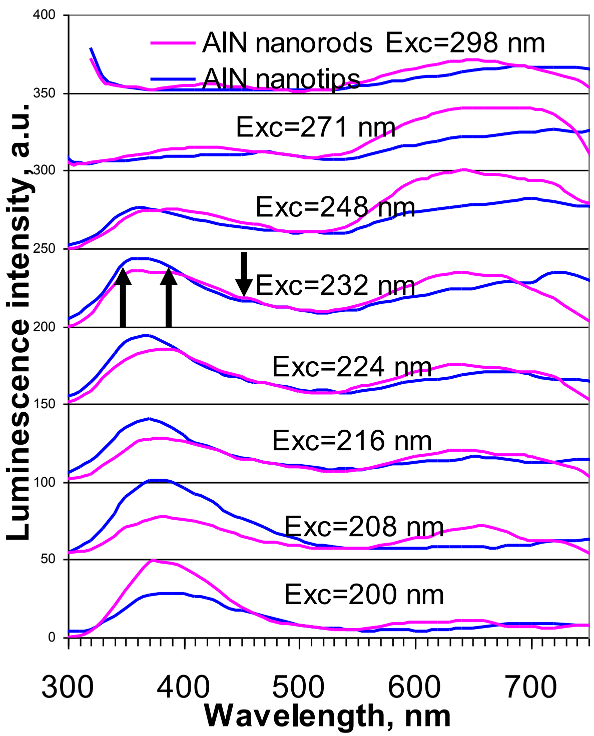
Using AIN for UV dosimetry, it is preferable to detect 480 nm emission band instead of 400 nm band, because

- Excitation falls into UV-B range, which is actual for personal protection
- TL signal rises
- Fading rate of the stored TL signal decreases.

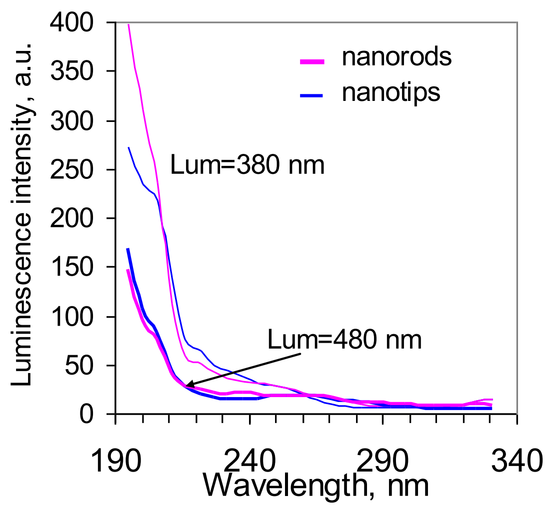
OSL is less suitable for practical application, because its fading rate is higher, than for TL.

AlN nanostructures. Photoluminescence

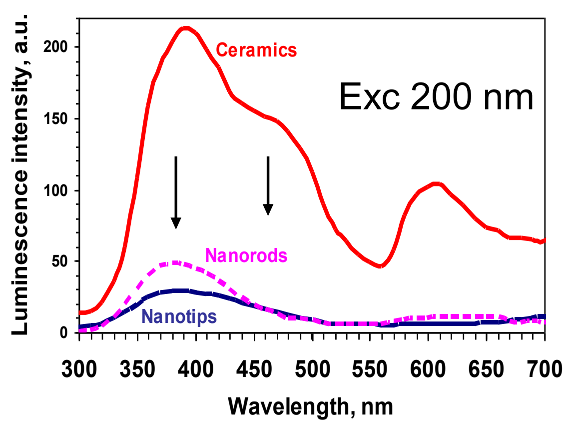
PL emission of AlN nanostructures



PL excitation of nanostructures



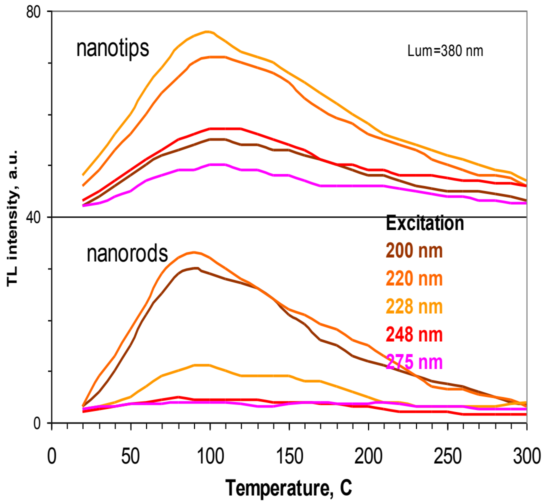
Comparison of PL emission of AlN materials



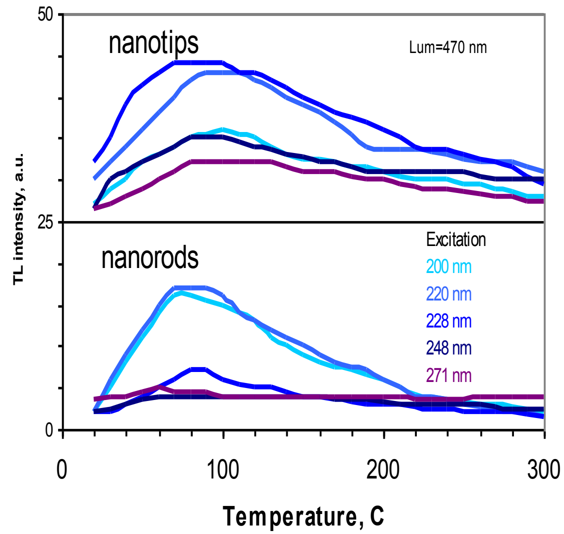
- **Properties of PL in AlN nanostructures**:
- 360+420, 480, 600 nm bands, excited mainly in the host lattice absorption region (200 nm) and weakly in the defect centres absorption bands (230 and 260 nm).
- PL intensity much weaker, than in AlN ceramics

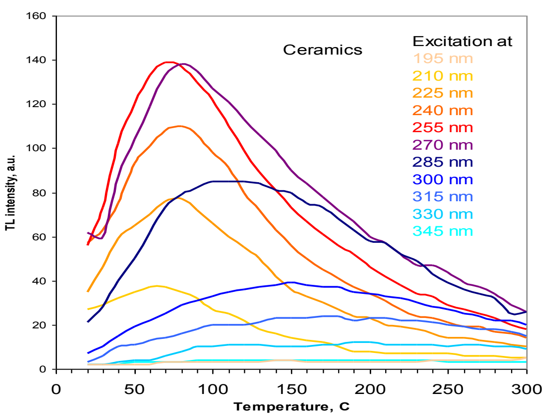
AlN nanostructures. Thermoluminescence

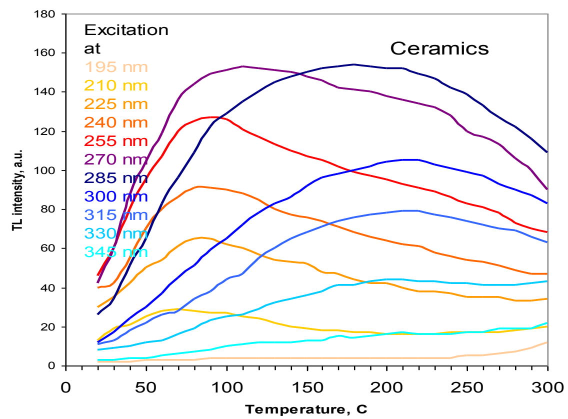
Lum=380 nm



Lum=470 nm





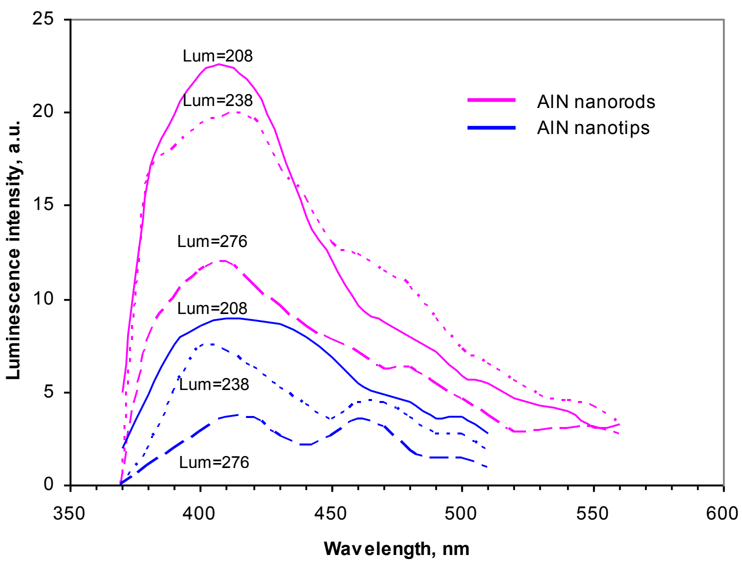


Ceramics: TL emission at 400, 480, (600) nm, each with its own excitation spectrum. Position of the TL peak depends on the emission and excitation wavelength. The optimal TL intensity and stability is observed at Exc=290 nm, Lum=480 nm.

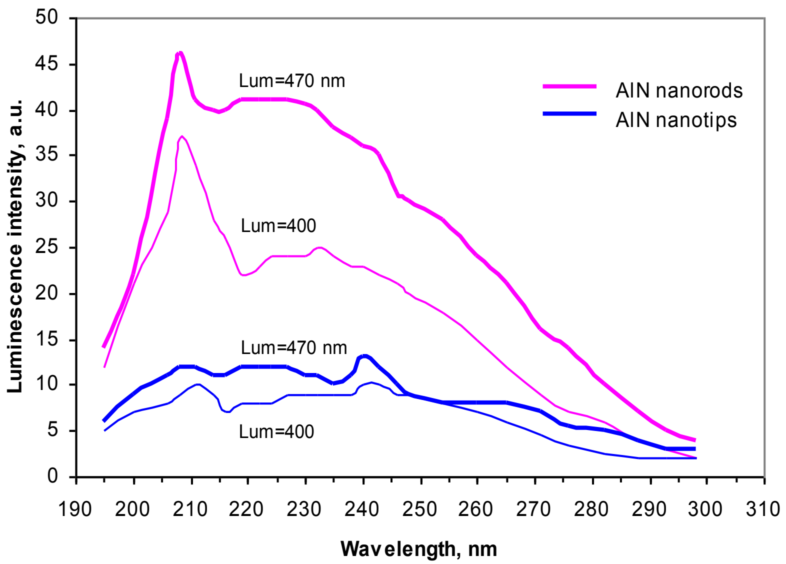
Nanostuctures: TL emission at 380, 420, 480 nm, excited mainly at 200-220 nm (nanorods) or 230 nm (nanotips). Position of the TL peak is 80ºC (nanorods) - 100ºC (nanotips) irrespective of the emission and excitation wavelength.

AlN nanostructures. Optically stimulated luminescence

OSL emission



OSL excitation



Nanostructures: OSL emission bands at 400, 470-480 nm, excited in the 200-300 nm region, with a dominant feature at 200-210 nm (nanorods).

AlN nanostructures. Summary of the main features

- Generally 360-420 and 480 nm luminescence bands of the nanostructures are similar to those of ceramics, evidently originating from recombination processes with participation of the oxygen-related centres.
- In nanostructures the luminescence processes (PL, TL, OSL) occur mainly through the excitation of the host lattice, probably due to the smaller content of the defects in the lattice.
- Absence of a high temperature part in the TL curves of nanostructures means a lack of the deep trap levels, implying the more perfect crystal lattice. In the same time it makes TL signal less stable during storage.
- Slight mutual differences in the luminescence properties of nanorods and nanotips are explained by the modifications of the lattice structure and different surface defects.
- AlN nanostructures are less suitable for the dosimetric application than AlN ceramics due to lower signal and higher fading rate.

Conclusions. Estimation of AlN applicability for solid state dosimetry

- AlN is a wide-gap material whose spectral properties are determined by presence of uncontrolled impurities and intrinsic defects, the main role played by oxygen-related defects.
- AlN ceramics is a material, highly sensitive to irradiation with both ionizing radiation and UV light. The dose of the obtained irradiation can be retrieved using TL and OSL methods. In many aspects AlN ceramics outperforms other actually used TL and OSL dosemeters.
- However, this material has an important drawback, hampering its practical application in dosimetry area – it is high fading rate of the signal during storage at room temperature.
- Selection of the 480 emission band (excited in UV-B region) for detection of TL and OSL signal diminishes the fading effect, though does not eliminate it.
- The studied AlN nanostructures also demonstrate UV light induced TL and OSL. They are less suitable for practical application due to lower and less stable signal compared to AlN ceramics.
- The further studies of this potentially perspective material are planned.

Thank you for attention !