Doctoral (Ph. D.) Theses

Preparation and usability investigation of shape-tailored bismuth tungstate and bismuth vanadate photocatalysts

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1. INTRODUCTION AND OBJECTIVES

While in the developed countries the wastewater treatment is mostly solved, in the developing countries the major problem is even the production of water suitable for human consumption. In most of these cases, conventional water purification processes are irrelevant, so it is necessary to find another alternative purification method to remove the pollutants that cause this problem. Heterogeneous photocatalysis among advanced oxidation processes (AOP's) may be a good solution, which is one of the most dynamically developing recent water purification method.

Heterogeneous photocatalysis is based on a semiconductor material, a photocatalyst, which can be activated with light irradiation to initiate the degradation of organic pollutants. If the electron- and band structure of the semiconductor allows, the excitation can be induced by visible light, a lower energy irradiation, which is more advantageous for the utilization of sunlight than the higher energy UV light excitation. Consequently, the development of visible light active photocatalysts such as bismuth tungstate and bismuth vanadate are a major priority.

It is well-known that the photocatalytic efficiency highly depends on the physical-chemical properties of the photocatalyst, such as particle size, morphology, band-gap, etc. Among these properties, the photocatalyst (micro)morphology and hierarchical organization levels have a prominent role, making possible a fine-tuning of the properties by during their synthesis. This is the so-called controlled crystallization, i.e. shape-tailoring, which favors the development of such morphological properties which are favorable to a specific application.

During my research, I aimed to shape control bismuth tungstate and bismuth vanadate semiconductor photocatalysts. In case of bismuth tungstate, the effect of the synthesis time, the calcination, and the additives on the physical-chemical properties of the microcrystals and as well the photocatalytic activity were intensively investigated. Considering the results, a morphological series was sought, which was obtained by changing the used additives systematically during the synthesis, in order to explore a clear relationship between the produced morphological series, the used additives and the photocatalytic activities.

In the second part of my work, the synthesis of specific crystal facet dominated bismuth vanadate microparticles were targeted by solvothermal crystallization fine-tuned by the change of the pH of the synthesis mixture. The relationship between the amount of photocatalytically active (040) crystal facet and the photocatalytic degradation of two model pollutants, the rhodamine B and the oxalic acid was targeted. However, the crucial role of (040) crystal facet in other chemical reaction was also planned .

2. EXPERIMENTAL METHODS, PROCEDURES

During the bismuth tungstate crystallization process, I investigated the effect of the hydrothermal crystallization time (5, 15, 20 and 40 hours) and calcination (500 °C, 2 hours) on the physico-chemical properties and the photocatalytic activity. I investigated the importance of thiourea, and that of Triton X-100 (TRX) surfactant during the synthesis. Based on the results, thiourea was replaced with other additives of similar structure, but different molecular properties (Hückel polarity). The non-surfactant additives included urea, acetamide, thioacetamide, acetone, glycine, alanine and phenylalanine.

Preparing bismuth vanadate, the morphology was influenced while the crystal facet ratio of (040) was changed with the adjustment of the synthesis pH. After the crystallization, a crystal facet selective copper nanoparticles deposition was carried out.

The crystallographic properties of the as-prepared Bi_2WO_6 and $BiVO_4$ samples were analyzed by X-ray diffractometry (XRD) to verify the success of the synthesis procedures. The primary particle size was calculated from the X-ray diffractograms using the Scherrer equation, and information was also obtained on the crystalline phase composition of the materials and the ratio of the crystal facets. Micromorphological features were investigated by scanning electron microscopy (SEM), while optical properties were measured using a diffuse reflection spectroscope (DRS). From the diffuse reflection spectra, the Kubelka-Munk correlation was used to calculate the band-gap of the photocatalysts, and the first order derivation of the DRS spectra was applied in order to get information concerning the excitability of the samples.

Infrared spectroscopy (IR) was used to check the surface purity. Some samples were subjected to elemental analysis (XPS) to obtain more information about the oxidation state of the surface atoms and residual compounds on the surface. Nitrogen adsorption and Brunnauer-Emmet-Teller (BET) equation were used to determine the specific surface area of the samples. Structural changes were followed by Raman spectroscopy after the crystal facet selective copper deposition was carried out on bismuth vanadate, while the amount of deposited copper was monitored by X-ray fluorescence spectroscopy (XRF). The particle size distributions of bismuth vanadate samples were followed by dynamic light scattering spectroscopy (DLS). The photocatalytic activities of the samples were investigated by photocatalytic degradation of two model pollutants (rhodamine B and oxalic acid) in a double-walled Pyrex® glass reactor with continuous stirring, air inlet and thermostated at 25 °C under UV and visible light irradiation. UV light excitation was provided by 6×6 W fluorescent lamps (λ_{max} = 365 nm), while the visible light sources were 4×24 W conventional energy saving light bulbs (λ> 400 nm). The residual rhodamine B dye was monitored by UV-Vis spectrophotometer ($\lambda_{det.}$ = 553 nm), while the residual oxalic acid concentration was monitored by high performance liquid chromatography (HPLC).

3. SUMMARY OF NEW SCIENTIFIC RESULTS

T. 1. The hydrothermal crystallization parameters (such as the crystallization time, the presence of Triton X) influences the morpho-structural properties of bismuth tungstate (crystal geometry, surface purity), and by this their photocatalytic activity

1.1. Bismuth tungstate microparticles with different morphology were produced by systematic change of the reaction conditions in order to demonstrate that we can produce stable particles by controlled synthesis with high photocatalytic activity under both UV and visible light irradiation.

1.2. The applied additives play a prominent role during the synthesis on the photocatalytically good morphology and the activity. It was proved that thiourea is not only responsible for the formation of ordered morphology, but also for the formation of more effective light absorption, while the surfactant Triton X-100 is responsible for curling the anisotropic sheets and increasing the interplanar space. At the same time, both additives are necessary to form the appropriate morphology.

1.3. The effect of residual thiourea on photocatalytic activity was investigated by infrared spectroscopy, and it was found that the thiourea does not affect the efficiency of photocatalytic degradation, furthermore the remaining thiourea can simultaneously decompose photocatalytically together with the rhodamine B. The type of residual thiourea on the surface was examined by X-ray photoelectron spectroscopy, and it has been found, that the thiourea decomposition product, dithioxyamide accumulated on the surface of the bismuth tungstate by increasing the synthesis time, resulting in the formation of C-C bonds, thus increasing the surface Bi^{5+} ratio compared to Bi^{3+}.

T. 2. The non-surfactant additives' structure defined by the Hückel-polarity affects the properties of $BiVO_4$ such as the morphology quantified by the rosality empiric approach which was observed directly in the photocatalytic activity.

2.1. Two bismuth tungstate morphological series were produced by the systematic change of non-surfactant additive molecules. The physico-chemical properties of the crystallized photocatalysts were studied in the frame of the used additive molecule. The grouping of the samples was carried out in two series based on the Hückel polarity of the additives, which was linked successfully with the light absorption properrties of the photocatalysts, which were increasing with the value of the Hückel polarity. I investigated the photocatalytic activity of the materials under both visible and UV light, and it was shown that the activity trends are closely related to the Hückel polarity values. However, this trend was destroyed, when Triton X-100 was added to the synthesis mixture, nevertheless the usage of TRX has given rise to better defined morphology.

2.2. I have successfully developed an empirical procedure and a relationship expressed in the form of an equation to quantify the morphological peculiarities, i.e. the level of order in the rose-shape. The equation, if the input conditions are met, can tell how the particle approaches the perfect rose-like shape. The base of the R_{SDC} (Rose Similarity Decay Constant) equation is the periodicity, or surface roughness, which was determined by the transformation of scanning electron microscopy images in case of many catalyst particles, averaged over four axis profile.

2.3. A clear correlation between the periodicity values of TRX-free samples and the Hückel polarity values of the used non-surfactant additives was found, and it was proved, that a well-chosen additive can fine-tune the morphological properties of the nascent particles. With the applied R_{SDC} equation to TRX-assisted bismuth tungstate, I proved and quantified, that the rose-like shape, namely, the periodically repetitive spherical particles which is built from individual anisotropic plates have the highest photocatalytic activity.

T. 3. Bismuth vanadate photocatalysts preparation can be linked directly with the appearance of (040) crystal facet which defines the degradation of specific model pollutants and other non-photocatalytic reactions as well.

3.1. I have successfully crystallized different bismuth vanadate photocatalysts with tunable (040) ratio by modifying the pH of the synthesis mixture. I have found that the pH change of the synthesis mixture has a drastic effect on the morphology of the formed particles, which can be followed by the r changes in the XRD patterns of the samples. I investigated the photocatalytic activity of the bismuth vanadate under visible light irradiation by photocatalytic degradation of two different model pollutants, rhodamine B and oxalic acid. A relationship between the presence of the (040) crystal facet intensity and the degradation and transformation amount of oxalic acid. The decomposition of oxalic acid is (040) crystal facet dependent, which confirms the direct reaction with the photogenerated holes. There was no such relationship with the degradation trend of rhodamine B.

3.2. By the crystal facet dependence of copper deposition I proved that the (040) crystal facet plays a key role not only in photocatalysis, but also in other chemical reaction as well. Knowing that copper deposition is (040) crystal facet dependent, the XRF measurements affirmed that the amount of deposited copper also increases with the amount of (040) crystallographic plane Raman spectroscopy measurements and photocatalytic decomposition results, directly confirmation was received that the (040) crystal facet is the most important crystallographic plane in case of bismuth vanadate.

3.3. From the results of the DLS measurements, it was determined that the samples with higher aggregation degrees have higher photocatalytic activity for the degradation of rhodamine B than the samples with more individual particles. However, as the hydrodynamic particle size increases, the photocatalytic conversion of rhodamine B also increases. I have found that the pollutants, which preferred to degrade by photogenerated holes, the (040) crystalline plane plays a crucial role, but it is not the only crucial particle property which influences the photoactivity.

4. APPLICABILITY OF THE SCIENTIFIC RESULTS

During my doctoral research, I worked on the production of two types bismuth oxide-based photocatalysts, and with the presented results, I hope that I contributed to the development of visible light driven photocatalysts, thereby identifying the way for future researchers and of course myself.

Based on my results, bismuth tungstate has a good chance to be used in photocatalytic water treatment in case of more complex, multi-component contaminated water, as two pollutants can degrade simultaneously. The literature has so far failed to quantify the morphological features, which I have tried to replace in my current research by creating a mathematical link which is a good start to create a more complex equations for other morphologies as well. I have highlighted, that the bismuth tungstate potentially can be used as an optical sensor, too, which has good selectivity, but further investigations are needed here.

We learned a lot about bismuth vanadate crystallographic features, promoting the creation of new composites materials. These new materials most likely can increase the bismuth vanadate photocatalytic stability.

5. SCIENTIFIC PUBLICATIONS

Hungarian Scientific bibliography (MTMT) identifier: 10052552

Publications related to the scientific topic of the dissertation:

[1] **Kása Zs.;** Saszet K.; Dombi A.; Hernádi K.; Baia L.; Magyari K.; Pap Zs.: *Thiourea and Triton X-100 as shape manipulating tools or more for Bi_2WO_6 photocatalysts?*

MATERIALS SCIENCE IN SEMICONDUCTOR PROCESSING 74 pp. 21-30., 10 p. **(2018);** DOI: 10.1016/j.mssp.2017.10.001

Citations: 1 (1) **I.F.$_{2017}$: 2.593**

[2] **Kása Zs.;** Baia, L.; Magyari K.; Hernadi K.; Pap Zs.*: Innovative visualization of crystal morphology effects on semiconductor photocatalysts. Tuning the Hückel polarity of the shape-tailoring agents: the case of Bi_2WO_6*; CRYSTENGCOMM 21 pp. 1267-1278. 12 p. (**2019**); DOI: 10.1039/C8CE01744A

Citations: 0 (0) **I.F.$_{2017}$: 3.304**

[3] **Kása Zs.;** Almási E. E.; Hernádi K.; Gyulavári T.; Baia L.; Veréb G.; László Zs.; Pap Zs.: *New insights into the photoactivity of shape-tailored $BiVO_4$ semiconductors via photocatalytic degradation reactions and classical reduction processes*

SUBMITTED FOR PUBLICATION−SCIENTIFIC REPORTS

(I.F.$_{2017}$: 4.122)

[4] **Kása Zs**.; Gyulavári T.; Veréb G.; Kovács G.; Baia L.; Pap Zs.; Hernádi K.: *Novel Applications and Future Perspectives of Nanocomposites* In: Khan M. M.; Pradhan D.; Sohn Y.: *Nanocomposites for Visible Light-induced Photocatalysis*. Heidelberg: Springer International Publishing, **2017**. pp. 333-398. (ISBN:978-3-319-62446-4) (*Book chapter*)

Citation 3 (3)

Σ I.F.$_{2017}$: 10.019

Other international publication:

[5] Vajda K.; **Kása Zs**.; Dombi A.; Németh Z.; Kovács G.; Danciu V.; Radu T.; Ghica C.; Baia L.; Hernádi K.; Pap Zs.*: "Crystallographic" holes: new insights for a*

beneficial structural feature for photocatalytic applications NANOSCALE 7: 13 pp. 5776-5786. , 11 p. (**2015**); DOI: 10.1039/C4NR07157C

Citations: 7 (4) **I.F.$_{2017}$: 7.233**

[6] Vajda K.; Saszet K.; Kedves E. Zs.; **Kása Zs.;** Danciu V.; Baia L.; Magyari K.; Hernádi K.; Kovács G.; Pap Zs.: *Shape-controlled agglomeration of TiO_2 nanoparticles. New insights on polycrystallinity vs. single crystals in photocatalysis* CERAMICS INTERNATIONAL 42 : 2 pp. 3077-3087. , 11 p. (**2016**); DOI: 10.1016/j.ceramint.2015.10.095

Citations: 19 (13) **I.F.$_{2017}$: 3.057**

[7] Magyari K.; Pap Zs.; Tóth Zs. R.; **Kása Zs.;** Emilia L.; Vodnar D. C.; Hernadi K.; Baia L.: *The impact of CuO nanoparticles on the structure and applicability of bioactive glasses*

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6. PRESENTATIONS, POSTER, CONFERENCE PARTICIPITATIONS

1. **Kása Zsolt***: Szén nanocsövön növesztett titán-dioxidok jellemzése fotokatalitikus tulajdonságainak vizsgálata*; SZTE-TTIK Környezettudományi Diákköri Konferencia, Szeged, Hungary (**2014**) (II. prize)

2. **Kása Zsolt**: *Szén nanocsövön növesztett titán-dioxidok jellemzése fotokatalitikus tulajdonságainak vizsgálata*; XIV. Országos Felsőoktatási Környezettudományi Diákkonferencia; Pécs, Hungary, (**2014**)

3. **Zsolt Kása**; Krisztina Vajda; Zsolt Pap; András Dombi; Klára Hernádi ; Gábor Kovács; Virginia Danciu; Lucian Baia: *TiO_2 mikrokristályok alakjának finomhangolása különböző szénfajták segítségével:* XX. International Conference on Chemistry; Kolozsvár, Romania (**2014**)

4. **Kása Zsolt** ; Pap Zsolt; Gábor Kovács; Székely István ; Kedves Zsolt ; Kata Saszet ; Hampel Boglárka ; Szilvia Fodor ; Zsejke-Réka Tóth ; Eszter Orbán et al.: *Nanoszerkezetű fotokatalizátorok és kompozitjaik - alakszobrászat és aktivitás;* XX. International Conference on Chemistry, Kolozsvár, Romania (**2014**)

5. **Kása Zsolt**; Hernádi Klára; Zsolt Pap; András Dombi; Vajda Krisztina; Gábor Kovács; Virginia Danciu; Lucian Baia: *Különböző morfológiájú TiO_2 nanokristályok előállítása és fotokatalitikus aktivitásuk vizsgálata:* XX. International Conference on Chemistry; Kolozsvár, Romania (**2014**)

6. **Kása Zsolt**; Kata Saszet; Zsolt Pap; Gábor Kovács; András Dombi; Klára Hernádi; Lucian Baia; Virginia Danciu: *Examination of the Photocatalytic Activity of Differently Shaped Bismuth Tungstate Microcrystals* - 21st International Symposium on Analytical and Environmental Problems*;* Szeged, Hungary (**2015**)

7. **Kása Zsolt**; Kata Saszet; Zsolt Pap; Gábor Kovács; András Dombi; Klára Hernádi; Lucian Baia; Virginia Danciu: *The crystallization pathway and photocatalytic activity of Bi_2WO_6 microflowers;* EYEC monograph: 4th European Young Engineers Conference, Warsav, Poland (**2015**)

8. **Kása Zsolt**; Hernádi Klára; Zsolt Pap; András Dombi; Krisztina Vajda; Lucian Baia; Virginia Danciu: *TiO_2 – C composite materials for photocatalytic applications;* 18th I. S. in Intercalation Compounds, Strasbourg, French (**2015**)

9. **Kása Zsolt**; Kata Saszet; Zsolt Pap; Gábor Kovács; András Dombi; Klára Hernádi; Virginia Danciu; Lucian Baia: *Morphological fine tuning of bismuth tungstate microcrystals and the evaluation of their photocatalytic activity*; IV. Interdisciplinary Doctoral Conference Pécs, Hungary (**2015**)

10. **Kása Zsolt**; Kata Saszet; Zsolt Pap; Gábor Kovács; András Dombi; Klára Hernádi; Lucian Baia; Virginia Danciu: *Morphology controlled synthesis and characterization of Bi_2WO_6 photocatalysts with advanced visible light photocatalytic activity:* European Materials Research Society, Lille, French (**2015**)

11. **Kása Zsolt;** Saszet Kata, Pap Zsolt; Kovács Gábor; Dombi András; Hernádi Klára; Baia Lucian; Virginia Danciu: *Examination of the Photocatalytic Activity of Differently Shaped Bismuth Tungstate Microcrystals;* The First International Conference on New Photocatalytic Materials for Environment Energy and Sustainability, Göttingen, Germany (**2016**)

12. **Kása Zsolt**; Kata Saszet; Zsolt Pap; András Dombi; Klára Hernádi; Lucian Baia: *Különböző funkciós csoportokkal rendelkező adalékanyagok hatása a bizmut-volframát tulajdonságaira és a fotokatalitikus aktivitására;* XXII. International Conference on Chemistry; Temesvár, Romania (**2016**)

13. **Kása Zsolt**; Kata Saszet; Zsolt Pap; Gábor Kovács; András Dombi; Klára Hernádi; Lucian Baia; Virginia Danciu: *Különböző polaritású adalékanyaggal előállított nagy fotokatalitikus aktivitású bizmut-volframát előállítása és vizsgálata;*

Procedings of the 22nd International Symposium on Analytical and Environmental Problems, Szeged, Hungary (**2016**)

14. Gyulavári Tamás és **Kása Zsolt**; Veréb Gábor; Saszet Kata; Pap Zsolt; Dombi András; Baia Lucian; Hernádi Klára: *Vízkezelés látható fényre aktív titán-dioxid és bizmut-volframát fotokatalizátorokkal;* V. Symposium on Environmental Chemistry, Tihany, Hungary (**2016**)

15. **Kása Zsolt**; Pap Zsolt; Dombi András; Hernádi Klára; Baia Lucian: *Synthesis of morphology controlled bismuth vanadate microcrystals and their photocatalytic activity – The effect of the matrix pH;* 5th European Conference on Environmental Applications of Advanced Oxidation Processes, Prague, Czech Republic (**2017**)

16. **Kása Zsolt**; Bárdos Enikő; Pap Zsolt; Hernádi Klára; Baia Lucian: *Synthesis and stability investigations of bismuth oxide containing mixed oxides;* XXIII. International Conference on Chemistry; Déva, Romania (**2017**)

17. **Kása Zsolt**; Pap Zsolt; Hernádi Klára; Baia Lucian: *Bizmut-vanadát fotokatalizátorok előállítása és stabilitásvizsgálata:* Proceedings of the 23rd I.S. on Analytical and Environmental Problems; Szeged, Hungary (**2017**)

7. CO-AUTHOR CONFERENCE PARTICIPATIONS

18. Pap Zsolt; Krisztina Vajda; Klára Hernadi; András Dombi; Zoltán Németh; Gábor Kovács; Virginia Danciu; Lucian Baia; **Zsolt Kása**; Teodora Radu: *Shaping of titania nanocrystals with carbon materials for photocatalytic applications* – XXVIII. International Winterschool on Electronic Properties of Novel Materials, Kirckberg, Ausztria (**2014**)

19. Pap Zsolt; Kovács Gábor; István Székely; Kedves Zsolt; Saszet Kata; Hampel Boglárka; Fodor Szilvia; Tóth Zsejke-Réka; Orbán Eszter; Kovács Zoltán; **Kása Zsolt** et al.: *Nano-sized photocatalysts and their composites – shape tailoring and activity,* XX. International Conference on Chemistry, Kolozsvár, Romania (**2014**)

20. Kedves Zsolt; Saszet Kata; Vajda Krisztina; **Kása Zsolt**; Kovács Gábor; Pap Zsolt; Dombi András; Hernádi Klára; Danciu Virginia; Vulpoi Adriana et al.: *Synthesis of differently shaped TiO_2 nanocrystallines with hidrothermal treatment and the analysis of their photocatalytic activity;* XX. International Conference on Chemistry, Kolozsvár, Romania (**2014**)

21. Zsolt Pap; Gábor Kovács; Zsejke-Réka Tóth; Krisztina Vajda; Éva Karácsonyi; **Zsolt Kása**; Szilvia Fodor; Zsolt Endre Kedves; István Székely; Kata Saszet; et al.: *The Functioning Mechanism of Photocatalytic Systems from the Charge Transfer Point*

of View. "The Adventure of the Electron, XXI. International Conference on Chemistry, Csíksomlyó, Romania (**2015**)

22. Saszet Kata; **Kása Zsolt**; Gábor Kovács; Zsolt Pap; Danciu Virginia; Adriana Vulpoi; Klára Magyari; András Dombi; Klára Hernádi; Lucian Baia: *Bi_2WO_6 nanovirágok hidrotermális előállítási paramétereinek finomhangolása és alkalmazhatósági spektrumának vizsgálata,* XXI. International Conference on Chemistry, Csíksomlyó, Romania (**2015**)

23. Gábor Veréb; Tünde Alapi; Krisztina Schrantz; Krisztina Vajda; Gergő Simon; Éva Karácsonyi; Eszter Arany; Emese Szabó; Ákos Kmetykó; Erzsébet Illés; Károly Mogyorósi; Georgina Rózsa; Tamás Gyulavári; **Zsolt Kása;** Et al.: *New challenges in water purification: Innovative methods for the elimination of oily contaminants pesticides and pharmaceuticals:* Water and Wastewater Treatment on Industry: II. Soós Ernő Scientific Conference, Nagykanizsa, Hungary (**2015**)

24. Zsolt Pap; Szilvia Fodor; Tamás Gyulavári; Gábor Kovács; Zsejke-Réka Tóth; **Zsolt Kása**; Enikő Bárdos; Georgina Rózsa; Gergő Simon; Zsuzsanna Kozmér; et al.: *New nanocomposites and nanostructures in water cleaning:* XXII. International Conference on Chemistry, Temesvár, Romania (**2016**)

25. Ravasz Alpár; Kedves Zsolt; **Kása Zsolt**; Kovács Gábor; Pap Zsolt; Magyari Klára; Hernádi Klára; Lucian Baia: *$BiVO_4$ „TiO_2 és WO_3 ternáris kompozitrendszerek előállítása és fotokatalitikus aktivitásának vizsgálata*, XXII. International Conference on Chemistry, Temesvár, Romania (**2016**)

26. Pap Zsolt; Fodor Szilvia; Gyulavári Tamás; Kovács Gábor; Tóth Zsejke; **Kása Zsolt**; et al.: *New nanocomposites and nanostructures in water cleaning II.*, XXIII. International Conference on Chemistry, Déva, Romania (**2017**)

27. K. Saszet; Zs. Kedves; Zs. Pap; **Zs. Kása**; G. Kovác ; V. Danciu; K. Magyari ; A. Dombi; K. Hernádi; L. Baia: *Study of photodegradation intermediated using specific TiO_2-based nanostructures*; 5th European Conference on Environmental Applications of Advanced Oxidation Processes, Prague, Czech Republic (**2017**)

28. Gábor Veréb; Krisztina Vajda; **Zsolt Kása**; Zsolt Kedves; Kata Saszet; Klára Hernádi; Zsolt Pap: *Preparation and characterisation of „TiO_2-C" hybride materials,* European Conference on Environmental Applications of Advanced Oxidation Processes (EAAOP5), Prague, Czech Republic (**2017**)

29. Alpár Ravasz; Endre-Zsolt Kedves; **Zsolt Kása**; Lucian Baia; Zsolt Pap: *Hydrothermal synthesis and photocatalytic activity of ternary $BiVO_4$, TiO_2 and WO_3 composite systems*, European Conference on Environmental Applications of Advanced Oxidation Processes (EAAOP5), Prague, Czech Republic (**2017**)