

Nickel-based catalysts: Dependence of properties on nickel loading and modification with palladium

Vesna D. Nikolić¹, Željko J. Kamberović², Marija S. Korać², Zoran M. Anđić³, Aleksandar M. Mihajlović¹, Jelena B. Uljarević¹

¹Innovation Center of the Faculty of Technology and Metallurgy, University of Belgrade, Belgrade, Serbia

²Faculty of Technology and Metallurgy, University of Belgrade, Belgrade, Serbia

³Innovation Center of the Faculty of Chemistry, University of Belgrade, Belgrade, Serbia

Abstract

The aim of this study was comparative analysis of catalysts with 5 and 20 wt.% Ni loading, with and without Pd additive, and it included: determination of reduction degrees, investigation on microstructure by scanning electron microscopy and chemical analysis by energy-dispersive spectroscopy. Ni/Al₂O₃ and Ni-Pd/Al₂O₃ catalysts supported on α -Al₂O₃-based foams were synthesized by aerosol assisted impregnation. Impregnation of the foams by ultrasonically aerosolized chlorides was followed by drying at 473 K and hydrogen reduction at 533 and 633 K. Lower Ni loading resulted in higher reduction degrees. Ni loading of 20 wt.% enabled relatively uniform coverage of the foam with a metallic coating. Nearly complete reduction was achieved at both temperatures with activity modifier (Pd). Reduction degrees reached 99.4 and 98.2 wt.% at 533 K for 5 and 20 wt.% of Ni, respectively.

Keywords: catalysts, Ni, Pd, modification, α -Al₂O₃-based foam, aerosol assisted impregnation.

Available online at the Journal website: <http://www.ache.org.rs/HI/>

Metal-ceramic composite systems have been intensively researched in recent years because properties of a base material can be significantly enhanced by a small quantity of additive. For instance, there are composites such as metals reinforced with ceramic particles, dispersed in a metal matrix. Multiple strengthened Cu-Al₂O₃ systems, that could be suitable for using at elevated temperatures, are produced from ultra-fine and nanocomposite Cu-Al₂O₃ powders. Those powders can be synthesized by using novel method that includes spray drying of dissolved metal salts [1,2]. Composite catalytic materials with metallic particles dispersed on oxide supports are applied in various heterogeneous catalysis processes [3,4]. As an example, Ni/Al₂O₃ catalysts are commonly used for reforming of hydrocarbons [5–7]. Metallic particles are supported on γ -Al₂O₃ powder [5,8], reticulated α -Al₂O₃ foams [9,10], etc. Reticulated ceramic foams have high pore volumes and exhibit good mass transfer, good heat conductivity and low pressure drops for the duration of the fluid flow. Therefore, industrial application of ceramic foams as catalyst support could be favorable [10–12].

Although catalysts based on noble metals are common due to excellent catalytic activity, stability and

selectivity [8], they are expensive for using in industry [13]. Ni is an effective alternative for noble metals because of low cost [6,7,14] and excellent catalytic properties [15]. Ni-based catalysts can be rapidly deactivated due to deposits that form on active sites and sintering of Ni at elevated temperatures. However, different supports, synthesis methods and catalytic activity modifiers (additives), improve catalytic activity, deactivation resistance and selectivity [6,7,14,15]. Modification with low amount of Pd (0.01 [16] to 0.3 wt. % [6]) greatly enhances catalysts properties. Catalyst's activity and selectivity can be drastically deteriorated if metals are unevenly dispersed on support [6,16]. Conventional thermochemical methods for Ni-based catalysts synthesis include oxidative calcination and reduction by hydrogen (activation). Calcination causes formation of NiAl₂O₄ spinel phase, especially in the case of impregnation and γ -Al₂O₃ support. NiAl₂O₄ is hardly reducible and it can inhibit catalytic activity because of low amount of Ni particles [5–7]. As reported by Gayan *et al.* [17], α -Al₂O₃ support minimizes NiAl₂O₄ presence in the Ni/Al₂O₃ system obtained by impregnation. Novel methods that involve ultrasonically generated aerosols provide synthesis of fine spherical particles with precise stoichiometry and high purity [18–20]. Ultrasonic spray pyrolysis is favored in large scale production in industry [21]. Gurmen *et al.* [22] pyrolyzed and reduced ultrasonically aerosolized NiCl₂ and FeCl₂ solution in hydrogen flow. They obtained fine

SCIENTIFIC PAPER

UDC 544.47:66.097:543:546.47

Hem. Ind. **70** (2) 137–142 (2016)

doi: 10.2298/HEMIND140928090N

Correspondence: V.D. Nikolić, Innovation Center of the Faculty of Technology and Metallurgy, University of Belgrade, Karnegijeva 4, 11120 Belgrade, Serbia.

E-mail: vnikolic@tmf.bg.ac.rs

Paper received: 28 September, 2014

Paper accepted: 23 December, 2014

spherical Fe–Ni particles in a single step, without calcination.

Previously, Ni/Al₂O₃ and Ni–M/Al₂O₃ catalysts, modified by additives M = Pd, Cu or Fe, were synthesized by aerosol assisted impregnation [23]. Oxides and chlorides were used as precursors for metals. Reduction of chloride precursor at 633 K, without previous calcination, resulted in optimal morphology of Ni coating. Modification with Pd enabled the highest reduction efficiency of all precursors [23] due to the hydrogen spillover effect [24, 25]. Chloride precursors for Ni–Pd coating reached significantly higher reduction degree (98.2 wt. %) than oxide precursors (20.0 wt. %) at 533 K [23].

On the basis of previously obtained results [23], Ni-based catalysts were synthesized by aerosol assisted impregnation using chlorides as precursors. Catalysts were supported on previously prepared α -Al₂O₃-based foams [26,27]. The aim of the presented research was to investigate dependence of catalysts properties from different Ni loading and from modification by Pd additive. Comparative analysis of the catalysts included determination of reduction degrees in hydrogen stream and microstructure development, which was investigated by scanning electron microscopy (SEM). An additional investigation included point analysis of reduced Ni–Pd/Al₂O₃ samples by energy-dispersive spectroscopy (EDS).

EXPERIMENTAL

Non-modified Ni/Al₂O₃ and modified Ni–Pd/Al₂O₃ catalysts with 5 and 20 wt.% Ni loading were synthesized by aerosol assisted impregnation method. Considering Ni–Pd/Al₂O₃ catalysts, Ni was modified with 0.1 wt.% of Pd. Used materials included: NiCl₂·6H₂O, PdCl₂ (MERCK, pro analysis) and α -Al₂O₃-based foams. The foams were produced by polymer replication route that is described elsewhere [26,27]. First, the foams were pre-heated at 473 K in a tubular quartz reactor and then impregnated with aerosolized chloride solutions at that temperature. Subsequently, samples were dried at 473 K for 60 min in air. The catalysts were reduced by hydrogen at 533 and 633 K for 90 min. Set-up for the aerosol assisted impregnation and the synthesis method details are presented elsewhere [23].

Reduction degrees of chlorides on the foam surface were determined on the basis of chlorine mass loss before and after hydrogen reduction. After the impregnation and subsequent drying at 473 K for 60 min, the samples were stored in desiccators with silica gel until the mass measurements to prevent absorption of moisture from the air. Mass ratio of chlorides and the foam was a known value. The reduction experiments were performed under static conditions as follows. First, the

samples were set into a quartz tube and reaction area was purified by nitrogen flow. Then, nitrogen flow was stopped and hydrogen was introduced into the tube. Hydrogen flow was conducted through system for purification and drying that was consisted of: columns with silica gel, Cu powder and CaCl₂ and wash bottle with H₂SO₄. A tubular electric resistance furnace was heated to the selected temperature and then the tube was put in. Measuring of reduction time started at that moment. Reduction of the dried samples was carried out at 533 and 633 K for 90 min (H₂ flow rate 20 L/h). Reduced samples were placed in desiccators, cooled down to room temperature and their mass was determined afterward.

Microstructures of the samples after drying and after reduction experiments were investigated with a JEOL JSM-5800LV scanning electron microscope (SEM) equipped with an energy-dispersive spectrometer (EDS). SEM/EDS point analysis was carried out for Ni–Pd/Al₂O₃ catalysts with different metal loading, reduced at 533 K for 90 min.

RESULTS AND DISCUSSION

Reduction degrees of NiCl₂ on the foam surface are presented in Table 1.

Table 1. Reduction degrees, wt.%, of NiCl₂ on α -Al₂O₃ based foam after 90 min

Sample description	Temperature, K	
	533	633
5 wt.% of Ni	60.2	67.7
20 wt.% of Ni	23.7	35.2

As expected, higher reduction degrees were obtained at 633 than at 533 K for all the samples. However, when Ni loading was lower, significantly higher amount of NiCl₂ was reduced at both temperatures due to the better exposure of NiCl₂ particles to H₂ flow when NiCl₂ content was lower. These results could be explained with microstructures of the samples before and after hydrogen reduction (Figs. 1–3).

SEM photographs of the dried samples are presented in Fig. 1a and b.

After SEM analysis of the dried sample with 5 wt.% Ni loading, presence of relatively unevenly distributed, fine, submicron-sized NiCl₂ particles was noted, as shown in Fig. 1a. Agglomerates were not detected. Despite relatively uneven particle distribution, satisfying reduction degrees of NiCl₂ were achieved at both temperatures (Table 1). These results could be explained by obtained morphology, because fine particles were non-agglomerated and, consequently, they had good exposure to H₂. In addition, good mass and heat transfer through the foam was one of the reasons for

satisfying reduction degrees. Figure 1b shows microstructure of the dried sample with 20 wt.% Ni loading. Higher NiCl_2 amount resulted in formation of cracked NiCl_2 crust, which completely covered the foam surface. This crust appeared to be thick on certain sites. A main reason for obtained cracks was dehydration of $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$ during the drying procedure, while filter-like behavior of the foam provided complete coverage with NiCl_2 . Higher Ni loading resulted in lower reduction degrees at both temperatures (Table 1), due to absence of small individual NiCl_2 particles.

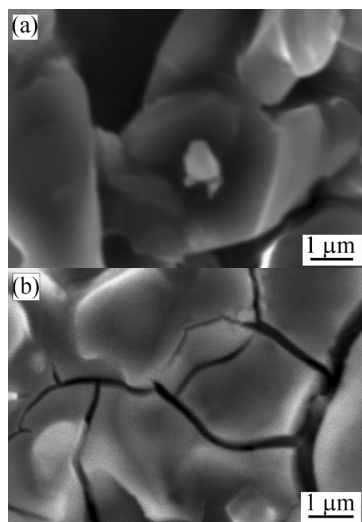


Figure 1. Sample dried at 473 K – $\text{NiCl}_2/\text{Al}_2\text{O}_3$, Ni loading: a) 5 and b) 20 wt.%.

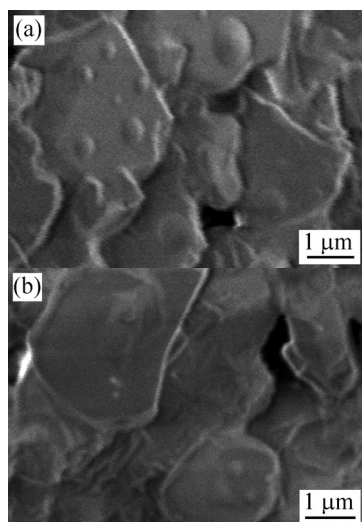


Figure 2. $\text{Ni}/\text{Al}_2\text{O}_3$ reduced at 533 K, nominal Ni loading: a) 5 and b) 20 wt.%.

SEM photographs of the $\text{Ni}/\text{Al}_2\text{O}_3$ samples reduced at 533 K are presented in Fig. 2a and b.

SEM photographs of the $\text{Ni}/\text{Al}_2\text{O}_3$ samples reduced at 633 K are presented in Fig. 3a and b.

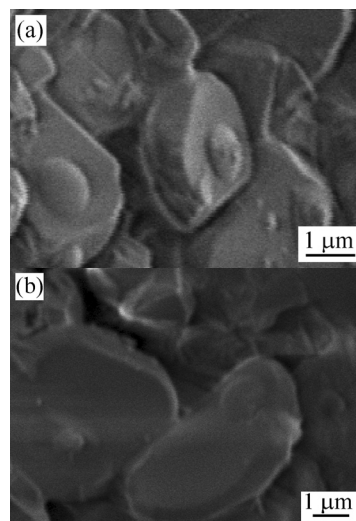


Figure 3. $\text{Ni}/\text{Al}_2\text{O}_3$ reduced at 633 K, nominal Ni loading: a) 5 and b) 20 wt.%.

After the reduction of $\text{Ni}/\text{Al}_2\text{O}_3$ with 5 wt.% Ni loading at both temperatures, small, island-like Ni particles were formed. Ni islands were isolated and sub-micron-sized particles were noted, as presented in Figs. 2a and 3a. No significant changes in the microstructures were noted after the reduction at each temperature. With 20 wt.% Ni loading, the samples reduced at both temperatures had similar microstructures: Ni coating relatively uniformly covered the foam surface, as presented in Figs. 2b and 3b. The reduction with H_2 led to disappearance of visible cracks. Formation of these microstructures could be explained by complex mass transport mechanism that occurred at elevated temperature during the reduction. At the reduction conditions, Ni particles gained sufficient mobility and migrated, which resulted in formation of contacts between particles and their growth. No agglomerates occurred in any of the reduced samples.

Reduction degrees of NiCl_2 with added PdCl_2 , supported on the foam surface, are presented in Table 2.

Table 2. Reduction degrees, wt.%, of NiCl_2 with PdCl_2 on $\alpha\text{-Al}_2\text{O}_3$ based foam after 90 min

Sample description	Temperature, K	
	533	633
5 wt.% Ni, 0.1 wt.% Pd	99.4	99.7
20 wt.% Ni, 0.1 wt.% Pd	98.2	99.6

Results of the reduction experiments showed minor differences in reduction degrees of the $\text{Ni-Pd}/\text{Al}_2\text{O}_3$ samples. All the samples were almost completely reduced at both investigated temperatures (Table 2). Although slightly higher reduction degrees were achieved at 633 K, obtained results indicate that lower reduction temperature can successfully be used for the catalysts synthesis, if the catalysts are modified with

Pd. The main reason for increased reduction efficiency was modification by Pd additive, which caused the hydrogen spillover effect. In general, this effect occurs in the presence of Pd surface, when H₂ molecules dissociate into atoms and rapidly diffuse into crystal lattices of Ni species [24,25].

SEM photographs of Ni–Pd/Al₂O₃ catalysts with 5 and 20 wt.% Ni loading, reduced at 533 K, are presented in Fig. 4a and b, respectively.

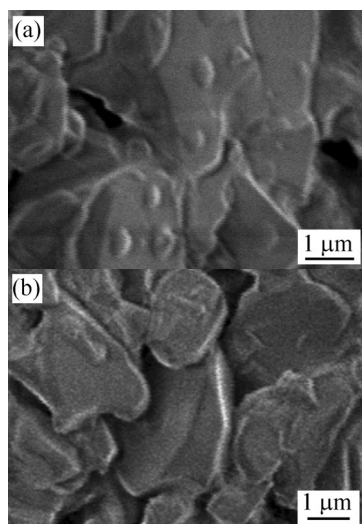


Fig. 4. Ni–Pd/Al₂O₃ reduced at 533 K, nominal Ni loading: a) 5 and b) 20 wt. %.

Similar microstructures were obtained for Ni–Pd/Al₂O₃ catalysts, reduced at 533 K, and for Ni/Al₂O₃ catalysts, reduced at 533 and 633 K. Ni–Pd/Al₂O₃ samples with 5 wt.% of Ni had fine, submicron-sized and isolated island-like particles dispersed on the foam surface, as presented in Fig. 4a. With higher Ni loading (20 wt.%), metallic coating relatively evenly covered the foam, as presented in Fig. 4b. Particles agglomeration did not occur in any of the samples. Modification with 0.1 wt.% of Pd enabled almost completed hydrogen reduction at 533 K. Ni-based catalysts prepared by conventional thermochemical methods are calcined before hydrogen reduction. The calcination treatment (*e.g.*, at 823 K [6] or 873–923 K [5, 7]) provides forming of oxide mixtures that contain oxide precursors for catalytically active components. During that treatment, especially in the case of impregnation method, hardly reducible NiAl₂O₄ phase is commonly formed [5–7]. In addition, the authors concluded that Ni-based catalysts, prepared by using aerosol assisted impregnation and calcined at 773 K, had undesirable agglomerates on the α-Al₂O₃-based foam surface. Ni-based catalysts prepared by using the same method and obtained from chloride precursors reached significantly higher reduction degrees at same temperatures [23].

Point SEM/EDS analysis was performed for Ni–Pd/Al₂O₃ catalysts with different Ni loading, reduced at 533 K. Analyzed points are marked in Fig. 5a and b. The results are presented in Table 3.

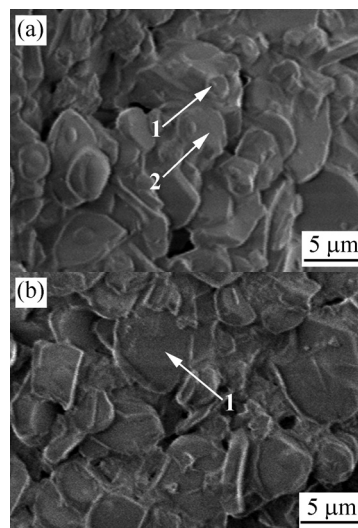


Figure 5. SEM micrographs of Ni–Pd/Al₂O₃ reduced at 533 K, nominal Ni loading: a) 5 and b) 20 wt. %.

Table 3. Point SEM/EDS analysis (wt. %) of Ni–Pd/Al₂O₃ reduced at 533 K

Metal loading	Mark	Ni	Al	Si	O
5 wt.% Ni, 0.1 wt.% Pd	1	93.50	1.97	0.63	3.90
	2	1.50	40.32	5.16	53.02
20 wt.% Ni, 0.1 wt.% Pd	1	94.21	1.53	0.58	3.68

Considering the sample with 5 wt.% Ni loading, island-like metallic particle mainly consisted of Ni. Pd was not detected due to very low concentration. Small amounts of Al, Si and O from the α-Al₂O₃ based foam were also detected. EDS analysis of point with no visible metallic particles showed predominance of elements that correspond to oxides in the α-Al₂O₃ based foam. With Ni loading of 20 wt.%, chemical analysis of a random point revealed that the main element in the metal coating was Ni. Similar to the sample with lower metal content, Pd was not detected because of low concentration. In addition, small amounts of Al, Si and O from the foam were noted. These results confirmed that higher metal loading enabled relatively uniform dispersion of metallic particles over the foam surface.

The conclusion is that Ni–Pd/Al₂O₃ catalysts supported on the α-Al₂O₃ based foams can successfully be produced by hydrogen reduction of chloride instead of oxide precursors, without the calcination step. Chloride precursors for Ni and Pd can be reduced at very low temperature (533 K). Optimal metal loading is 20 wt.%

of Ni modified by 0.1 wt.% Pd due to the relatively even coverage of the foam surface with metallic coating.

CONCLUSION

A comparative analysis of catalysts with 5 and 20 wt.% of Ni, with Pd and without additive, was conducted. The catalysts were supported on α -Al₂O₃-based foams and prepared by aerosol assisted impregnation. Before hydrogen reduction, samples with 5 wt.% of Ni had submicron-sized, non-agglomerated, isolated NiCl₂ particles. Cracked NiCl₂ crust enveloped the whole foam surface in the case of 20 wt.% of Ni. Higher reduction degrees were reached with lower Ni loading at 533 and 633 K. Reduced Ni/Al₂O₃ and Ni-Pd/Al₂O₃ samples had similar microstructures for the same Ni content. With 5 wt.% of Ni, isolated, island-like, submicron-sized particles were formed. Sample with 20 wt.% of Ni had metallic coating without agglomerates and visible cracks that relatively evenly covered the whole foam surface. Ni-Pd/Al₂O₃ catalysts were nearly completely reduced at both temperatures. Their reduction degrees reached 99.4 and 98.2 wt.% for 5 and 20 wt.% Ni loading, respectively, at 533 K. Optimal metal loading in the produced catalysts is 20 wt.% of Ni and the addition of 0.1 wt.% Pd because the foam surface is relatively uniformly covered with metallic coating. Obtained results indicate that Ni-based catalysts, modified with Pd, can successfully be synthesized without the calcination step and can be reduced at lower temperature.

Acknowledgements

This work was financially supported by the Ministry of Education, Science and Technological Development of the Republic of Serbia and is a result of project No. 34033, Innovative synergy of by-products, waste minimization and clean technologies in metallurgy.

REFERENCES

- [1] Z. Anđić, M. Korać, M. Tasić, Ž. Kamberović, K. Raić, Synthesis and sintering of Cu-Al₂O₃ nanocomposite powders produced by a thermochemical route, *Metall. Mater. Eng.* **13** (2007) 71–81.
- [2] M. Korać, Ž. Kamberović, Z. Anđić, M. Filipović, Sintered Materials Based on Copper and Alumina Powders Synthesized by a Novel Method, *InTech, Rijeka*, 2011.
- [3] C. B. Alcock, *Thermochemical Processes: Principles and Models*, Butterworth-Heinemann, Oxford, 2001.
- [4] B.S. Kwak, J. Kim, M. Kang, Hydrogen production from ethanol steam reforming over core-shell structured Ni_xO_y-, Fe_xO_y-, and Co_xO_y-Pd catalysts, *Int. J. Hydrogen Energy* **35** (2010) 11829–11843.
- [5] A.J. Akande, R.O. Idem, A.K. Dalai, Synthesis, characterization and performance evaluation of Ni/Al₂O₃ catalysts for reforming of crude ethanol for hydrogen production, *Appl. Catal. A* **287** (2005) 159–175.
- [6] L.P.R. Profeti, J.A.C. Dias, J.M. Assaf, E.M. Assaf, Hydrogen production by steam reforming of ethanol over Ni-based catalysts promoted with noble metals, *J. Power Sources* **190** (2009) 525–533.
- [7] C.K. Cheng, S.Y. Foo, A.A. Adesina, Steam reforming of glycerol over Ni/Al₂O₃ catalyst, *Catal. Today* **178** (2011) 25–33.
- [8] M.A. Goula, S.K. Kontou, P.E. Tsiakaras, Hydrogen production by ethanol steam reforming over a commercial Pd/ γ -Al₂O₃ catalyst, *Appl. Catal., B* **49** (2004) 135–144.
- [9] J. Grosse, B. Dietrich, G. Incera Garrido, P. Habisreuther, N. Zarzalis, H. Martin, M. Kind, B. Kraushaar-Czarnetzki, Morphological characterization of ceramic sponges for applications in chemical engineering, *Ind. Eng. Chem. Res.* **48** (2009) 10395–10401.
- [10] M.V. Twigg, J.T. Richardson, Theory and applications of ceramic foam catalysts, *Chem. Eng. Res. Des.* **80** (2002) 183–189.
- [11] F. Ribeiro, J.M. Silva, E. Silva, M. Fatima Vaz, F.A.C. Oliveira, Catalytic combustion of toluene on Pt zeolite coated cordierite foams, *Catal. Today* **176** (2011) 93–96.
- [12] P. Ciambelli, V. Palma, E. Palo, Comparison of ceramic honeycomb monolith and foam as Ni catalyst carrier for methane autothermal reforming, *Catal. Today* **155** (2010) 92–100.
- [13] D.K. Liguras, D.I. Kondarides, X.E. Verykios, Production of hydrogen for fuel cells by steam reforming of ethanol over supported noble metal catalysts, *Appl. Catal., B* **43** (2003) 345–354.
- [14] Y. Zhang, Y.H. Tang, E.L. Zhang, L.W. Lin, L.Z. Pei, Preparation of Ni/MgO catalysts for carbon nanofibres by self-propagating low temperature combustion process, *Mater. Sci.-Poland* **28** (2010) 805–815.
- [15] A.N. Fatsikostas, D.I. Kondarides, X.E. Verykios, Production of hydrogen for fuel cells by reformation of biomass-derived ethanol, *Catal. Today* **75** (2002) 145–155.
- [16] K. Yoshida, K. Okumura, T. Miyao, S. Naito, S. Ito, K. Kunimori, K. Tomishige, Oxidative steam reforming of methane over Ni/ α -Al₂O₃ modified with trace Pd, *Appl. Catal., A* **351** (2008) 217–225.
- [17] P. Gayan, L.F. de Diego, F. Garcia-Labiano, J. Adanez, A. Abad, C. Dueso, Effect of support on reactivity and selectivity of Ni-based oxygen carriers for chemical-looping combustion, *Fuel* **87** (2008) 2641–2650.
- [18] M. Watanabe, H. Yamashita, X. Chen, J. Yamanaka, M. Kotobuki, H. Suzuki, H. Uchida, Nano-sized Ni particles on hollow alumina ball: Catalysts for hydrogen production, *Appl. Catal., B* **71** (2007) 237–245.
- [19] X. Wei, P. Hug, R. Figi, M. Trottmann, A. Weidenkaff, D. Ferri, Catalytic combustion of methane on nano-structured perovskite-type oxides fabricated by ultrasonic spray combustion, *Appl. Catal., B* **94** (2010) 27–37.
- [20] G. Matula, J. Bogović, S. Stopić, B. Friedrich, Scale-up of the Ultrasonic spray pyrolysis (USP) process for nanopowder production (Part 1), *PIM International* **7** (2013) 75–78.

- [21] B. Ebin, E. Arig, B. Ozkal, S. Gurmen, Production and characterization of ZnO nanoparticles and porous particles by ultrasonic spray pyrolysis using a zinc nitrate precursor, *Int. J. Min. Metall. Mater.* **19** (2012) 651–656.
- [22] S. Gurmen, B. Ebin, S. Stopić, B. Friedrich, Nanocrystalline spherical iron–nickel (Fe–Ni) alloy particles prepared by ultrasonic spray pyrolysis and hydrogen reduction (USP-HR), *J. Alloys Compd.* **480** (2009) 529–533.
- [23] V. Nikolić, Ž. Kamberović, Z. Anđić, M. Korać, M. Sokić, V. Maksimović, Influence of the synthesis method and the addition of modifiers on the properties of Ni-based catalysts supported on reticulated ceramic foams, *Int. J. Min. Metall. Mater.* (2014), DOI: 10.1007/s12613-014-0000-0.
- [24] M. Šušić, S. Stopić, I. Ilić, D. Uskoković, Kinetics of hydrogen absorption by nickel powder with added palladium, copper, and nickel from nickel-chloride reduction by hydrogen, *Int. J. Hydrogen Energy* **22** (1997) 661–667.
- [25] C. Amorim, M.A. Keane, Catalytic hydrodechlorination of chloroaromatic gas streams promoted by Pd and Ni: The role of hydrogen spillover, *J. Hazard. Mater.* **211–212** (2011) 208–217.
- [26] V. Nikolić, Ž. Kamberović, Z. Anđić, M. Korać, A. Vujović, M. Sokić, Alumina based catalytically active components carriers with improved properties, in: *Proceedings of the 44th International October Conference on Mining and Metallurgy, Bor, Serbia, 2012*, pp. 395–400.
- [27] V. Nikolić, Ž. Kamberović, Z. Anđić, M. Korać, M. Sokić, Synthesis of α -Al₂O₃ based foams with improved properties as catalyst carriers, *Mater. Tehnol.* **48** (2014) 45–50.

IZVOD

Katalizatori na bazi nikla: zavisnost svojstava od sadržaja nikla i modifikacije paladijumom

Vesna D. Nikolić¹, Željko J. Kamberović², Marija S. Korać², Zoran M. Anđić³, Aleksandar M. Mihajlović¹, Jelena B. Uljarević¹

¹Inovacioni centar Tehnološko–metalurškog fakulteta, Univerzitet u Beogradu, Beograd, Srbija

²Tehnološko–metalurški fakultet Univerziteta u Beogradu, Beograd, Srbija

³Inovacioni centar Hemijskog fakulteta, Univerzitet u Beogradu, Beograd, Srbija

(Naučni rad)

Cilj predstavljenog istraživanja bila je uporedna analiza svojstava katalizatora na bazi nikla, sa sledećim sadržajem metala: 5 i 20 mas. % Ni, bez dodatka i sa dodatkom Pd. Ni/Al₂O₃ i Ni–Pd/Al₂O₃ katalizatori su sintetisani metodom impregnacije potpomognute ultrazvučnim raspršivanjem. Kao nosači katalitički aktivnih materija korišćene su keramičke pene na bazi α -Al₂O₃. Uporedna analiza svojstava sintetisanih katalizatora obuhvatila je: određivanje stepena redukcije uzoraka u struji vodonika, praćenje razvoja mikrostrukture metodom skenirajuće elektronske mikroskopije i ispitivanje hemijskog sastava u tački metodom energetske disperzivne spektroskopije. Radi sinteze katalizatora, nosači odn. pene su predgrejane u cevnom reaktoru na 473 K i impregnirane ultrazvučno raspršenim rastvorima hlorida metala. Nakon impregnacije, uzorci su sušeni na 473 K radi dehidratacije, a zatim redukovani u struji vodonika na 533 i 633 K. Kod uzoraka sa nižim sadržajem Ni, dobijeni su viši stepeni redukcije. Svi uzorci sa dodatim Pd, kao modifikatorom aktivnosti, gotovo su u potpunosti bili redukovani na obe ispitivane temperature. Kod ovih uzoraka su na temperaturi od 533 K stepeni redukcije dostigli 99,4 i 98,2 mas.% za sadržaj Ni od 5 i 20 mas.%, redom. Optimalni sastav proizvedenih katalizatora je 20 mas.% Ni i dodatak od 0,1 mas.% Pd, čime se postiže relativno ravnomerna prekrivenost cele površine pene slojem metala. Dobijeni rezultati ukazuju na to da se katalizatori na bazi Ni, modifikovani dodatkom Pd, uspešno mogu sintetisati bez koraka žarenja i redukovati na izuzetno niskoj temperaturi (533 K).

Ključne reči: Katalizatori • Ni • Pd • Modifikovanje • Pena na bazi α -Al₂O₃ • Impregnacija aerosolom