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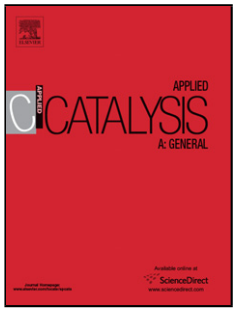

Title: The Roles of Co-Precipitation pH, Phase-Purity and Alloy Formation for the Ammonia Decomposition Activity of Ga-Promoted Fe/MgO Catalysts

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PII: S0926-860X(17)30432-5
DOI: http://dx.doi.org/10.1016/j.apcata.2017.09.004
Reference: APCATA 16398

To appear in: *Applied Catalysis A: General*

Received date: 14-6-2017
Revised date: 26-8-2017
Accepted date: 2-9-2017

Please cite this article as: Denise Rein, Klaus Friedel Ortega, Claudia Weidenthaler, Eckhard Bill, Malte Behrens, The Roles of Co-Precipitation pH, Phase-Purity and Alloy Formation for the Ammonia Decomposition Activity of Ga-Promoted Fe/MgO Catalysts, Applied Catalysis A, Generalhttp://dx.doi.org/10.1016/j.apcata.2017.09.004

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ACCEPTED MANUSCRIPT

The Roles of Co-Precipitation pH, Phase-Purity and Alloy Formation for the Ammonia Decomposition Activity of Ga-Promoted Fe/MgO Catalysts

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Highlights:

- Active Ga-promoted Fe/MgO ammonia decomposition catalysts are derived from co-precipitated hydrotalcite-like precursors
- Variation of pH strongly changes the catalytic properties due to differences in the reduction properties
- By-phases formed below pH 9.5 disturb the reduction compared to phase-pure samples
- Compositional uniformity of Fe-Ga nitrides with different Ga-contents greatly influences the activity

Abstract

A series of mesoporous $MgFe_{1.75}Ga_{0.25}O_4$ mixed spinel oxides obtained upon calcination of hydrotalcite-like precursors was investigated in the ammonia decomposition reaction at 1 atm after reduction in H_2 atmosphere. The corresponding precursors were synthesized from metal salt solutions at five constant pH values in the range between 8.5 and 10.5 by co-precipitation in aqueous media to study the impact of pH variation on the catalyst's structure and activity. N_2 physisorption, thermogravimetric analysis, powder X-ray diffraction, Mössbauer spectroscopy, and temperature programmed techniques (H_2-TPR and NH_3-TPD) were applied to gather information about the textural, (micro-)structural, and adsorption properties of the samples. While phase purity in the precursor and oxide stages is only observed for pH = 10, undesired by-phases ($MgFe_2O_4$ and/or Fe_3O_4) are additionally formed during co-precipitation at the remaining pH values. This is partly related to an incomplete precipitation of Mg^{2+} cations in less alkaline environments. *In situ* XRD measurements during reduction revealed that Fe-Ga alloys are formed between 500 and 600 °C. The absence of by-phases avoids the

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