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Ni-Co-B非晶态合金的结构和催化活性的理论研究

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摘要 利用一系列原子簇模型Ni~xCo~3~-~xB~2(x=1～3)对Ni-Co-B非晶态合金的电子结构进行了SCC-DV-Xα计算。结果表明,Ni-B非晶态合金的催化加氢活性可通过Co的引入而得到改进。结合EXAFS和活性试验的结果,得出Ni和Co对非晶态Ni-Co-B合金的结构的稳定性有一种协同作用的结论。

关键词 镍合金 钴合金 硼合金 催化活性 催化加氢 电子结构 协同效应

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Theorectical study on the structure and catalytic activity of Ni-Co- B amorphous alloy

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Abstract In the literature, there is little work could be found about the calculation of amorphous alloy, since the structure of amorphous alloy is ambiguous now. In this paper, a series of models were selected to reflect both the character of amorphous alloy and the change of cobalt content in Ni-Co-B amorphous alloy. These models were calculated SCC-DV-Xα method, the catalytic activity and the EXAFS structure of Ni-Co-B amorphous alloy could be interpreted by the calculation results very well. The calculated results showed that the relationships between the Fermi energy and the density of state near Fermi level to cobalt in Ni-Co-B amorphous alloy are also present in a volcano-shape curve. It could be inferred from the calculation that the FMO energy of catalysts will be most close to FMO of H~2 when the contents of cobalt and nickel in the Ni-Co-B amorphous alloy are near. In the reaction of catalytic hydrogenation, the H-H bond would be weakened by the transfer of the electron from HOMO of catalyst to the LUMO of hydrogen. Thus the closer of these two orbital energy levesl, the higher the catalyitc activity of the catalyst. The volcano-shape curve of the catalytic activity versus their cobalt content is due to the volcano-shape of their Fermi level versus their cobalt content. It is known that the amorphous-orming ability and the interaction of metal-metalloid are related to it's Fermi level and density of states near Fermi level. Our calculation also shows that the amorphous-forming ability is strongest when the contents of Ni and Co are near in the Ni-Co-B amorphous alloy and meantime Ni-B interaction is strongest. All the results confirmed that there is a synergism of Ni and Co to the structure of Ni-Co-B amorphous alloy and certainly these results will give a new idea to the design of novel catalyst.

Key words NICKEL ALLOYS COBALT ALLOYS BORON ALLOY CATALYTIC ACTIVITY CATALYTIC HYDROGENATION ELECTRONIC STRUCTURE SYNERGISTIC EFFECT

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