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**Interaction of A^V₂B^{VI}₃ semiconductor compounds
with metals**

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It is shown that as well as in the cases of II – VI, III – VI, IV – VI and I – VI group semiconductors the comparison of experimental results about phase relations of A^V₂B^{VI}₃ semiconductor compounds with metals and given thermodynamic calculations testifies that the calculations of thermodynamic Gibbs potential changes in the systems A^V₂B^{VI}₃ – Me allow to determine the presence or absence of exchange interaction in each particular system with sufficient reliability. Such estimation becomes more reliable at the simultaneous examination of the phase diagrams of binary systems limiting each particular ternary system and taking into account the stability of all existing binary and ternary phases. The reasons of discrepancies between experimental data and thermodynamic calculations are discussed.

Keywords: interaction, thermodynamic calculation, semiconductor compounds, metals, discrepancy.

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The metals are widely used in the semiconductor material sciences for the contact forming, as the solvent for single crystals growth of refractory semiconductor compounds and their solid solutions and for the doping of semiconductors. But their practical using is bounded and complicated in case of chemical interaction of metal with semiconductor. Therefore the problem of chemical interaction in the systems «semiconductor – metal» is one of the main problems of semiconductor material sciences.

Chalcogenides of Group VA elements have good thermoelectric properties and are used at the manufacturing of thermoelectric devices of a good quality [1]. While operating, a working semiconductor element is always in contact with current-carrying metal units; at the contact spot various phisycocochemical interactions can occur to form eutectics, solid solutions, or new phases [2-4]. The nascent compounds can differ in properties from both the semiconductor and the metal, causing pore formation and cracking in the contact area.

The selection of commutation materials that would be chemically inert toward the semiconductor and provide stable performance is a challenging problem in semiconductor engineering. To make the selection, the engineer should understand the character of the interaction of semiconductor with various metals.

The interaction of other semiconductor chalcogenides with metals were generalized in our previous works [5-11]. This paper is devoted to the systematization and generalization of phase equilibria in the A^V₂B^{VI}₃ – Me systems and comparison of existing experimental data with computation of Gibbs energy in the next reactions:



The computation of Gibbs energy was performed taking into account that AsS (congruent melting) and AsSe, SbTe, BiSe and BiTe (incongruent melting) are formed also in the V – VI binary system:



The Table 1 compiles all experimental data

available on the character of physicochemical interaction of arsenic, antimony and bismuth sesquichalcogenides with metals; they are labeled with asterisk. This table also includes our predictions of exchange interaction based on our computations of the Gibbs free energy by using the procedure in [6]. Compounds included in the thermodynamic calculations are specified in the Table 1 also. For Ga, Nd, Sm, Gd, Dy, Fe and Ni the Gibbs free energy of exchange reactions were calculated with allowance for the formation of GaB^{VI} , $\text{Nd}_2\text{B}^{\text{VI}}_3$, $\text{Sm}_2\text{B}^{\text{VI}}_3$, $\text{Gd}_2\text{B}^{\text{VI}}_3$, $\text{Dy}_2\text{B}^{\text{VI}}_3$, FeB^{VI}_2 and NiB^{VI}_2 respectively. Identical values were obtained with allowance for the formation of $\text{Ga}_2\text{B}^{\text{VI}}_3$, NdB^{VI} , SmB^{VI} , GdB^{VI} , DyB^{VI} , FeB^{VI} and NiB^{VI} (see Table 1).

Examination of experimental data indicates that at the interaction of $\text{A}^{\text{V}}_2\text{B}^{\text{VI}}_3$ semiconductor compounds with copper and silver Cu(I) and Ag(I) chalcogenides are formed [12-16] and the interaction of gold with As_2Te_3 leads to the formation of AuTe_2 [17]. Gallium monotelluride is formed at the interaction of bismuth sesquitelluride with gallium [20]. Interaction of rare earth elements with arsenic, antimony and bismuth sesquichalcogenides leads mainly to the formation of corresponding lanthanide monotelluride [26-39, 41-44] and only at the interaction of gadolinium with As_2S_3 Gd(III) sulfide is formed [40]. The interaction of Bi and As_2S_3 leads to the formation of Bi_2S_3 [45], Co and Bi_2Te_3 – to the formation of BiTe [51] and Ni and Bi_2Te_3 – to the formation of Ni_3Te_4 [49]. The compounds both of FeB^{VI} -type (Sb_2Se_3 -Fe system) and FeB^{VI}_2 -type (Sb_2Te_3 -Fe system) can form in the case of iron interaction with $\text{A}^{\text{V}}_2\text{B}^{\text{VI}}_3$ semiconductor compounds [46-48]. The other of the experimentally studied $\text{A}^{\text{V}}_2\text{B}^{\text{VI}}_3$ -Me systems are quasi-binary [As_2Te_3 -Tl, Sb_2Te_3 -Au(Yb, Fe, Co), Bi_2Se_3 -Au and Bi_2Te_3 -Tl(Fe)] (Table 1).

The Gibbs free energies were calculated for all of the $\text{A}^{\text{V}}_2\text{B}^{\text{VI}}_3$ -Me systems studied, except for gold sulfide and selenide systems (their thermodynamic properties have not been determined adequately). Although only one of the nine arsenic, antimony and bismuth sesquichalcogenide systems of the type

$\text{A}^{\text{V}}_2\text{B}^{\text{VI}}_3$ -Me have been studied experimentally, we calculated exchange interactions for all nine such systems. Having done this, we are able to predict the character of physicochemical interaction in systems of this type.

Up to now, 36 of the $\text{A}^{\text{V}}_2\text{B}^{\text{VI}}_3$ -Me systems have been studied experimentally and only for two systems (As_2Te_3 -Au and Sb_2Te_3 -Yb) there are discordances between experimental results and thermodynamic calculations. It is necessary to note that both these systems are needed additional experimental investigations.

Thus, the occurrence or absence of exchange chemical reactions for arsenic, antimony and bismuth sesquichalcogenides with metals can reliably be predicted based on temperature variations of the Gibbs free energy with allowance for the character of interaction of the components of the boundary binaries of a ternary system.

Taking into account the obtaining results and the data of earlier published our papers [5-11] it is possible to analyse the reasons of discrepancies between the experimental data and thermodynamic calculations in the systems «chalcogenide semiconductor – metal». The number of experimentaly investigated systems and the number of discrepancies between experimental data and thermodynamic calculations are given in the Table 2. The next reasons can explain the existing discrepancies.

1. The conditions of experiment realization (the open or closed system). This reason explain the existing discrepancies in the systems ZnSe -Cu, CdSe -Cu(Ga) and Cu_2Se -Zn(Cd);

2. The formation of ternary compounds, intermetallides and metal chalcogenides for wich the thermodynamic properties are absent. This causes the discrepancies for the systems GeTe -As (formation of GeAs_2), GeTe -Co (GeCo_2), SnTe -Ni (Ni_3Sn_2), PbTe -Cr (CrTe or Cr_3Te_4 but not Cr_3Te_2), PbTe -Ni (Ni_3Te_2 but not NiTe), Ga_2Te_3 -Co (Co_3Te_4 but not CoTe), Ga_2Te_3 -Ni (Ni_3Te_4 but not NiTe), Tl_2S -Bi (Tl_2Bi_3 , $\text{Tl}_4\text{Bi}_2\text{S}_5$, TlBiS_2), Ga_2S_3 -Pb (PbGa_2S_4), Ga_2Te_3 -Sb(Bi) (GaTe but not Ga) and InSe-Sb (InSb , In_2Se_3);

Table 1.
Interaction of $A^V_2B^{VI}_3$ semiconductor compounds with metals

	As_2S_3	As_2Se_3	As_2Te_3	Sb_2S_3	Sb_2Se_3	Sb_2Te_3	Bi_2S_3	Bi_2Se_3	Bi_2Te_3
Cu (Cu_2B^{VI})	+ + [12]	+	+	+	+	+	+	+	+
Ag (Ag_2B^{VI})	+	+	+	-/+ + [14]	+	+	-/+ + [15]	+	+
Au (AuB^{VI}_2)			- + [17]			- [18]		- [19]	-
Ga ($Ga_2B^{VI}_3$)	+	+	+	+	+	+	+	+	+ [20]
Tl (Tl_2B^{VI})	+	+	+	+	+	+	+	+	+ - [24, 25]
La (LaB^{VI})	+	+	+	+	+	+	+	+	+ + [26]
Nd (NdB^{VI})	+	+	+	+	+	+	+	+	+ + [29]
Sm (SmB^{VI})	+	+	+	+	+	+	+	+	+ + [38]
Eu (EuB^{VI})	+	+	+	+	+	+	+	+	+
Gd (GdB^{VI})	+	+	+	+	+	+	+	+	+
Dy (DyB^{VI})	+	+	+	+	+	+	+	+	+ + [41, 42]
Tm (TmB^{VI})	+	+	+	+	+	+	+	+	+
Yb (YbB^{VI})	+	+	+	+	+	+	+	+	+
Bi ($Bi_2B^{VI}_3$)	+/- + [45]	+/-	+/-	-	+/-	+/-			
Fe (FeB^{VI})	+/-	+/-	+/-	+/-	+/- + [46]	+/- - [47, 48]	+/-	+/-	+/- - [49]
Co (CoB^{VI}_2)	+/-	+/-	+/-	+/-	+/-	+/- - [50]	+/-	+/-	+/- + [51]
Ni (NiB^{VI})	+/-	+/-	+/-	+/-	+/-	+/-	+/-	+/-	+/- + [52, 53]

Note: + means that a chemical interaction occurs; - means that no chemical interaction occurs; and +/- means that a chemical interaction occurs at low temperatures and does not occur at high temperatures. The compounds included in thermodynamic calculations are parenthesized.

3. Contradictory information about congruent or incongruent melting of some chalcogenides (systems with GeTe and GeSe participation);

4. Formation in the ternary systems extensive region of solid solutions ($Pb_xSn_{1-x}Se$ solid solutions in the Pb-Sn-Se system);

5. Partial experimental investigations of

Table 2.

Discrepancies between experimental results and thermodynamic calculations of Gibbs energy for the system «chalcogenide semiconductor – metal»

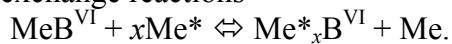
System	Investigated	Discrepancies	Reference
A ^{II} B ^{VI} -Me	50	45	[5, 7]
A ^{IV} B ^{VI} -Me	76	63	[6, 7]
A ^{III} ₂ B ^{VI} ₃ -Me	27	18	[7, 8]
A ^{III} B ^{VI} -Me	35	30	[7, 10]
Tl ₂ B ^{VI} -Me	17	15	[7, 9]
A ^V ₂ B ^{VI} ₃ -Me	36	34	This work
A ^I ₂ B ^{VI} -Me	45	41	[11]
Total	286	246	[5-11]

some systems [SnTe-W, In₂Te₃-Mg(Zn, Cd, Fe), InTe-Sb, Tl₂Se-Ag, As₂Te₃-Au(Tl), Sb₂Te₃-Yb, As₂Se₃-Tl and AuTe₂-As systems)];

6. Uncertain thermodynamic parameters for some chalcogenides [GaS(InSe, InTe)-Mn systems; uncertain values of thermodynamic parameters for manganese chalcogenides)].

Thus, the occurrence or absence of chemical interaction in a particular «chalcogenide semiconductor – metal» system can be predicted unambiguously by simulations of

temperature variations of the Gibbs free energy of exchange reactions



The prediction will be more reliable if the thermodynamic calculations will be complemented by an analysis of phase diagrams of the boundary binaries of every ternary system and the stabilities of all existing binary and ternary phases will be accounted for.

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Взаємодія напівпровідниківих сполук типу A^V₂B^{VI}₃ з металами

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Показано, що, як і у випадку напівпровідниківих сполук типу A^{II}B^{VI}, A^{IV}B^{VI}, A^{III}B^{VI}, A^{III}₂B^{VI}₃, Tl₂B^{VI} і A^I₂B^{VI}, порівняння експериментальних результатів про фазові рівноваги в системах A^V₂B^{VI}₃ – метал та даних проведених термодинамічних розрахунків свідчить про те, що розрахунки зміни потенціалу Гіббса у вказаних системах дозволяють визначити наявність або відсутність обмінної взаємодії в кожній конкретній системі з достатньою надійністю. Надійність такої оцінки значно підвищується, якщо одночасно проаналізувати діаграми стану бінарних систем, що обмежують конкретну потрійну систему та врахувати стабільність всіх існуючих бінарних та потрійних фаз. Обговорюються причини неспівпадінь термодинамічних розрахунків з експериментальними результатами.