NEUTRON ACTIVATION ANALYSIS OF MANGANESE-MERCURY TELLURIDE

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Introduction

The triple semiconductor compound $Mn_xHg_{1-x}Te$ is a worthwhile material for the development of infra-red detectors. It's properties, as well as other semiconductors ones depend on impurity elements content. So, analytical Purity control of $Mn_xHg_{1-x}Te$ is required. Some procedures of the neutron activation analysis of Te[1], Hg [2] and that of their compounds ($Cd_xHg_{1-x}Te$ [3], for instance) have been developed recently. However, no papers on the NAA of $Mn_xHg_{1-x}Te$ have been found.
This paper describes the procedure of the NAA of $Mn_xHg_{1-x}Te$ based on anion-exchange chromatographic separation of impurity and matrix elements.
Experimental
The following reagents were used: commerciality available strongly based anion-exchange resin AW-17 (50-100 meash), hydrochloride hydrasine of analytical grade, solutions of HCl and HNO3 prepared from concentrated acids of reagent grade.
Separation processes were studied by radioactive tracers. The gamma-activity was measured on a HPGe detector GC1518 (efficiency 15 %, resolution 1,7 keV at 1332,5 keV line of ^{60}Co) using DSA-1000 digital mulichannel analyzer (Canberra, USA).
Radioactive tracers were produced by irradiation of metals, salts or oxides of corresponding elements in WWR-SM water-water nuclear reactor.
Irradiation of $Mn_xHg_{1-x}Te$ samples and standards was carried out in a channel of WWR-SM reactor with a neutron flux density of 1.10^{14} $cm^2.s^{-1}$ for 10 h.
Solutions of manganese and mercury were prepared by dissolving their oxides in concentric hydrochloric acid. Solutions of tellurium were prepared by dissolving metallic tellurium in $HCl:HNO_3$ (3:1) mixture. Solution obtained was evaporated to dryness, the reside was dissolved in conc. HCl. To reduce tellurium to Te(IV) 1-2 mg of hydrazine were dissolved in solution at slight heating. Then, distilled water was added in a required quantities to get a desired concentration of HCl.
The chromatographic behaviour of matrix and impurity elements was studied using chromatographic columns, filled with AW-17 resin (columns' i. d. 1.2 cm , resin layer height 10 cm, mobil phase volume 5.5 cm^3), prepared according to recommendations of [4]. 10 ml of the solution containing 61 mg of Te, 86 mg of Hg and 2.6 mg of Mn (That corresponds to 150 mg of $Mn_xHg_{1-x}Te$) and radioactive tracers were placed into chromatographic column and then impurities were eluted with 4 M HCl. Eluation rate was about 0.5-0.6 ml/min. Eluated was collected in a portions of 1 ml, followed by measuring it's gamma-activity.
To study the distribution of matrix radionuclides along the column their activity was measured using a lead collimator.

Results and Discussion

A number of long – lived radionuclides of manganese, tellurium, mercury as well as those of iodine and antimony are formed during the irradiation of $Mn_xHg_{1-x}Te$ samples by neutrons in nuclear reactor (Table 1). Specific activity of these radionuclides exceeds 10^7-10^9 Bk/g. So,

realization of instrumental neutron activation analysis of $Mn_xHg_{1-x}Te$ is difficultly and radionuclides of matrix elements should be separated from that one of impurities.

Table 1 Nuclear-physical property of Mn, Hg, Te

Radionuclide	**Nuclear reaction**	**Half-life**	**Energy, keV**
^{56}Mn	$^{56}Mn(n,\gamma)$	2,58h	846,9; 1238,2; 1810,7
^{54}Mn	$^{55}Mn(n,\gamma)$	312d	834,8
^{121}Te	$^{120}Te(n,\gamma)$	16,8d	470,5; 507,6; 573,1
^{123m}Te	$^{122}Te(n,\gamma)$	120d	159
^{125m}Te	$^{124}Te(n,\gamma)$	58d	35,5; 109,3
^{127m}Te	$^{126}Te(n,\gamma)$	109d	57,6
^{129}Te	$^{128}Te(n,\gamma)$	1,16d	27,8; 459,6; 487,4
^{129m}Te	$^{128}Te(n,\gamma)$	33,6d	695,9
^{131}Te	$^{130}Te(n,\gamma)$	24,8min	149,7; 452,3; 492,7; 602; 654,3; 948,5; 997,2
^{131m}Te	$^{130}Te(n,\gamma)$	1,25d	102,1; 200,6; 240,9; 334,3; 773,7;782,5; 793,8; 852,2; 1125,5; 1206,6
^{131}J	$^{131}Te \rightarrow \beta^-$	8,04d	80,2; 284,3; 364,5; 637; 722,9
^{122}Sb	$^{122}Te(n,p)$	2,71d	564; 692,6
^{124}Sb	$^{124}Te(n,p)$	60,2d	602,7; 645,8; 713,8; 722,3; 968,2; 1045,1; 1325,5; 1368,2; 1691
^{197m}Hg	$^{196}Hg(n,\gamma)$	2,67d	77,4; 191,4
^{197}Hg	$^{196}Hg(n,\gamma)$	23,8h	133,9
^{203}Hg	$^{202}Hg(n,\gamma)$	46,6d	279,2

Preliminary estimation, based on measuring of matrix radionuclides activity, shoos that separation factors should not be less then 10^7 for tellurium, 10^9 for iodine, 10^9 for mercury.
Half – life of ^{56}Mn (2,58 h) allows one to decrease it's activity by cooling of irradiated sample for 2-3 days. Activity of ^{54}Mn negligible due to small activation cross – section of $^{55}Mn(n,2n)\ ^{54}Mn$ reaction. Besides, total amount of manganese in $Mn_xHg_{1-x}Te$ is rather small, considering x is used to be about 0,1.
Therefore, radiochemical procedure should ensure separation of impurity elements from tellurium, mercury, iodine and antimony. The most convenient method for such effective separation is chromatography.
It is well known [4], that mercury, iodine and antimony (V) form chloro-complexes, which are readily sorbed by strongly-based anion-exchange resin AW-17 (it's properties are analogues to those of Dowex 1 resin) from 3-9 M HCl. Distribution ratious for these elements exceed 10^2, while many impurities under this conditions are poorly sorbed (D<1). So, there exists a principal possibility of anion-exchange chromatographic separation of radionuclides of impurities from that one of matrix, listed above.
Distribution ratious of some elements in AW-17 – HCL system are depending on hydrochloric acid concentration, so separation conditions have been experimentally studied to ensure the most selective and effective separation.

As follows from date, published in [4],the most selective separation of mercury and telluriym from other elements can be carried out when 1 M HCL is used. In this case mercury and tellurium can be separated from Na, K, Cs, Ba, Sc, rear-earth elements, Cr, Fe, Co, Ni, Cu, Ga, As, Zr, Hf. But our experimental data differ from that one of [4]. Namely, it has been found, that D_{Te} is strongly depending on Te and HCL concentration (Fig.1).It is in our opinion – due to hydrolyzes of tellurium at 1-2 M HCL.

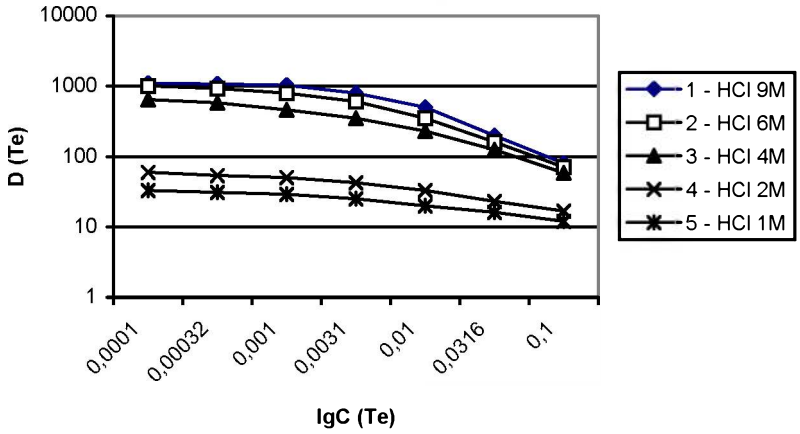

Fig. 1 The distribution coefficient of Te by AW-17 sorption from HCL

Distribution ratio for Sb(V) increases while HCL concentration increases, and it reaches 10^2 when C_{HCL}=4 M, it provides effective separation.

According to mentioned above, concentration of hydrochloric acid should not be less then 4 M. This concentration seems to be close to an optimal one, because at highest values of C_{HCL} the distribution coefficient of Co, Cu, As, Zr, Hf exceed and their separation from matrix becomes labored.

Elution curves of impurity elements are shown at Fig.2. Matrix radionuclides (except ^{54}Mn) completely retained by column (Fig.3). The separation factors of impurities from mercury, tellurium and antimony exceed 10^9.

The major part of iodine is removed during evaporation of solution of investigated sample. The residue of iodine, as well as ^{131}J formed from ^{131}Te during chromatography separation, is almost completely retained in the column. Also, a small amounts of iodine, slightly contaminated concentrate of impurities, can removed by distillation during preparation of eluate for gamma-activity measurements.

Thus, anion-exchange chromatography can be used to separate simultaneously a number of impurities from macroamounts of Hg, Te and their daughter radionuclides of latter (^{122,124}Sb, ^{131}J) in NAA of $Mn_xHg_{1-x}Te$.

The following NAA procedure for manganese-mercury telluride has been developed on basis of the investigations performed.

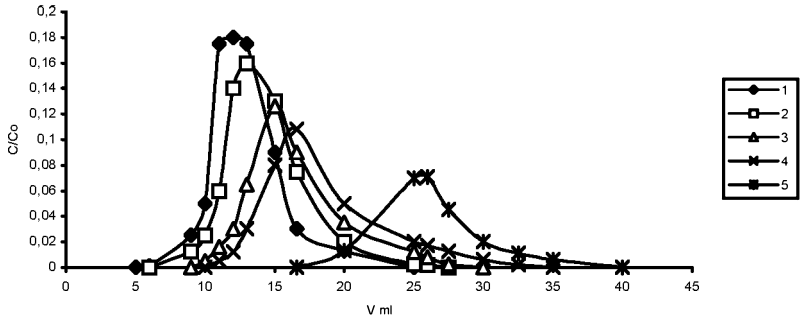

Fig. 2. Elution curves of impurity elements at the presence of 150 mg $Mn_xHg_{1-x}Te$.
1 – Na, K, Cs, Rb, Ba, La+Ln; 2 – Cr, Mn; 3 – Co, Hf; 4 – As; 5 – Cu

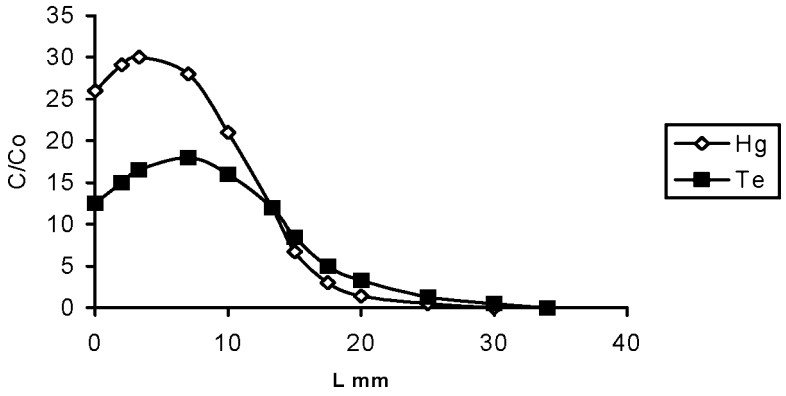

Fig.3 The macrocomponents distribution by length chromatography column after doped element eluation.

Procedure of analysis.

The $Mn_xHg_{1-x}Te$ sample (150-200 mg) and the standards of the elements to be determined are irradiated in the channel of water-water reactor for 10 h. After irradiation the sample is etched in some portions of concentrated HCL and HNO_3 mixture ($HCl:HNO_3$=3:1) and distilled water. Then the sample is transferred into a distillation apparatus, the carriers of elements to be determined (10^{-4}-10^{-5} g) are added, and the sample is dissolved in 10 ml of concentrated HCL and HNO_3 acids ($HCl:HNO_3$=3:1) mixture. The solution obtained is evaporated to a dry salts, adding 2 or 3 portions (1 ml each) of concentrated HCL at the end of drying to remove the rest of nitric acid. The dry residue is dissolved in 4 ml of 10 M HCL, and 1-2 mg of hydrazine is dissolved in solution at slight Heating to reduce tellurium to Te(V). Then 6 ml distilled water is added and solution is placed into chromatography column with AW-17 resin (resin layer height is 10 cm, column diameter 1,2 cm) prewashed with 40 ml of 4 M HCL. Sample solution is passed through column at 0,5-0,6 ml/min rate and impurities are eluted

with 40 ml of 4 M HCL at the same rate. Next, 5-10 mg of potassium iodide and 5-10 ml of concentrated nitric acid are added to eluate and the latter is evaporated to 5 ml volume, following by gamma-activity measurements.

Radioactive tracers experiments showed the chemical yields for all elements to be determined were 90-95 %, the relative standard deviation of determination, S_r, were about 0,15-0,25. The detection limits of impurities in procedure developed are listed in Table 2.

The procedure suggested can also be used in NAA of Hg, Te and HgTe.

Table 2. Detection limits of impurities in NAA of $Mn_xHg_{1-x}Te$.

Element	Detection limit, % weight	Element	Detection limit, % weight
Na	$5 \cdot 10^{-8}$	Cs	$3 \cdot 10^{-7}$
K	$2 \cdot 10^{-6}$	Ba	$8 \cdot 10^{-6}$
Sc	$5 \cdot 10^{-9}$	La	$6 \cdot 10^{-8}$
Cr	$3 \cdot 10^{-6}$	Ce	$6 \cdot 10^{-7}$
Co	$1 \cdot 10^{-7}$	Sm	$7 \cdot 10^{-10}$
Ni	$1 \cdot 10^{-5}$	Eu	$8 \cdot 10^{-10}$
Cu	$9 \cdot 10^{-8}$	Gd	$5 \cdot 10^{-7}$
As	$4 \cdot 10^{-8}$	Tb	$4 \cdot 10^{-7}$
Rb	$1 \cdot 10^{-5}$	Dy	$3 \cdot 10^{-7}$
Zr	$8 \cdot 10^{-6}$	Hf	$8 \cdot 10^{-8}$

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