THERMODYNAMICAL MODELING OF SLAG SYSTEMS FOR THE COMBUSTION PROCESSES OF A FUEL FROM WASTE

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Introduction

In a light of the progressing tendency to globalization and integration the binding point is an environmental protection in the each region, so that the local safety measures for Atmosphere Air and World Ocean are becoming the most importance. According to a theory of the sustainable development for a national economy in terms of the permanent international cooperation the practical aspects for energy and resource saving are necessary also in the branch of the administration for municipal solid waste (MSW) treatment and recycling.

It is evident [1-3], that the limited doses for the deposition in the ground are reachable only after the thermal incineration. We regard the MSW as an energy bearer, which could be ignited and burned without additives of fuel. Thus for the reducing of CO2- pollutions we require the “green” propellant, which substitutes the natural ones. The result of temperature influence is slag and it is a filling material for the building tools. In the case of fly ash (the products of a filter separation with high content for water soluble salts of the toxic metals) we expect the glassed and cemented granulate.

1. Metal containing rest substances (MeRS)

1.1 Fuel from waste

We start with an elementary content of MSW, which is used as an initial compound for a treatment in incineration plants. It includes 25 mass % of coal, 4% of hydrogen, 25% of inert materials and 30% of water. 12-15% of oxygen are fixed in an inflammable component and near 0,5% of nitrogen presented in different species.

The enthalpy of MSW increased in the last time because of an input for synthetic materials in the consume circulation for industrial countries. We talk about the igniting mix of 10 MJ/kg and this value grows together with the separation of rest substances. In [1] they are energy fractions of paper, gummy, plastics and also glass, metals and biomass.

The minimal, maximal and median values for total concentrations of elements in a fuel from waste are drawn in the table 1 with the quantity of regarded experiments N.

Table 1: Statistical evaluation of waste probes in mg/kg

Element	Min	Max	Median	N
K	841	21130	8608	82
Ca	1596	59441	38654	82
Ti	18	2475	1271	82
Cr	30	301	166	82
Mn	57,4	1442	238	82
Fe	316	18592	7327	82
Co	4,9	149	31,3	78
Ni	18,3	254	70,9	82
Cu	32,2	1549	98,7	82
Zn	46,4	2026	487	82
As	1,85	62	7,64	47
Mo	1,71	24,3	8,69	58
Cd	2,52	21,5	7,12	55
Sn	8,13	1042	25,5	60
Sb	6,39	117	18,4	59
Tl	2,49	36,4	6,17	25
Pb	33,8	7795	381	60

Covering near 90% of the global outcome lignin cellulose is the most popular type of bio-fuel. Its pyroluse is directed to the substitution of energetic compounds such as H2 and CO. [2] differ the bio-masse of two categories:

1- traditional (non commercial) quantitative with wood and char-coal, straw, hay, field rests, leaves and dung;
2- modern (fossil) qualitative with MSW.

Domestic rests consist of 20-30 volume % for artificial substances and they are responsible for only 6-12 mass % of the common stream [3]. Some of them can inherit the Cl and Br containing components. In the case of chlorine polyvinylchloride (PVC) is widely used in a production of roll materials on the textile basis. In the fraction of plastics 5-7 mass % of PVC cover near 40-50% of general Cl volumes [3]. Bromine is imputed in the compounds for fire suppressing. Such waste is characterized with the high enthalpy > 30 MJ/kg. Furthermore synthetic tools include the metal essences, pigments and catalyses, the presence of which is the source of heat and toxic complexes during co-combustion.

Thus MSW is the mix fuel, according to energetic characteristics which is similar [4] to brown coal. For example, if the efficiency coefficient for an electricity production is equal 21% in the plant with boiler, so the result of waste combustion is exceeded the own requirements in the 5-6 times. But during its using we should pay attention to the ecological meaning of the energy translation method. The progress of industrialization proposes us not only the Goods of new artifacts for consumption, but threatens with danger of their influence on environment. Solving in water medias metals develop double-face of their toxity in form of cations and oxy-anions. One of the most aggressive ones is thallium, which joins MSW from special alloys, semi-conductors and veterinary poisons. In the industrial waste it follows from technological slurry of lead production. Substances of Tl take place in color glasses, signal rackets and anti-detonation additives. Di-thallium trioxide is the mineral avicenite.

1.2. Bottom ash

The most important slag ingredients after thermal treating are [5] oxygen, silicium, iron, calcium, aluminium, sodium, potassium and coal. The total concentrations for each of them is more than 10 g pro kg of bottom ash. The secondary elements are presented from 1 till 10 g/kg. They are magnesium, zinc, cuprum, lead, titanium, manganese and chrome. Tin, vanadium, molybdenum, nickel, cobalt, cadmium, thallium, arsenic, antimony and mercury leave only a sigh and are less than 1 g/kg. In the table 2 the elementary content is investigated in the N probes (mg/kg) of TAMARA [6] combustion substances for MSW.

Table 2: Statistical evaluation of slag ingredients

Element	Min	Max	Median	N
K	13008	25532	18157	42
Ca	74255	14022	99179	42
Ti	3160	6848	4471	42
Cr	–	–	–	–
Mn	333	1714	647	42
Fe	14503	27241	20795	42
Co	2,55	204	74	42
Ni	56,3	277	99	42
Cu	355	2712	539	42
Zn	915	2943	1491	42
As	1	80,8	24,1	38
Mo	6,89	104	19,9	42
Cd	–	–	–	–
Sn	37,4	332	72,4	42
Sb	22,3	2302	65,2	41
Tl	7,69	2518	17,2	10
Pb	423	2171	1098	40

Since the aim of the temperature influence for waste is the production of building tools with no difference in their chemical composition from natural grounds, so the primary measures in the burning chamber are directed to the vaporization of toxic compounds and their accumulation in the filter ash. This effect could be reached as a result of the staged input of the technical air and the additives of chlorine, bromine and sulphur containing species. We become the higher level of slag quality, the practical assessment of which should explain the table 3.

Table 3. Slag of the different thermal processes in compare with cement and main vulcanic earths

Mass. %	Portland cement [10]	Metallurgy slag [8]	Coal combustion slag [7]	Al production [10]	Bottom ash [4]	Fly ash [11]	Ultrabase	Base	Acid
							Vulcanite [4]		
SiO_2	21,0	6,5	52,92	20,24	43,25	39-58	44,07	49,45	54,39
TiO_2	0,3	–	–	6,22	1,83	–	2,86	1,8	1,29
Al_2O_3	5,03	32	27,51	29,1	11,25	9-12	13,69	15,35	15,74
Fe-O	2,83	–	11,27	24,85	6,58	4-6	12,34	12,23	9,6
Mn-O	0,06	–	–	–	0,29	–	0,19	0,15	0,18
MgO	2,0	6,5	2,65	0,124	2,88	1,3-1,6	10,53	7,71	4,99
CaO	63,9	53	5,63	3,54	25,36	11-13	10,67	11,01	8,8
Na_2O	0,24	–	–	14,25	3,43	1,2-1,9	3,28	2,57	2,95
K_2O	0,65	–	–	0,118	2,23	2-3	1,67	0,52	1,04
P_2O_5	0,16	–	–	0,155	2,9	–	0,71	0,21	0,21
S	3,0	2	–	0,523	–	2,1-2,5	–	–	–
Cl	–	–	–	0,142	–	1,5-1,7	–	–	–

During the treating of MSW wollastonite and other silicates are building in the slag after the isothermal crystallization at 1400°C. The phase equilibrium is characterized (see figure 1) with gehlenite and anortite, aluminates $CaAl_2O_4$, $Ca_3Al_2O_6$, $CaAl_4O_7$ and mullite at the high content of Al_2O_3.

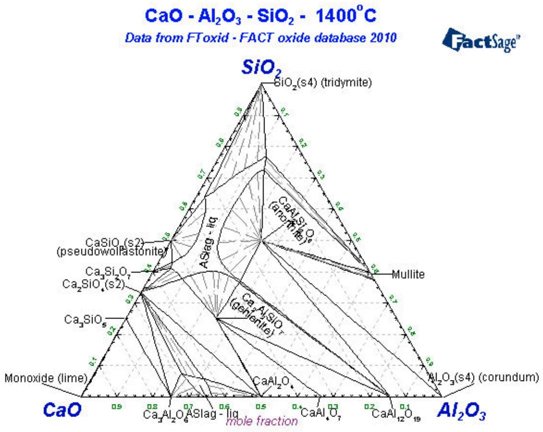

Figure 1: Calculations of equilibrium concentrations for fly and bottom ashes

In the work [4] the melilite were noticed as the main mineral too. Its composition is compared with pyroxenes in the table 4.

Table 4: Composition of mix crystals

Oxides	Melilite	Pyroxene
Na_2O	3,21	–
MgO	7,80	12,93
Al_2O_3	4,07	4,82
SiO_2	40,2	43,91
K_2O	–	0
CaO	38,94	28,63
FeO	–	–
ZnO	2,65	0,22
Fe_2O_3	1,01	3,59
Cr_2O_3	–	0,7
P_2O_5	0,58	0,42

Seok [7] calculated mullite at 1440 °C. The mole brakes increased with cooling of the system till 1200 °C, at which the appearance of silica, anortite and corderite took place. 980 °C it didn't exist any more.

It is high time to conclude the processes running in the chemical reactor named the waste incineration plant. Above we noticed the local transformation cases for fuel with help of additives to increase the heat effect and to destruct some complex existences in chlorides, bromides and sulphates. But except of these the global thermodynamic potential takes place in the test facility and it initiates the flying of metal substances from molten body. Kjellquist [8] calculated an accident of the purification for Fe-C alloys in slag and proved the fact of decreasing for the Al concentration in the steel during the contact with ash. In the thermal treating that means the diffusion of toxic species in the peripheral area, where the product quality is less than the conventional one because of excess for additional components.

We consider that the building of mix crystals with an unchangeable content of main minerals takes place in the burning chamber and they inject the secondary compounds. In terms of this work we regard thallium in the group of aluminium. The analogy of disposition in the periodic table of elements and development of chemical properties gives us a chance of thermodynamic modeling for essences, which include this metal in their composition melted in the silicate or the cement matrix – like these $(Al\text{-}Tl)_6Si_2O_{13}$, $Ca(Al\text{-}Tl)SiO_6$, $Ca_2(Al_{0,67}\text{-}Tl_{0,67})SiO_6$, $Ca(Al_{0,33}\text{-}Tl_{0,33})Si_2O_6$ или $Ca_2(Al\text{-}Tl)SiO_7$, $Ca(Al\text{-}Tl)Si_2O_8$.

1.3 Fly ash and water soluble species

We divide the gas phase in two parts: Filter separations are the condensed at 300 K existences and the essences, which are passing the acid and water washers. The elementary content of MeRS is in the table 5 [9] and the chemical composition is in the table 3 [11].

Table 5: Fly ash of waste combustion

Element	Concentration		
	Min	Max	Median
K	12000	74000	43000
Ca	30000	90000	60000
Ti	–	–	–
Cr	101	1318	594
Mn	1700	3040	2900
Fe	28000	40000	34000
Co	6,7	98	43
Ni	101	586	289
Cu	134	3461	1487
Zn	13315	22190	15016
As	13	108	64
Mo	24	45	35
Cd	42,4	414	316
Sn	11	1596	724
Sb	76	1134	800
Tl	1	2,1	2
Pb	4	82	4

Distribution of Cl differs [3], so that near 10-15% stay in the solid body, the equal quantity in the dust and 70-80% in the gas phase. In the case of Br: 40-50% in bottom and 30% in fly ash.

The water solubility of slag for high temperature treatment is characterized with chlorides 0,024–0,6 mass % and sulphates 0,036–2 mass %. Practically all metals (the total concentrations are drawn in table 6), which are presenting in solutions in the form of cations and oxy-anions, build difficult soluble hydro-oxides and oxy-hydrates in the weak base medias.

Table 6: Statistical evaluation of water solubility for rest species

Element	Min	Max	Median	N
K	19,2	76,0	40,3	22
Ca	164	742	467	22
Ti	0,17	0,17	0,17	1
Cr	0,1	0,48	0,24	21
Mn	0,04	0,07	0,06	2
Fe	0,04	0,37	0,06	17
Co	0	0,12	0,03	19
Ni	0	0,1	0,06	19
Cu	0	0,09	0,04	20
Zn	0,04	1,46	0,15	22
As	0,01	0,04	0,03	6
Mo	0,12	0,39	0,26	13
Cd	0,04	0,05	0,04	2
Sn	0,04	0,16	0,07	3
Sb	0,03	0,1	0,06	4
Tl	–	–	–	–
Pb	0,04	2,15	0,24	21

The appearance of chromates, molibdates, arsenates, antimonates in the conditions of thermal treating for waste was investigated [11] in the aspect of their solubility in acid and base environments. In the work [12] Vehlow confirmed the content of Me- anions in the solutions. The creation for aluminates [13,14] in the condensed phase of the burning products for solid propellants and the own computations (see figures 2-4) allow obtaining of solved salts in MSW incinerators – potassium and calcium thallates as a result of the following reactions:

Reaction products	$[\mathrm{TlO2}]^{-}$	$K_2O + Tl_2O_3 = 2\cdot KTlO_2$
		$CaO + Tl_2O_3 = Ca(TlO_2)_2$
	$[\mathrm{TlO3}]^{3-}$	$3\cdot K_2O + Tl_2O_3 = 2\cdot K_3TlO_3$
		$3\cdot CaO + Tl_2O_3 = Ca_3(TlO_3)_2$
	$[\mathrm{Tl4O7}]^{2-}$	$K_2O + 2\cdot Tl_2O_3 = K_2Tl_4O_7$
		$CaO + 2\cdot Tl_2O_3 = CaTl_4O_7$

The calcium thallates (presented in the figure 2) have the two times lower activity in compare with the aluminates. They are detected at the higher temperature 2000-2500 K, than the potassium ones in the figure 3, the existence area of which likewise extends on the all interval of heat cultivation. The figure 4 shows the phase diagram in the system K-Ca-Tl-O at 1500 K. The calculation confirms the thermodynamic prevalence of the Ca- compounds at the low values of the oxygen excess, but at the higher O- concentrations the synthesis of the K- complexes takes place.

With aim to compute the activities of combustion outputs for fuel from MSW [15,16] we produced and designed the program complex MeRS-data, which provided creating and visualizing databases, searching information, inserting new components and presenting graphics and tables.

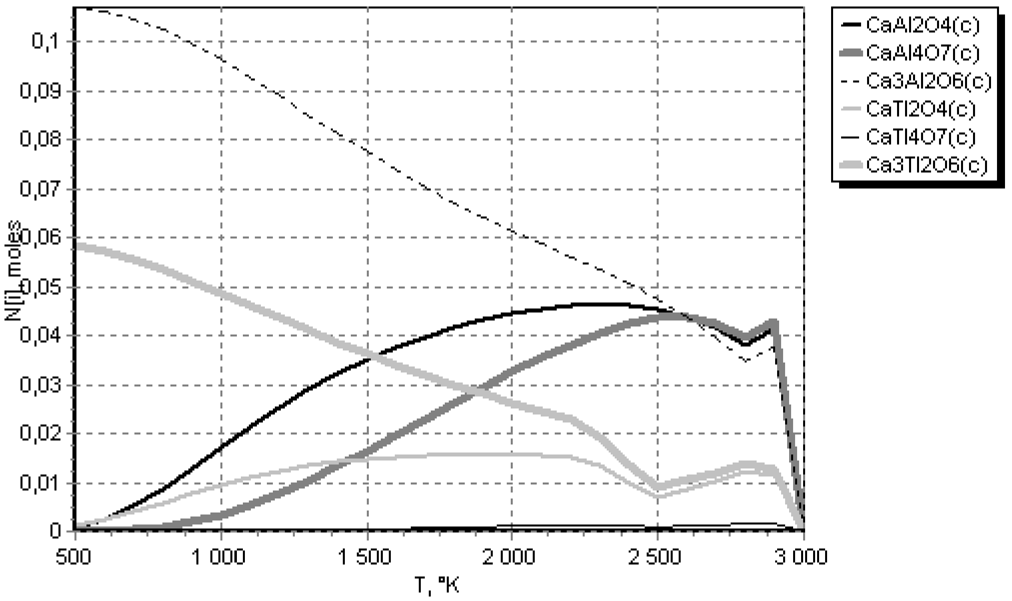

Figure 2: Phase diagram for species created in a chemical reactor of the system Ca-Al-Tl-O at the atmosphere pressure

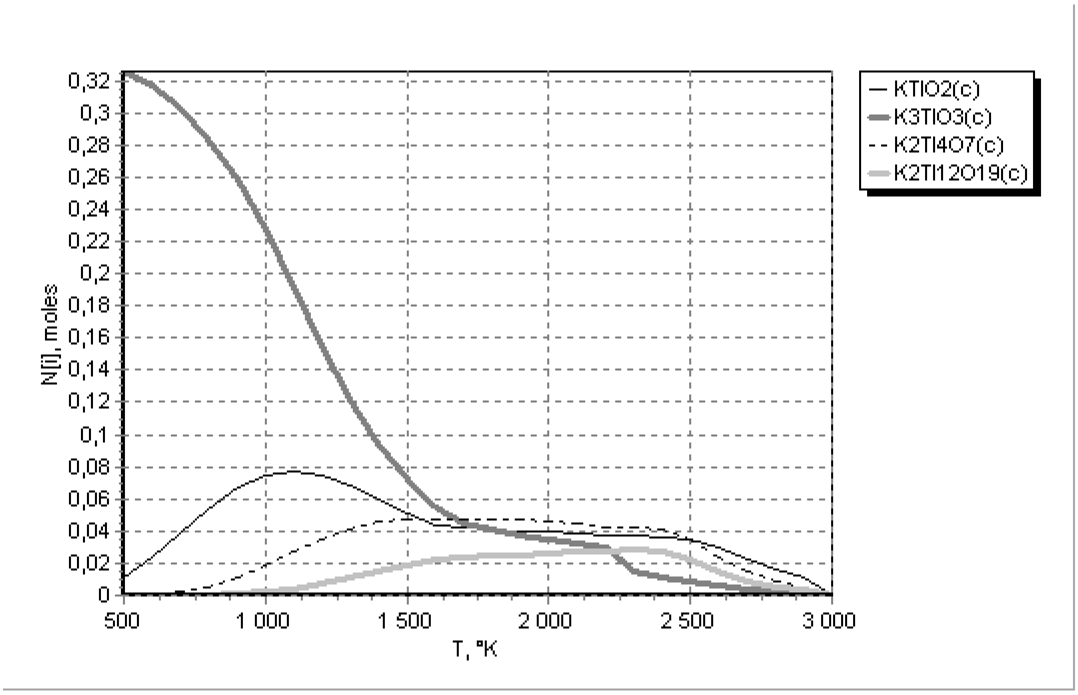

Figure 3: The activities of condensed phases for the potassium thallates in the conditions of a high-temperature transformation of MeRS

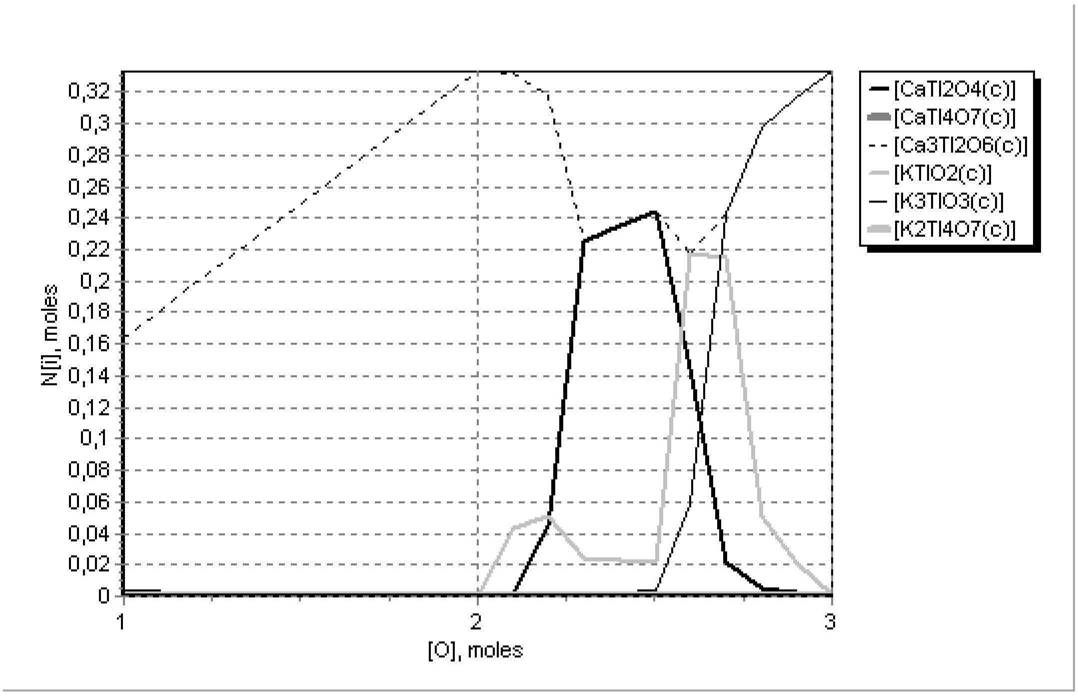

Figure 4: Dependence of the equilibria in slag Ca-K-Tl-O on the oxygen excess from 2 till 3 moles in the fuel at T=1500 K

1.4 Glassification and cementation

In this department we introduce the mechanic types of immobilization, which are directed to the isolation of a toxic part for slag in a silicate and cemented matrix or in an oxide film. The principle of these methods is well known from the theory [17], where the metals are divided in two groups: first are building the oxides and its melting temperature is higher than T_{melt} of pure substances, and mobile ones. We consider that after homogenization near 2000 °C the molten body is covered with complex compounds of the silicium and calcium oxides. It means that we become the species and they are not dangerous for the water environments. But it is not the case of materials because they are amorphous. We need the regular structure of aluminium and magnesium oxides. In this aspect the plasma spraying is actually, when we use the energetic fuels [17] on the basis of Al and Mg with aim to reach the anti-corrosion coatings.

Schütze [18] offered the experimental results for powder co-diffusion samples Al-Al_2O_3 and Al-Al_2O_3-Si to deposit on the Fe-Ni alloys at 900 °C. The model investigations [19] of corrosion in the boiler of waste incineration plant displayed a worse properties for di-aluminium tri-oxide, but the additives of magnesium one showed the better resistance. [10] confirmed the positive effect of stability against Cl- species after covering the steel surface with passivated layer of metallurgy slags, rest substances of aluminium production from boxites (table 3), Portland cement. In the article [20] the mix of milled bottom ash and cement were regarded as a hydraulic barrier for the waste of middle toxity level at high values of pH.

Experimental investigations for MSW incineration and glassification (cementation) of MeRS were tested in the pilot plant TAMARA [6,21]. The united grates had the length of 3,2 m and the width of 0,8. The combustion areas were separated to the four zones (reviewed in the table 7) by flowing a technical air from both sources 400 and 800 m^3/hour. The material balances after distribution of the treatment outputs in relative unities were 400 kg bottom, 20 kg fly ash and 10 kg washer slurry. The research results displayed the good resistance against the base influence with pH 9-11 of slag after thermal treatment at 850-1000 °C. The later increasing of the transformation temperature till 1300 °C showed no effects. Thus the return of the condensed phase in the burning chamber and its re-inertization produced the substances, which did not differ from the earths in the total concentrations of toxic elements.

Table 7: Methods of thermal incineration for waste in compare

Temperature °C	Inert atmosphere	Gasification tools	Oxygen excess
< 150	Drying	Drying	Drying
< 500	Low-T-Pyroluse		
500 – 800	Pyroluse	Gasification	
> 800	Hihg-T-Pyroluse		Combustion

2. Modeling of phase equilibrium in the slag systems

The task of a computation for the equilibrium concentrations of substances, which arise during a combustion of MSW in the incineration plants, is equal to the calculation of the activities for species appearing and destroying at the fixed temperature, pressure and elementary content. It means that the system of equations for the chemical potentials $\mu_{\text{равн}} = \frac{-\Delta_r G(T,p,n)}{R \cdot T}$ of all reactions reaches the global minimum of the free Gibbs energy. n is the substantial composition in the burning chamber of the treatment facility.

In the case of a condensed phase let us write Φ°(T)=-G(T)/T+H(298,15)/T in the polynomial form

$$\Phi^\circ(T) = A + B \cdot T + C \cdot \ln(T) + \frac{D}{T} + \frac{E}{T^2}, \ (1)$$

introducing the approximation curve for experimental dependences of the heat capacity on the temperature.

The discretization errors e_h are drawn in the table 8 together with the consistence $\max\limits_{T \in I} \|e_h\| \leq L \cdot \max\limits_{T \in I} \|\tau_h\|$ of the numerical algorithm for the start value task (SVT).

$$A_h \Phi^\circ(T) = f(T, \Phi^\circ(T)), \ \Phi^\circ(298{,}15) = A_{T_0} \quad (2)$$

$A_{298,15}$ is a constant of integration and A_h is a differential operator.

Table 8: Global and local convergences

Method Step h	Euler $\|e_h\|$	Heun $\|e_h\|$	Runge-Kutta $\|e_h\|$	$\|\tau_h\|$
0,01	after 1200 °K all no convergence and after 2300 °K no consistence			3,169
0,001	0,168	0,337	0,373	0,384
0,0001	0,0158	0,0321	0,032116	0,0384
0,00001	0,0016	0,00317	0,003156	0,00384

The function (1) is continuous in the interval $I = (T_0, T_0 + H)$. According to the convergence of the second difference $\|f(T, \Phi^\circ(T)) - f(T, \Phi'(T))\| \leq L \cdot \|\Phi^\circ(T) - \Phi'(T)\|$ the solution of SVT (2) exists and is singular. We find it with the Euler, Heun and Runge-Kutta methods. It is global and extended on the all H. We are pleased to notice that the exponential compatibility takes place too.

The estimation of $\|\tau_h\| \leq \frac{1}{2} h \cdot \max\limits_{T \in I} \left\| \frac{2D}{T^3} + \frac{6E}{T^4} - \frac{C}{T^2} \right\|$ introduces the case, when the first order approximation is not sufficient at the start temperatures and h=0,01. The precision of the step width till 0,001 eliminates this problem.

Thus we studied the important property of this function in respect to the main problem of the chemical engineering [22] – investigation for the content of complex compounds in the gas and condensed phase in terms of modeling for thermodynamics of the real technical processes.

Grounding to the foregoing we write the difference schema of the rand value task for the system of i- linear differential equations

$$A_h n_{h,i}(b_j) = c_i, \ 0 < b_j < \kappa_j, \ n_i(0) = 0, \ n_i(\kappa_j) = \upsilon_i, \ \kappa_j, \upsilon_i \in I = (0, \infty) \ (3)$$

at the elementary balance condition [22,23] $\sum\limits_{i \in I_T} a_{ji} n_i = b_j, \ j = \overline{1,m}$, where m is the quantity of chemical elements, which build the pure quiddity, a_{ji} is the quantity of j- atoms in a i- substance.

The value $n_l = c_l \sum_{l=1}^{L} h_{li}$ is found with help of Ritz's variation method in the approach of the rounding error with the order 10^{-7}. The activity c_i is the existence ability for each composition in the reactions following to a minimization of the Gibbs energy.

(3) has the singular solution in the interval $\lfloor 0, \kappa_j \rfloor$ and is stable at all computed values for the inconstant width of the step. Thus the concentration change is always equal to the multiplication of its activity on h in the each iteration for the temperature layers.

The figure 5 display the calculation results for the system K-Al-O in the conditions of the thermal treating for MSW. The thermodynamic functions (heat capacity, T- divided enthalpy change and Φ°(T)) for KAl_9O_{14} are presented with help of properties IVTANTHERMO [14] (index 'ivt') in the comparison with calorimetrical investigations [24] and show the good compatibility of dates. The program for the numerical modeling of coefficients (1) is developed with aim of the interpolation for Φ°(T). We reach the precision of 10^{-3} for the mix oxides $0,5K_2O \cdot 4,5Al_2O_3$. In the figure 6 the model experiments is graphed in compare with calcium aluminates, which prevail also as thallates at the low oxygen excess α.

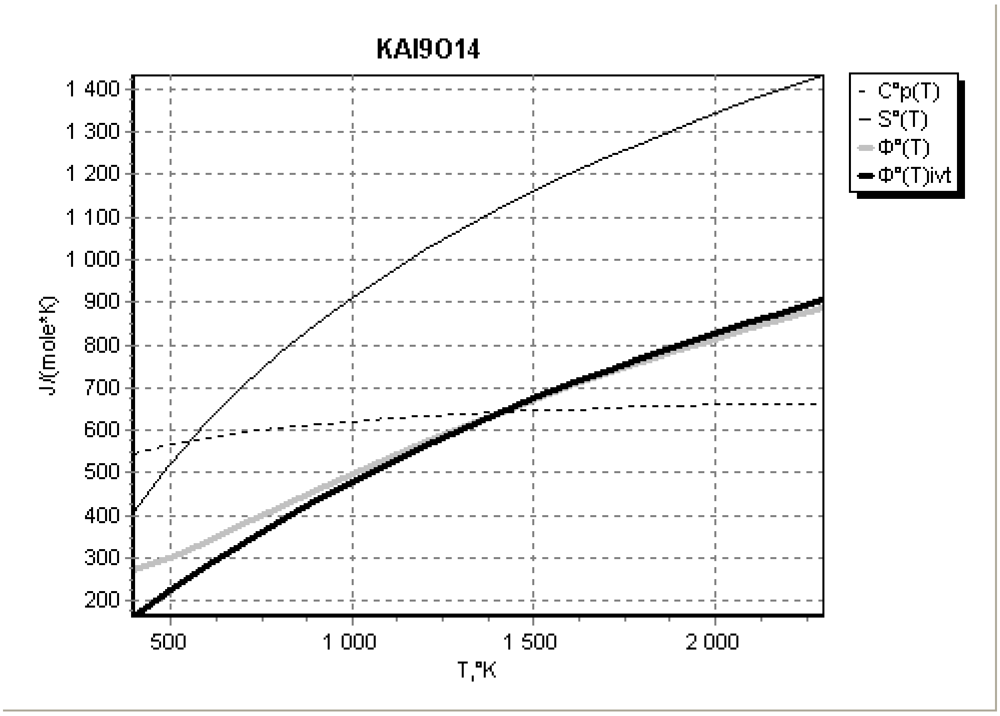

Figure 5: Compatible computation of the approximation terms for the thermodynamic functions of the oxide mix $0,5K_2O \cdot 4,5Al_2O_3$ IVTANTHERMO and potassium tetraluminate KAl_9O_{14} FACT [25]

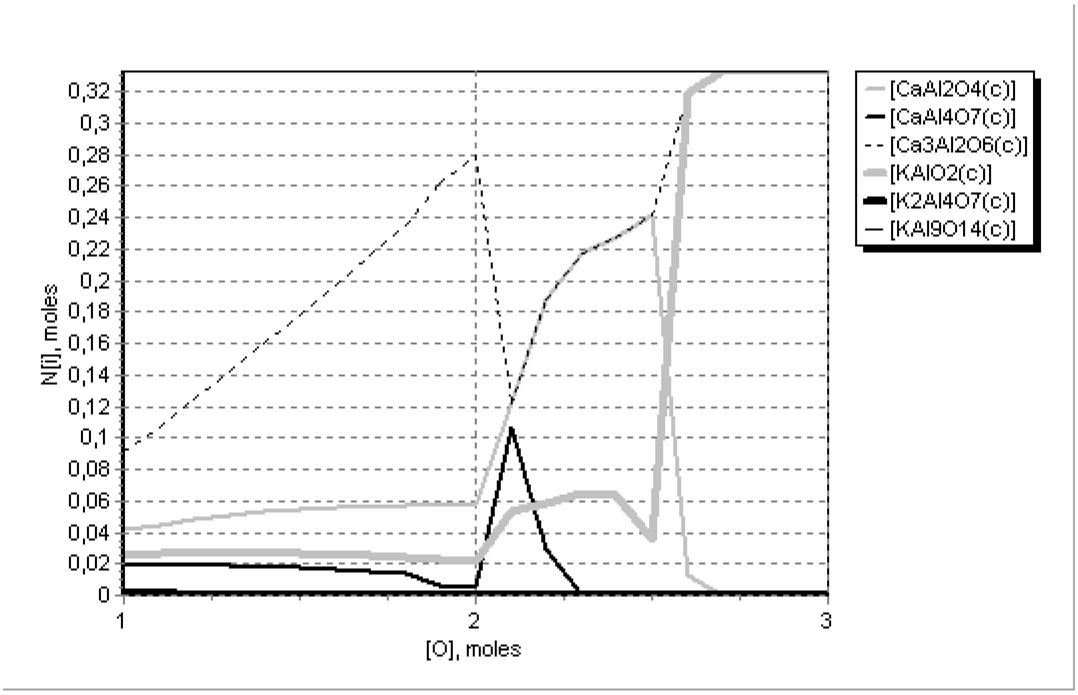

Figure 6: Molar brakes for the aluminates at T=1500 K and the change of the potassium content from 1 mole till 3 with the step 0,1

Summary

The statistic assessment of elementary content for MSW and the fraction analysis displayed that waste was the substitute fuel on the basis of the energetic rest substances for production and consummation in the industrial countries. Its using as an energy bearer is rational in the plants equipped with boilers. The result of the combustion process is slag, which don't differ from natural grounds in its chemical and phase composition. Thus the thermal treatment methods are directed to the fixing of soluble salts in the silicate and cemented matrix of the out coming products. It means that the binding of thallates takes place analogous with calcium alumina-silica to prevent the mobilization of oxy-anions in the acid and base environments. The primary measures initiate the vaporization of toxic metals in the compounds with chlorine, bromine species in the burning chamber and also the purification of the melted bottom ash from the excess additives in a periphery.

For example of aluminium oxide the dates of the thermodynamic modeling [14] and [24] show the possibility of building for the complexes of thallium with Ca and potassium, which dissociate in the form of anions containing Tl and O. Their control in the total concentrations should be corrected with a real toxity of $(TlO_2)^-$, $(TlO_3)^{3-}$, $(Tl_4O_7)^{2-}$. Own calculations confirm the appearance of the mix crystals in the all interval of conditions for waste incineration, so that the lack of oxygen influences the pyrolyze for the system Ca-Tl-O, but its excess serves the gasification for the K-Tl-O essences.

In terms of this article we'd like to notice the mathematical model for the high-T transformation of the fuel from waste in the TAMARA facility. The algorithm for the estimation of equilibrium activities was created with help of the permanent comparison of the numerical investigations with experimental ones. The program MeRS-data was designed and produced to analyze qualitatively and semi- quantitatively the phase structure of slag materials. According to calorimetrical measures of the warm capacities we constructed the difference schema for the integral dependence of enthalpies on temperature and solved it with the Euler, Heun and Runge-Kutta methods. As a part of energy balances the coefficients of the approximation equations for the T-divided Gibbs functions were determined with aim to minimize the computation errors for thermodynamic properties of pure substances.

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