

STRUCTURE AND PROPERTIES OF ^{137}Cs CONTAINING WASTE GLASS

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High level radioactive wastes (HLW) from both the nuclear power industry and military nuclear programmes are currently immobilised by vitrification. Borosilicate glass provides the very suitable medium for the majority of the species present in these wastes [1, 2]. It prevents leaching of radio nuclides by water-immobilisation effect. Environment protection legislation and increase of disposal costs makes the vitrification of the low level nuclear waste (LLW) favorable [3]. The treatment of such waste begins with reduction of their volume by incineration. The ash containing radio isotopes, is suitable for vitrification and ceramization by incorporating it into the composition of boro-aluminosilicate glass, similar to that used in nuclear power industry.

Our studies concerned the influence of Cs as a characteristic component of ash after combustion of LLW on the structure and properties of borosilicate glass. The effect of introducing Cs_2O or both Cs_2O and CaO on the properties of boro-aluminosilicate glasses is little known, as the compounds are not used in traditional glass manufacturing.

Four different compositions of glass were studied. (Table 1). CaO was present in glasses 1 and 2, and was added as the characteristic component of ash obtained from paper incineration. The other ash components: SiO_2 , Al_2O_3 were already present in the base glass (glass 3) composition. In two glasses a part of Na_2O (5 mass %) was replaced by Cs_2O (glass 2 and 4).

Table 1. The chemical compositions of investigated Glasses.

Chemical compositions [mass %]	Glass 1	Glass 2	Glass 3	Glass 4
SiO_2	48.5	48.5	56	56
B_2O_3	7.5	7.5	15	15
Al_2O_3	8	8	8	8
CaO	18	18	-	-
Na_2O	18	13	21	16
Cs_2O	-	5	-	5

Our results have shown that the introduction of relatively small amount of Cs_2O (5 % mass) instead of Na_2O induces considerable changes in the structure and the thermochemical properties of boroaluminosilicate $\text{Na}_2\text{O} - \text{B}_2\text{O}_3 - \text{Al}_2\text{O}_3 - \text{SiO}_2$ glass. FTIR spectrum of $\text{Na}_2\text{O} - \text{B}_2\text{O}_3 - \text{Al}_2\text{O}_3 - \text{SiO}_2$ glass is characterized by an intensive band of Si - O - Si bridges of silicate network at 1000 cm^{-1} , 729 cm^{-1} , and a band 1407 cm^{-1} of $[\text{BO}_3]$ groups which occur in its structure. The band

connected with the cation modifier Na^+ occurs at 458.8 cm^{-1} . Introduction of Cs_2O causes the shift of the main band to 1017 cm^{-1} and the glass modifiers band to 455.9 cm^{-1} . It means that the presence of Cs^+ increases the degree of polymerization of the network. Introduction of Ca causes the appearance besides of the Si-O-Si band (1021 cm^{-1}) the band at 946 cm^{-1} , which is characteristic for silicate and aluminate glasses, containing calcium. This indicates the formation in the network the domains containing Ca ; also yielding the shift of the $[\text{BO}_3]$ band to 1422 cm^{-1} . The band connected with the cation modifiers becomes shifted to 471 cm^{-1} . This means that the presence of 5% mass of Cs_2O , affects significantly the glass structure. The effect is stronger when Cs_2O and CaO are introduced simultaneously; then the degree of polymerization of the silicate network increases producing the observed shift of the Si-O-Si band toward higher frequencies (from 1000 to 1021 cm^{-1}).

Introduction of Cs into the basic glass $\text{Na}_2\text{O} - \text{B}_2\text{O}_3 - \text{Al}_2\text{O}_3 - \text{SiO}_2$ (waste glass) in order to immobilize its radioactive isotope induces changes in the structure and in the properties of the glass. This results from considerable crystallochemical differences between Cs^+ and Na^+ as the glass structure modifiers. In the presence of even a relatively small amount of Cs_2O the degree of polymerization of the glass network as well as its stability increases. Evidently this means the increase of the chemical durability of the glass. On the other hand above glass transformation temperature (T_g), cesium acts as the strong depolymeriser, increasing the tendency for crystallization of the basic glass. It becomes incorporated into the structure of the newly formed crystal phases such as the chemically resistant aluminosilicates. Similar changes are induced by the introduction of CaO into the glass. Our results confirm and explain the suitability of borosilicate glass for inactivation of the incinerated hospital and laboratory wastes contaminated with radioactive ^{137}Cs [4].

References

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