Atomic Energy Organization Of Iran

Application of Natural Zeolites as Pre-Materials for Immobilization of Radioactive Waste in Glass Matrix

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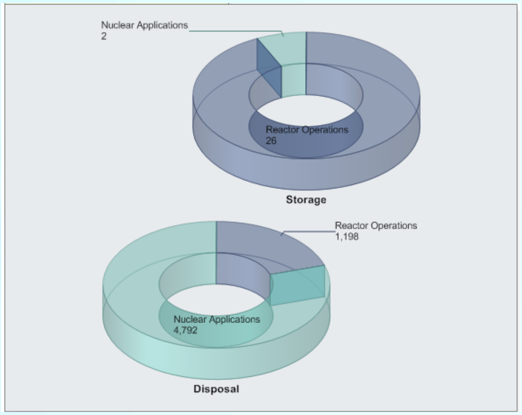
Outline

- **Nuclear Reactor operation in IRAN**
- **Waste Management Strategy In IRAN**
- **Application of Zeolite for Waste Treatment**
- **Application of Zeolite for Waste Immobilization**

Nuclear Reactor operation in IRAN

- The Bushehr Nuclear Power Reactor (BNPP)-915 MWe VVER-1000 type

- The Tehran Research Reactor (TRR)-5 MW pool-type research reactor



Waste Management Strategy In IRAN

❑ Iran Nuclear Waste Management Company (IRWA) is the only authorized company for radioactive waste management in Iran which acts under framework of Iran nuclear regulatory authority and AEOI.

- **Main activities:**
- Waste treatment
- Cementation
- Interim storage
- Near surface disposal

❑ Nuclear Science and Technology Research Institute (NSTRI) do research for establishment, development, promotion and optimization of methods and processes for waste management.

Waste Treatment

- Cation exchange, followed by a suitable solidification treatment, e.g., encapsulation in a cement matrix, is one of the most effective procedures to remove and safely storing hazardous contaminants present in radioactive waste streams.
- Owing to the fact that organic exchangers (resins) turned out poorly effective, due to their low radiation and thermal stability, natural zeolite exchangers have successfully been proposed as a valid alternative, also in consideration of their pozzolanic nature and the consequent ability to interact with Portland cement.

Waste Treatment

- Zeolites are one of the important inorganic cations exchangers and effectively used for selective removal of radionuclides, especially fission-product radionuclides of high activity such as ^{137}Cs.
- Their structures of aluminosilicate framework are durable in contacting with waste water and their ion selectivity is effective even in the solution containing other competing ions.
- An alternative way to safely store radionuclides, after taking up them in a zeolite framework, is based on a thermal treatment, which destroys zeolite structure and blocks the undesired species into a vitreous lattice or a non-exchanging crystalline phase.

Waste Treatment

- This study deals with the treatment of waste solution containing Cs^+ using natural zeolite as well as the Vitrification process for immobilizing of these radionuclides in borosilicate glass products.
- At first, radioactive ions were adsorbed in the zeolite framework, and then the waste loaded zeolite was used as pre-material for producing borosilicate glass.
- The evaluation of waste-loaded glass samples were carried out using X-ray techniques (XRF, XRD) and SEM analysis.
- The leaching of waste loaded borosilicate glass matrices have been also studied.

Natural Zeolite

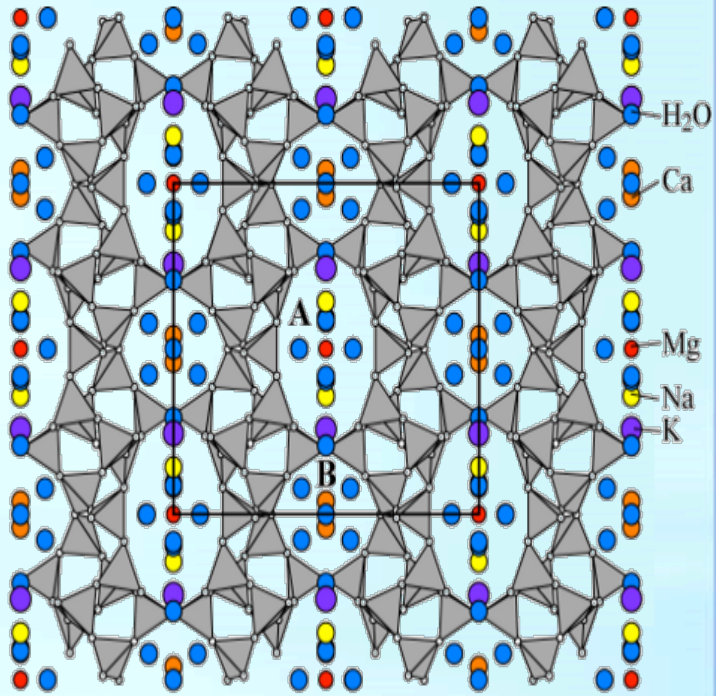
Clinoptilolite Zeolite

Clinoptilolite is a natural zeolite composed of a micro-porous arrangement of silica and alumina tetrahedra.

It has the complex formula:

$$|(Na,K,Ca_{0.5},Sr_{0.5},Ba_{0.5},Mg_{0.5})_6(H_2O)_{20}|[Al_6Si_{30}O_7]$$

Chemical composition (wt%)	
SiO_2	83.37
Al_2O_3	8.54
K_2O	1.67
Na_2O	1.27
CaO	2.3
MgO	0.65
Fe_2O_3	1.3
TiO_2	0.26
MnO	0.37



Natural Zeolite

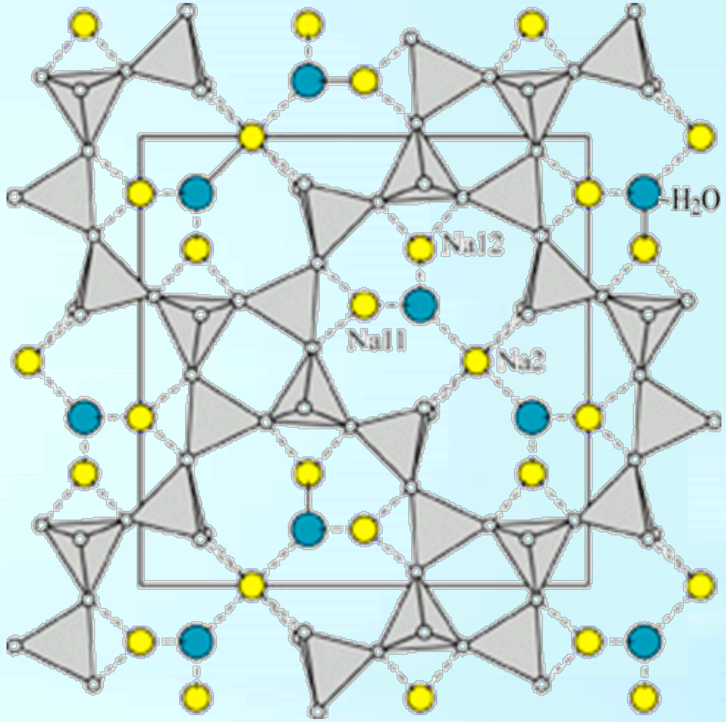
Analcime Zeolite

Analcime or analcite is a natural zeolite consists of hydrated sodium aluminium silicate in cubic crystalline form.

It has the formula:

$$|Na(H_2O)|\ [AlSi_2O_6]$$

Chemical composition (wt%)	
SiO_2	86.48
Al_2O_3	4.73
K_2O	2.23
Na_2O	0.05
CaO	2.47
MgO	0.01
Fe_2O_3	2.15
TiO_2	0.36
MnO	250 ppm



Adsorption Experiments

The effects of various parameters on the adsorption efficiencies of Cs has been studied systematically by batch experiments.

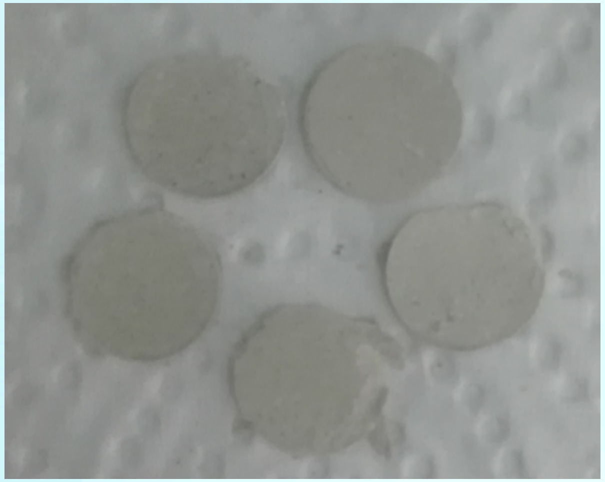
- Initial pH value of the solution
- Contact time
- Temperature
- Ionic strength of solution
- Interference ions
- The initial concentration of the metal ions

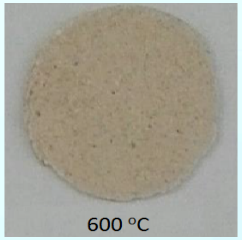
Max Cs adsorption capacity Of 125.40 $mg.g^{-1}$ for Clinoptilolite

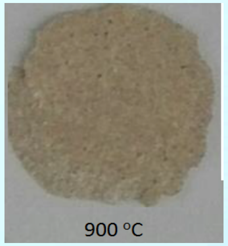
Max Cs adsorption capacity Of 105.12 $mg.g^{-1}$ for Analcime

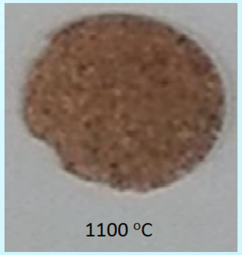
Heat treatment of Cs loaded Zeolites

- First the pellets of Cs loaded Clinoptilollite and Analcime were made.
- Then the heat treatment at different temperatures was performed.
- Finally the chemical durability of samples were investigated.

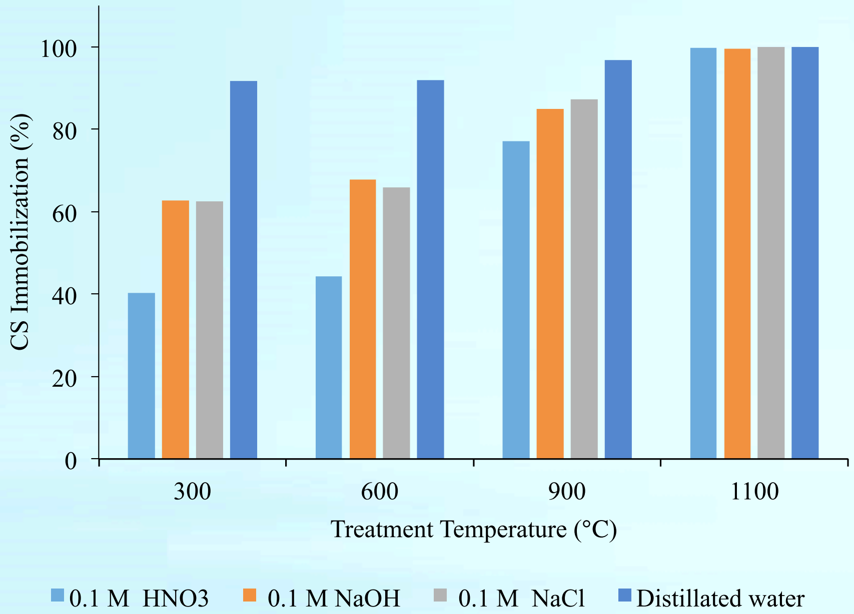


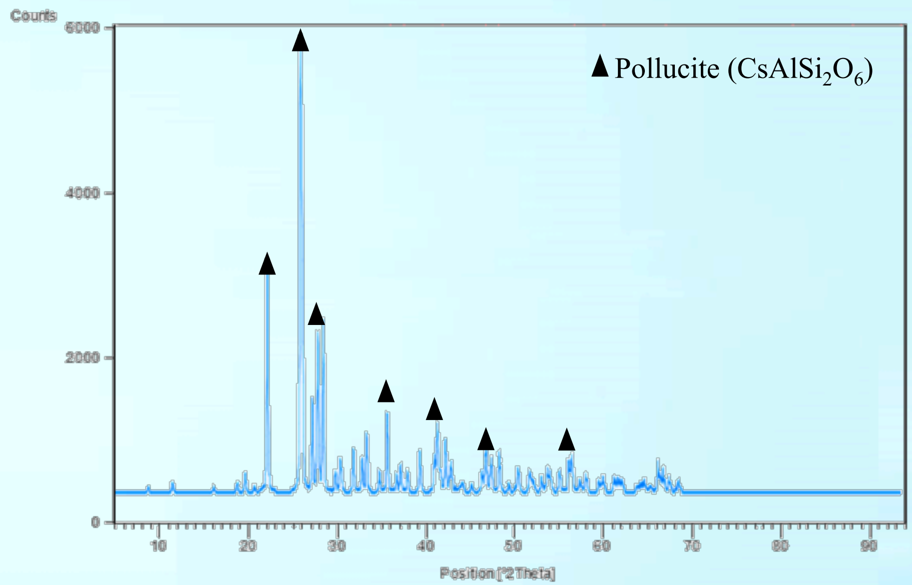




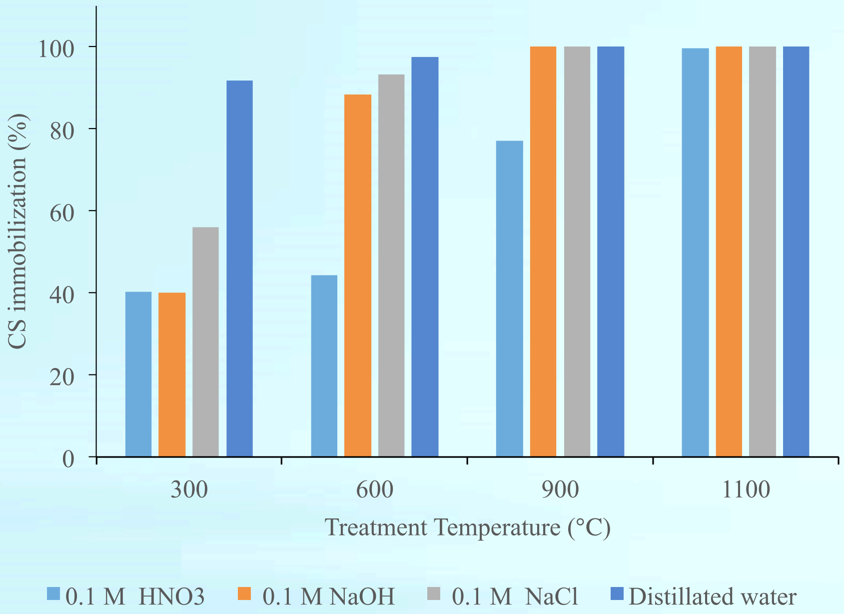


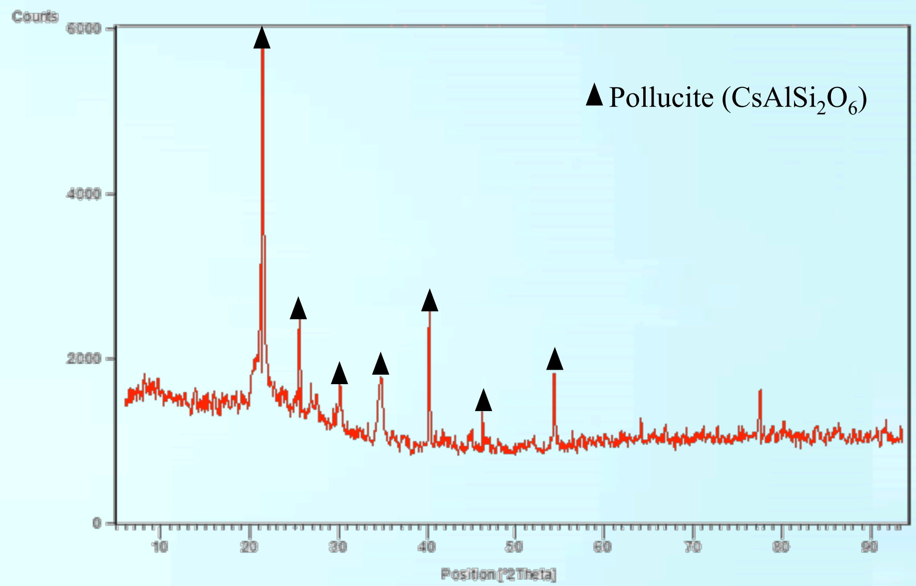
Heat treatment of Cs loaded Clinoptilollite



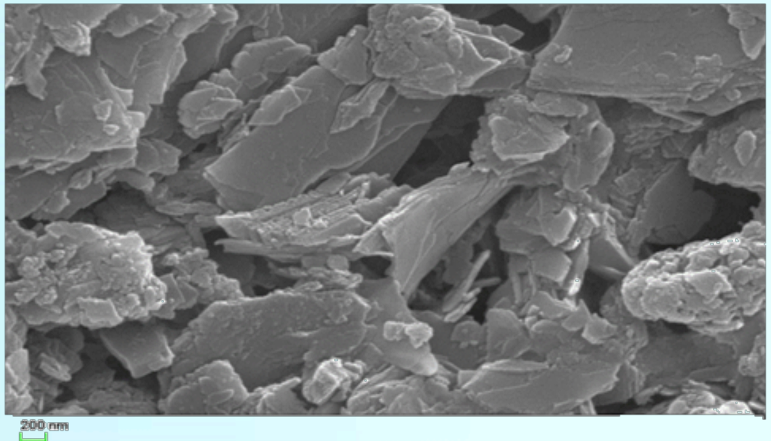


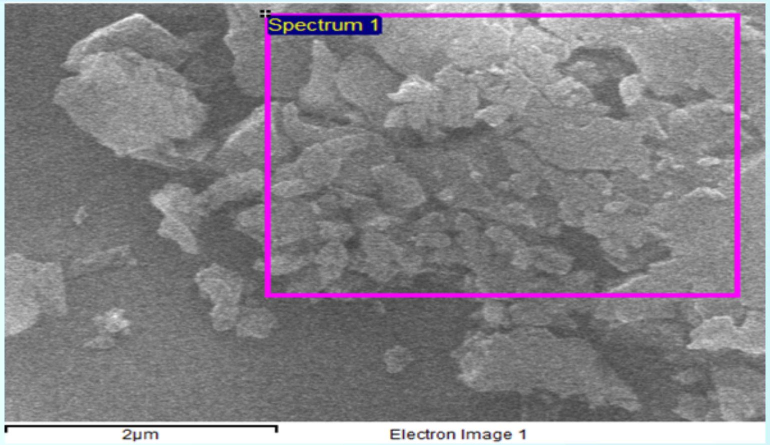
Heat treatment of Cs loaded Analcime

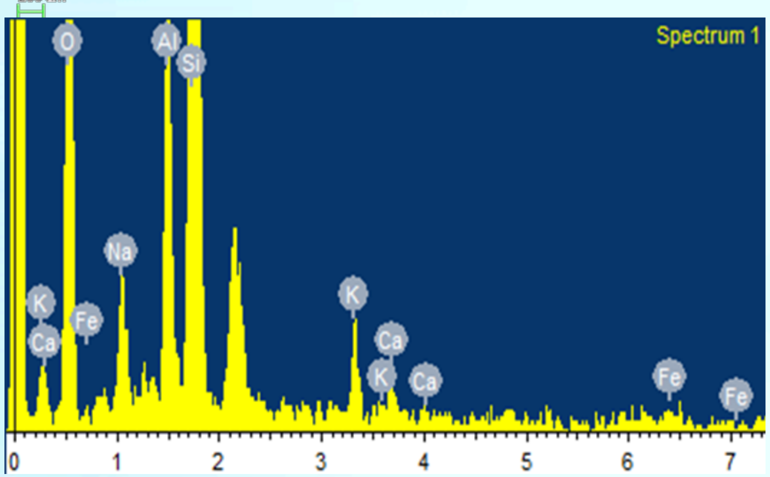


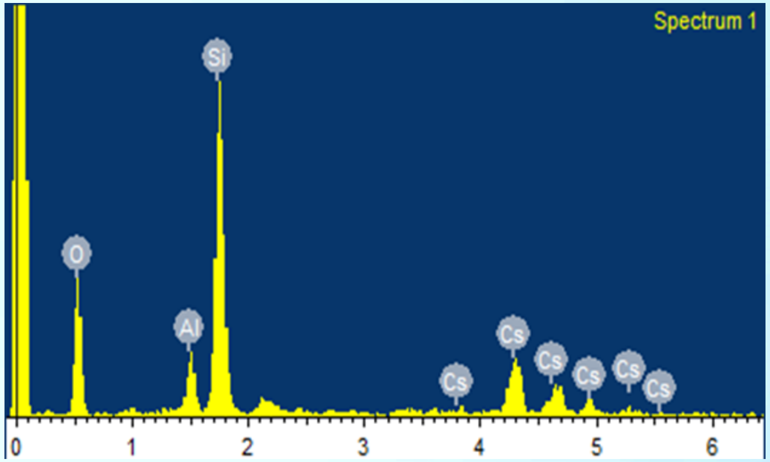


Heat treatment of Cs loaded Zeolites









Heat treatment of Cs loaded Zeolites

- The adsorption and immobilizing of radioactive Cs-137 was also studied.

Radioactive sample	Activity before adsorption (Bq/l)	Activity after adsorption (Bq/l)	0.1 M HNO_3	0.1 M NaOH	0.1 M NaCl	Distillated Water
Cs-137	180	10	0	0	0	0

The reported data demonstrate that a thermal treatment of a Cs-exchanged Clinoptilolite at suitably high temperatures, e.g., 1100°C, is an effective procedure to immobilize Cs+ inside zeolite structure.

It is of interest to note that analogous procedures, applied to Analcim gave similar results with even better immobilization performances.

Vitrification of Cs loaded Zeolites

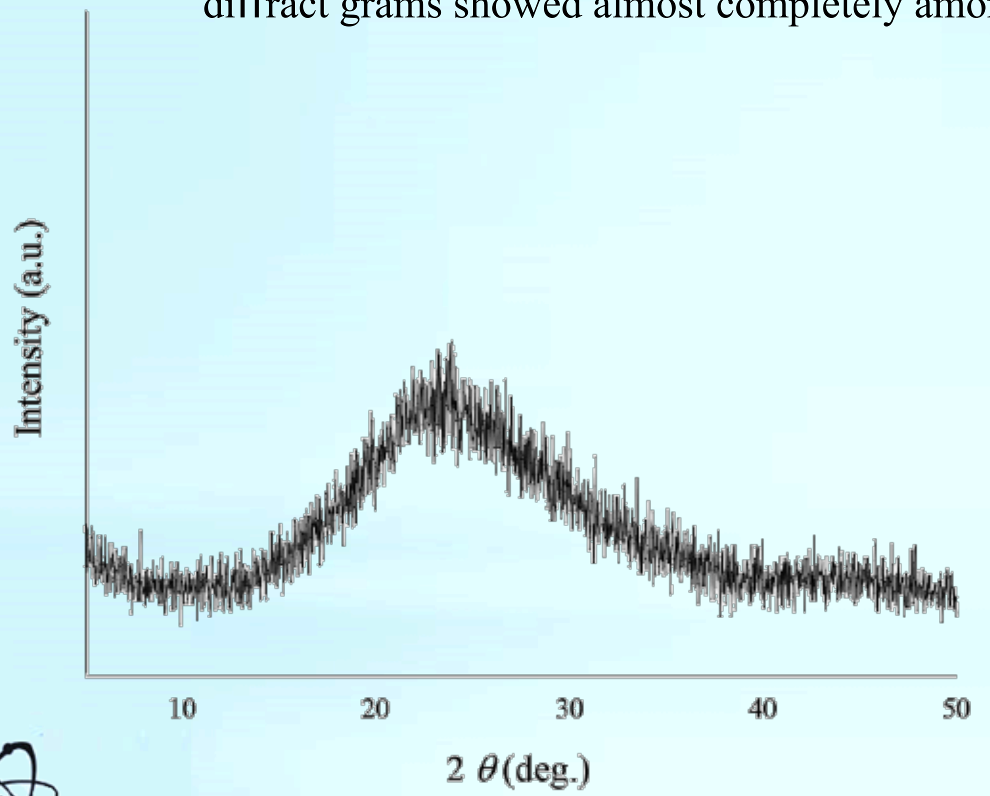
- For producing borosilicate glass, Cs loaded Clinoptilollite and Analcime were blended with some additives.
- The glass composition is considered as:

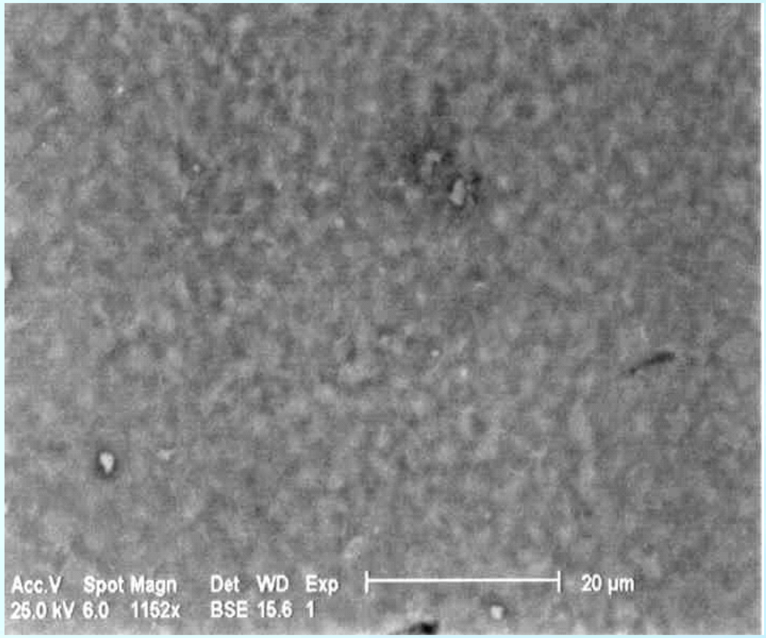
Al_2O_3: 9.46%, B_2O_3: 15.61%, CaO: 2.55%, MgO: 0.95%, Na_2O: 11.1%, SiO_2: 47.1%

- Considering Clinoptilollite and Analcime as sources of SiO_2 and Al_2O_3, the amount of these two zeolites and other components was calculated according to the XRF analysis of both zeolites.
- The mixture was melted for 3 h at 1200 ℃ and resulted glass was evaluated.

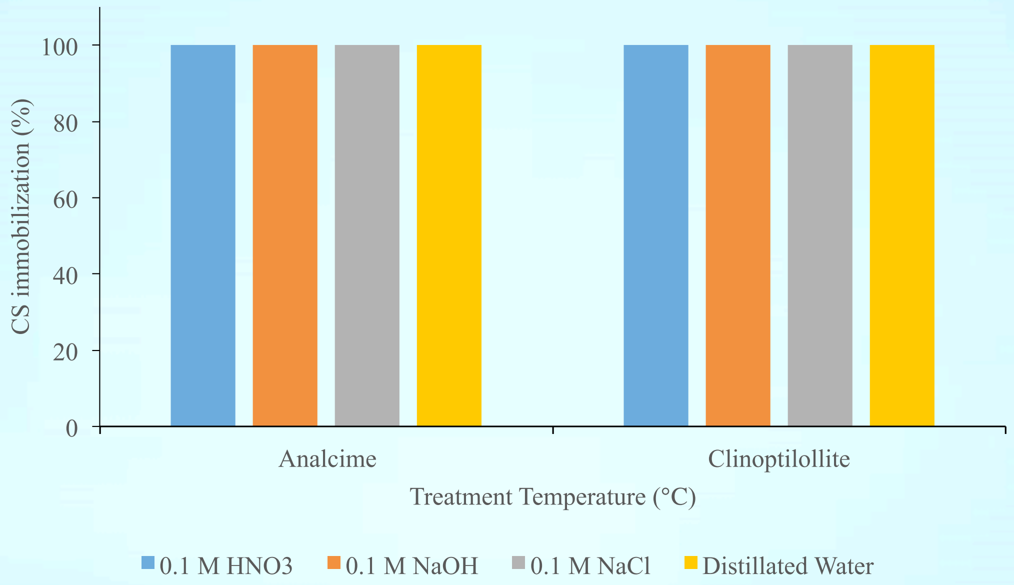
Vitrification of Cs loaded Zeolites

Although separated zeolite spots were observed in the glass products melted, the XRD diffract grams showed almost completely amorphous state for the glass product.





Vitrification of Cs loaded Zeolites



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Thank you for your attention