MCNP simulation for setting up experiments of electron beam irradiation on zeolite ZSM-5 and mordenite

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Abstract

In this work, we have simulated the interaction of electron beam with zeolite Mordenite & ZSM-5 in different experimental configurations to optimize the uniformity of the irradiating experiment for reducing errors in the study modifying structures of the zeolites by the electron beam. This simulation helps to investigate and control a number of problems such as the possibility of decelerating electron 10MeV of some common materials, the energy spectrum of electrons for irradiating the sample, the uniformity of the imparted energy of electrons in the sample, the heat generated during irradiation, irradiation time corresponding to dose levels. With the information obtained from the simulation process, we can set up the irradiation experiment for zeolite samples with the expected doses. In addition, the simulation data gives us information about the absorbed energy in the zeolite sample. This will support the interpretation of experimental results in a systematic way in future.

Keywords: Electron irradiation, ZSM-5, mordenite, MCNP

1. Introduction

Mordenite & ZSM-5 are aluminosilicate zeolites which have very widely applied in many industrial areas as catalytic, absorption and ion-exchange materials [1]. These zeolites have recently been studied and applied to handle the inorganic and organic liquid waste [2, 3]. Besides, they have been shown to be promising materials for the treatment of radioactive isotopes such as ^{152}Eu, ^{137}Cs, ^{131}I and ^{90}Sr from the liquid waste associated with the operation of the nuclear reactor [4]. The ability to process radioactive isotope ions of the zeolites depends strongly on the number of absorption and ion exchange centers [12, 13]. Studies of the denaturation of Mordenite & ZSM-5 by chemical and physical methods have been performed to produce materials containing defects that have superabsorbent potential for above radioactive isotopes [12, 13]. Recently, some studies on the effect of radiation on the structure of zeolite have been carried out [5, 6]. The most salient research in this direction on the zeolite clinoptilolite was performed by Yeritsyan et al, using 8MeV electron beam with flux up to 10^{16} e/cm^2. Results showed that the ion exchange capacity of the post-irradiation material increased by tens/hundreds of times [5]. The authors also point out that crystal network defects are the source of a very large number of ion exchange centers of zeolite after being modified by electron irradiation.

Vietnam has a great potential for kaolin [11] which is a source of raw materials for direct synthesis of zeolite ZSM-5 and Mordenite [7, 8, 10]. Synthesis and modification of these two types of zeolite by electron beam from accelerator is an important topic in this material development in Vietnam. In order to carry out experimental research, the experimental simulations to investigate and to control the physical parameters of the radiation-material interaction should be performed carefully and particularly.

In this study, we have simulated the interaction of electron beam with Mordenite & ZSM-5 in different experimental configurations. The objective of the work is to optimize the uniformity of the experiment that will reduce errors in the study of the change in zeolite material properties caused by electron beam irradiation. We simulated the irradiation experiment on these zeolite samples to investigate and control a number of problems such as the possibility of decelerating electron 10MeV of some common materials, the energy spectrum of electrons for irradiating the sample, the uniformity of the imparted energy of electrons in the sample, the heat generated during irradiation, and irradiation time corresponding to dose levels.

2. Simulation

We simulated electron irradiation experiment by MCNP4C (Monte Carlo N Particle, version 4C) code, using mode e and p: electrons from sources and photons which are generated from interaction of electrons with matter.

2.1. Accelerator system and simulation configurations

The process of irradiation on zeolite samples will be carried out on the linac model UERL-10-15S2, accelerating electrons by high frequency electromagnetic waves through resonators of accelerating structure. Electron beam energy is 10MeV. The linac has two symmetric scanners as shown in Fig. 1, with dose error of 5%, energy error of 2.5%.

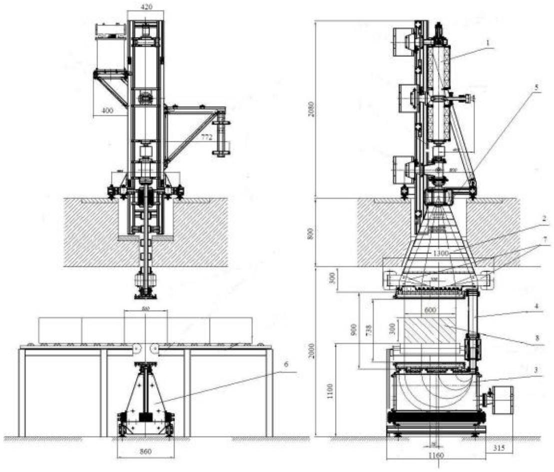

Fig. 1. LINAC model UERL-10-15S2:
1. Acceleration tube structure
2. Scanner 1
3. Scanner 2
4. Electron conductor
5. Electromagnetic field of scanner 1
6. Electromagnetic field of scanner 2
7. Output directional magnetic field
8. Sample location.

As indicated by Yeritsyan et al [5], the 8 MeV electron beam is optimal for generation of structural defects. For this reason, firstly, we investigated the possibility of decelerating 10 MeV electron down to 8 MeV of some common materials such as lead, copper, iron and aluminum. The simple configuration of the first simulation consists of a 10 MeV electron disc source directly irradiating to the metal plates with different thicknesses. Behind the metal plates is the detector of the electron energy (tally F8). The simulation results (Fig. 2) show the residual energy spectra of 10 MeV electron braked by different thickness panels of each material. Figure 3 shows the necessary thickness of the material panels to decelerate the electron energy to 8 MeV. The thickness is 5 mm for aluminium, and are from 1.3 to 2 mm for other material. The energy distribution of the electron beam braked through the aluminum layer is the most concentrated, simultaneously, the peak intensity (peak to background ratio) is significantly higher than that of the other materials. This leads to the conclusion that the electron deceleration ability from 10 MeV to 8 MeV of aluminum is best among simulated materials. This ability and some other properties of aluminum (such as low x-ray yield [14], heat-durability, easy for metalworking and fairly cheap) make aluminum the best material among the survey materials used for our experiment.

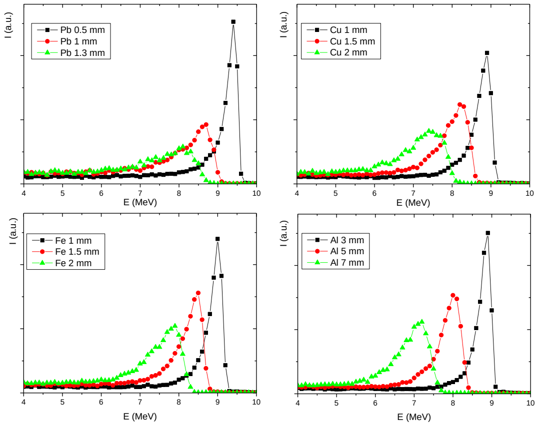

Fig. 2. Energy spectra of electrons decelerated through different material, with different thicknesses.

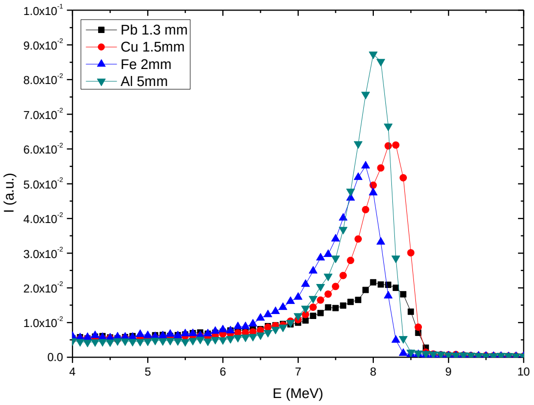

Fig. 3. Necessary thicknesses of the material panels to decelerate the electron energy to 8 MeV.

Based on actual conditions and the survey results above, the simulated configuration is set up as follows: 02 identical disc sources release/emit 10 MeV electrons irradiating on zeolite contained in aluminum container. We surveyed three geometrically symmetric containers which are relatively easy for processing: cylinder (r = 1.2 cm, h = 6 cm), sphere (R = 2.1 cm) and rectangular parallelepiped (2.7 x 2.7 x 6cm), aluminum thickness is 5 mm. These containers' net volumes are nearly equal. The distances from the two sources to the aluminum container are about 32cm (see Fig. 4). The tally F8 was used to study the electron energy spectrum inside the aluminum containers.

The simulation results shown in Fig. 5 indicate that the boxy and cylindrical configurations give similar energy spectra. These energy distributions peak at 8 MeV, concentrate in the range of 7.7-8.3 MeV and decrease rapidly in both directions. For the spherical aluminum container, the energy distribution is less concentrated. We removed the spherical and the boxy configuration, and chose the cylindrical configuration to perform following simulations, because of the better energy spectrum and the more symmetric geometry. The tally F2 is used to survey the electron flux inside (fi) and outside (fo) the aluminum container. The fi/fo ratio is 1.351 (Eq. 1), fi is higher than fo because secondary electrons are created when primary electrons interact with aluminum. At last part, this ratio of flux was used to calculate the irradiation time as well as the amount of energy that the electrons leave behind in the zeolite sample.

$$f_i/f_o = 1.351 \pm 0.003 \qquad (1)$$

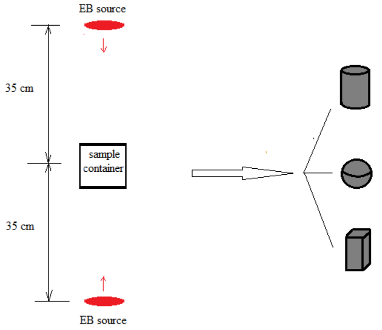

Fig. 4. Simulated configurations to survey electron energy spectrum inside aluminum container.

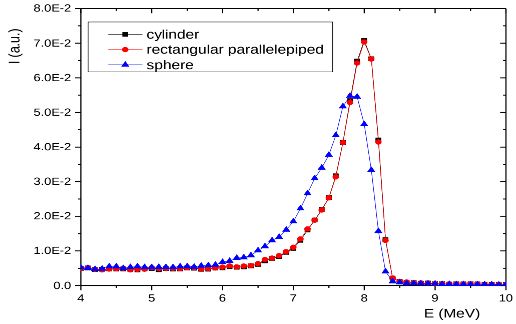

Fig. 5. Energy distribution of electron beam 10 MeV decelerated through aluminum containers.

2.2. Uniformity of interaction energy in zeolite sample

An important factor of the zeolite sample irradiation experiment is the uniformity of the energy that the electrons impart to the sample. We can consider this amount energy as the absorbed dose. If the dose is uniform on the whole irradiated sample, the error of the study on the post irradiation sample will decrease. This will reduce the systematic error and increase the reliability of the experiment. For this reason, the cylindrical aluminum container with thickness of 0.5 cm and radius 2 cm was used to study the uniformity of absorbed dose in the zeolite sample. We divided the sample in the container into several layers, then scored the energy deposited in each layer by the tally F8. The thickness of each layer is 0.5 cm and 0.2 cm corresponding to ZSM-5 and mordenite. Because the mordenite has a higher density, each layer must be divided thinner, so that material will be surveyed more detail. Zeolite composition [1]: ZSM-5 (density 0.75g/cm^3) $[Na_n (H2O)_{16}] [Al_nSi_{96-n} O_{192}]$ n<27; Mordenite (density 2.13 g/cm^3) $[Na_8 (H2O)_{24}] [Al_8Si_{40}O_{96}]$.

Fig. 6. Simulation cases for ZSM-5 sample. Case 1: 2 layers; Case 2: 4 layers; Case 3: 5 layers; Case 4: 6 layers; Case 5: 6 layers with air inside aluminum container; Case 6: 10 layers.

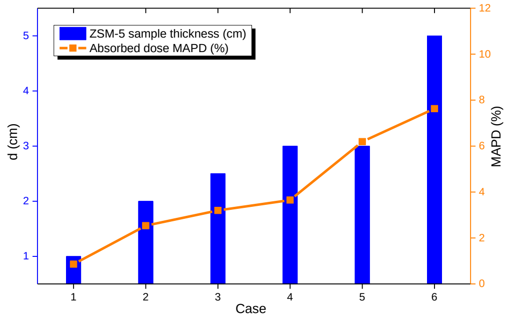

Fig. 7. Mean absolute percentage deviation of absorbed dose of ZSM-5 samples in the corresponding cases

For the zeolite ZSM-5 we examined six cases as shown in Fig. 6. to quantify the uniformity based on simulation data and calculated mean absolute percentage deviation (MAPD) of the absorbed dose in sample inside container. Simulation results in Fig. 7 shows that if the ZSM-5 sample thickness is less than 3cm and fully filled inside the container (Case 1, 2, 3, 4), the absorbed dose is uniform throughout the sample, with MAPDs are less than 3.7%. In Case 5, the sample is half filled the space inside the container, the remain is air, the absorbed dose is inhomogeneous, MAPD is about 6.2%. If the sample thickness is larger, the energy that the electrons impart in the sample is less uniform. As in Case 6, deviation is more than 7.6%.

This study performed simulations of different ZSM-5 densities (0.65-0.85 g/cm^3) for Case 4 in order to examine the influence of sample density on the uniformity of the absorbed dose. Results in Fig. 8 show no significant difference in the dose in the surveyed density range. The energy deposited is fairly uniform in the zeolite sample regardless of density in the range. The electron fluxes through sample layers are also equal. The deviation of these fluxes is about 1% and error is within the simulation error range (the simulation error is from 0.5% to 2% in the energy range of which we are interested).

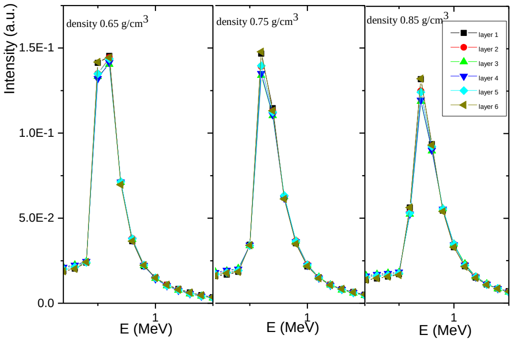

Fig. 8. The uniformity of energy absorbed in the ZSM-5 samples corresponding to different densities.

For mordenite, we have simulated six cases (see Fig. 9) for optimum sample thickness, ensuring uniformity in irradiation experiment and sufficient sample amount for post-irradiation survey measurement. Simulation results show that the more the sample amount is, the less uniformity of absorbed dose in the sample is. The sample thickness less than 1.4cm (7 layers, Case 4) is to ensure the uniformity of the electron energy imparted in the mordenite sample. If Case 4 is selected as the configuration for the mordenite irradiation experiment, the MAPD of absorbed dose will be about 6.6%. The corresponding value for Case 3 is 4.5% and for Case 5 is 10% (see Fig. 10).

Fig. 9. Simulation cases for mordenite sample. Case 1: 4 layers; Case 2: 5 layers; Case 3: 6 layers; Case 4: 7 layers; Case 5: 8 layers; Case 6: 10 layers

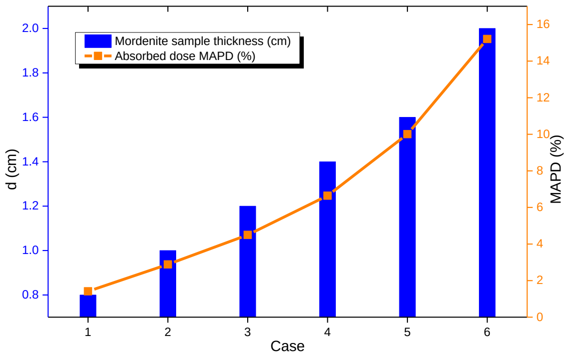

Fig. 10. Mean absolute percentage deviation of absorbed dose of Mordenite samples in the corresponding cases

The contribution of X-ray scattering from the shielding concrete around the irradiated area has also been investigated. The simulation results show that the contribution of scattered X-rays to the absorbed dose of the zeolite samples are from 0.9 to 1.2%, fairly small. On the other hand, the effect of the photon on the zeolite structure is unsignificant in comparison to the electron [9], so the contribution of the X-ray can be neglected.

2.3. *Irradiation time and temperature*

Finally, we used the simulation result of ZSM-5 (Case 4) to investigate the irradiation time and heat produced in the sample and aluminum shell. The normal operating intensity of the UERL-10-15S2 accelerator is 0.8 mA and beam size (diameter of beam spot at sample location) is 10 cm, so that the electron flux at the source is 6.366×10^{13} e/(cm^2.s). Thus, we have electron flux at container surface $f_o = 6.366 \times 10^{13} \times 2r/50 = 5.093 \times 10^{12}$ e/(cm^2.s), with r = 2 cm is radius of cylinder surface, and 50 cm is the scanning width of the beam. According to Eq.1, we have electron flux inside aluminum container $f_i = 6.880 \times 10^{12}$ e/(cm^2.s). For the target dose that causes a significant change in zeolite structure of about 10^{16} e/cm^2 [5], irradiation time should be 0.4 h.

Table 1

Parameters correspond to electron beam dose levels

Electron dose (e/cm^2)	Irradiation time	Total energy deposited in aluminum container (J)	Total energy deposited in ZSM-5 (J)	Temperature increasing rate in aluminum container (^{o}C/minute)
$1x10^{12}$	0.15 s	6.94	4.21	52.18
$1x10^{13}$	1.45 s	69.41	42.11	
$1x10^{14}$	14.53 s	694.13	421.05	
$1x10^{15}$	145.34 s	6941.29	4210.51	
$1x10^{16}$	0.40 h	69412.88	42105.05	

As indicated in Table 1 that total energy is deposited in the aluminum layer and in the ZSM sample corresponding to each electron dose level, by joule unit. Assuming that entire this energy converts to thermal energy. Because of aluminum thermal capacity of 880 J/kg.K and estimated aluminum container weight of 0.062 kg, the temperature increasing rate in aluminum container is 52.18°C/minute. At a high dose of 10^{16} e/cm^2, the temperature of this container increases beyond 600 °C after 0.4 hour. Unfortunately, the temperature increasing rate in ZSM-5 sample cannot be calculated because we lack of heat capacity of ZSM-5. However, the radiation energy absorbed by the ZSM-5 sample is significant smaller than the aluminum shell. Thus, the heat produced in this zeolite is expected to be lower than that of aluminum.

3. Conclusions

Based on the simulation results, it can be concluded as follow: Among common materials, aluminum is the best material to reduce electron beam energy. And the cylindrical aluminum container with a thickness of 0.5 cm is optimal for decelerating electron energy from 10 MeV down to 8 MeV. The energy distribution of decelerated electron reaches the peak at 8 MeV, concentrates in the range of 7.7-8.3 MeV and decreases rapidly on both directions. For ZSM-5 (density 0.65-0.85 g/cm^3), sample thickness less than 3 cm and fully filled inside aluminum container ensure homogeneity of the experiment with deviation less than 3.7%. For mordenite, the sample thickness for irradiating less than 1.2 cm is to ensure the uniformity of absorbed dose with deviation less than 6.6%. The heat generated during the irradiation in the aluminum shell and in the sample is moderate.

With the information obtained from the simulation process, we can set up the irradiation experiment: making of aluminum containers, irradiating zeolite samples with the desired doses. Samples of ZSM-5 and mordenite after irradiation will be investigated by means of positron annihilation spectroscopy (PAS), x-ray diffraction and some other physical and chemical methods in order to study the behavior of material structure under the effect of electron radiation. In addition, the simulation data gives us information about the absorbed energy in the zeolite sample. That will support the interpretation of later experimental results in a systematic way.

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