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Abstract

In heavy ion irradiation studies, the injected self-ion has recently been shown to suppress void nucleation in the ion deposition region. Previous theoretical calculations using steady state void nucleation theory have demonstrated that the predicted void number density is sensitive to small changes in the excess interstitial fraction, ε_i. In this paper the magnitude and distribution of ε_i is examined as a function of depth and incident ion energy for Ni irradiation of Ni. The magnitude of ε_i in the ion deposition region increases as the incident ion energy decreases. This increase is especially large below 4 MeV. The use of different electronic stopping power (esp) models in the damage calculations gave differences in ε_i of ~ 20% where the LSS esp gave higher results than the Brice esp. For low energy (< 5 MeV) ion irradiations there exists no part of the ion range free from the presence of excess interstitials while for the high energy (14 MeV) case the region less than 1.2 μm in depth appears to have a negligible ε_i value.

1. Introduction

The injected ions in a heavy ion irradiation damage study can affect the damage microstructure after they are deposited in the matrix. The injected ions come to rest in the solid as an interstitial without a vacancy partner. These excess interstitials have been shown to cause suppression of void nucleation and swelling in the ion deposition region. Brailsford and Mansur[1] first predicted that the injected ions would reduce the void swelling rate. This theoretical prediction has been expanded upon by Mansur[2,3] and also experimentally verified by Lee et al.[2] Plumton and Wolfer[5] have theoretically shown large reductions in void nucleation due to the excess interstitials. This reduction in the void number density in the peak damage region has been observed experimentally.[6,7] For example, Fig. 1 is a through range micrograph[7] of a Ni specimen irradiated at 450°C by 14 MeV Ni ions which illustrates the large suppression in void density possible in the ion deposition region. A review of the experimental evidence on the suppression effect of the injected interstitials has recently been presented by Garner.[8] The suppression increases whenever recombination is the dominant point defect loss mechanism and occurs at low temperatures and/or when the vacancy mobility is reduced by impurity trapping. Kumar and Garner[9] recently modeled the helium in dual ion irradiations as an additional excess interstitial because of the ability of helium in the matrix to trap a vacancy thereby freeing up an interstitial. This extra suppression to their void nucleation results suggests a possible explanation for previous experimental void number density anomalies.

The number of excess interstitials is a small fraction of the total number of damage produced interstitials (< 1%) so that the excess interstitials only become a significant portion of the interstitials reaching voids or void nuclei when most of the interstitials are recombining with vacancies. Previously it has been noted[5] that a factor of two difference in the excess interstitial fraction, ε_i (5×10^{-4} - 1×10^{-3}), can result in more than two orders of magnitude difference in the calculated void nucleation rate. That the inclusion of a few more hundredths of a percent to the total interstitial concentration can result in orders of magnitude differences in the nucleation rate indicates a highly nonlinear system. Calculating an accurate excess interstitial fraction is a necessity before good theoretical predictions on void nucleation and swelling during heavy ion irradiation can be obtained.

2. Theoretical Procedure

The damage rate or the excess interstitial fraction associated with a heavy ion irradiation can be calculated with damage codes such as the BRICE code[10] and the HERAD code.[11] From these codes one obtains an ion deposition distribution function, f(x), and a displacement energy distribution, $S_D(x)$. Both of these are a function of the depth, x, along the ion range. The displacement rate, I_D, can then be calculated by using a modified Kinchin and Pease model[12] where

$$I_D(x) = \frac{\phi K S_D(x)}{2\ \rho E_D} \quad . \tag{1}$$

Here ϕ is the incident ion flux, ρ is the atomic density and E_D is the effective displacement energy. To obtain accurate displacement values the displacement efficiency, K, should be taken as 0.3 in contrast to the traditional value of 0.8. A recent review by Kinney et al.[13] indicates that K is dependent on the incident ion energy, with K decreasing for increasing recoil energy. For high energy ($\gtrsim$ 1 MeV) neutron or heavy ion irradiations of FCC metals the efficiency is ~ 0.3 which reduces most previously cited damage values by a factor of 3/8. However, self-consistency requires the use of K = 0.8 since the low temperature work done to determine the fraction of defects escaping in-cascade recombination, E_{ff}, has already assumed K = 0.8.

The excess interstitial fraction has been taken[5] as the ratio of deposited ions to the interstitials produced by damage that survive in-cascade recombination. Therefore ε_i is

$$\varepsilon_i(x) = \frac{f(x)\phi}{E_{ff}\rho I_D(x)} \tag{2}$$

where E_{ff} is the fraction of defects that escape in-cascade recombination. The inclusion of E_{ff} into the formalism means that only those interstitials going to sinks or recombining after diffusion away from the cascade site are considered. This is a large reduction to the interstitial concentration since E_{ff} can be as low as 0.15[14] for FCC metals. The functional dependence of Eq. (2) can be seen through the use of Eq. (1). This gives ε_i as,

$$\varepsilon_i(x) = \frac{2\ E_D}{KE_{ff}} \frac{f(x)}{S_D(x)} . \tag{3}$$

The BRICE code and Eqs. (1) and (3) have been used to examine the interrelationship between ε_i and I_D for various incident ion energies for Ni on Ni. The distribution of $\varepsilon_i(x)$ as a function of depth is examined for decreasing ion energies. The values of ε_i versus incident ion energies are shown for various points along the ion range with the additional effect of two different electronic stopping power models, Brice[10] and LSS[15], included. Finally the two damage codes BRICE and HERAD are shown to affect the depth distribution of ε_i in significantly different ways. All damage code results are for a Ni on Ni heavy ion irradiation.

3. Results and Discussion

From Eq. (3) we observe competing trends. Both f(x) and $S_D(x)$ go through a maximum as the depth, x, is varied from the front surface to the end of the ion range. This can be observed in Fig. 2 where the BRICE code has been used to calculate the displacement value, Eq. (1), versus depth for 5 and 14 MeV Ni on Ni (solid line Fig. 2). Additionally it can be noted that as the incident ion energy is decreased, f(x) (dashed line Fig. 2) can completely overlap the damage profile. Plumton et al.(16) showed that for low energy ions this increased overlap causes increasing void nucleation suppression even though the displacement rate (i.e., $S_D(x)$) has increased.

The depth distribution of $\varepsilon_i(x)$ is shown in Fig. 3 for several incident ion energies. For consistency the $\varepsilon_i(x)$ values are plotted out to an end of range value coincident with a damage rate of ~ 10^{-6} dpa/s. For 0.5 MeV irradiations ε_i is extremely large which will give a large void suppression effect under even mild point defect recombination conditions. Under the appropriate irradiation conditions an excess interstitial fraction as low as 10^{-4} can have significant results.(5) Therefore, for Ni ions with incident energy $\lesssim 5$ there is no area free from the presence of the excess interstitials and free from the influence of the front surface. In contrast, for a 14 MeV ion irradiation, there exists a depth region from 0.4 μm to 1.2 μm where ε_i should have little effect.

Examination of Fig. 4, which compares ε_i as a function of depth between the two damage codes, BRICE and HERAD, shows a much larger ε_i value towards the front surface for the 14 MeV HERAD results as compared to the 14 MeV BRICE results. HERAD, which uses a more detailed physical modeling of the collision process coupled with the absence of any compromising assumptions regarding the solution of the transport equation, should result in a more accurate description of the ion deposition distribution function. The larger value of ε_i near the front surface for the HERAD results arises from a non-Gaussian shape for f(x) with a long tail towards the front surface. That a small value of f(x) should give such a large increase in $\varepsilon_i(x)$ also results from the decreasing value of $S_D(x)$ towards the front surface. The magnitude of ε_i, 10^{-6}-10^{-4}, that the 14 MeV HERAD code gives for the $\lesssim$ 1.4 μm depth region is only significant under conditions where point defect loss is extensively dominated by recombination (i.e., low temperatures). Therefore, the two damage codes will only give significantly different void nucleation and/or swelling results when the temperature is low and/or the vacancy mobility is reduced through impurity trapping. The 5 MeV results, Fig. 4, show again the trend of a larger value of $\varepsilon_i(x)$ towards the front surface for HERAD compared to BRICE calculations. Comparison between the 5 and 14 MeV HERAD results at the 1 μm depth, which is a typical depth for transmission electron microscopy analysis, shows that ε_i (5 MeV) is more than an order of magnitude larger than ε_i (14 MeV).

This low but non-negligible value of $\varepsilon_i(x)$ near the front surface might be responsible for some of the discrepancies observed between experimental results on nickel irradiated with 14 MeV Ni and the predictions of steady state void nucleation theory. Low temperature irradiations at 400°C(6) and 425°C(7) both showed that the void number density was suppressed for a depth of almost 2.5 μm. Void nucleation theory, using BRICE code data, predicted only ~ 1 μm(5,7) of suppressed region. Part of the discrepancy may be attributed to the BRICE code's use of a Gaussian distribution function. This Gaussian distribution gives too small a value for ε_i near the front surface when compared to the more accurate HERAD results.

The two remaining figures show BRICE code results for the log of ε_i versus incident ion energy. Figure 5 shows ε_i versus incident ion energy for the ion deposition peak and the damage peak. In the deposition peak, we see a smooth increase in ε_i as the ion energy decreases. In the damage peak, the competing trends that f(x) and $S_D(x)$ impose on $\varepsilon_i(x)$ cause a more complicated behavior. The dip in the ε_i values, at intermediate ion energies, occurs because $S_D(x)$ increases faster than f(x). In both peaks the LSS esp model gives higher ε_i values, ~ 20% greater than the Brice esp models. Finally, Fig. 6 shows $\varepsilon_i(x)$ versus ion energy for several damage rates in the ion deposition region. In all cases ε_i increases smoothly with decreasing ion energy. The 2 MeV ε_i value is about 50% larger than the 14 MeV ε_i value for the BRICE esp case, while the increase is ~ 70% for the LSS esp case. The LSS electronic stopping power models gives ε_i values 10-25% higher than the corresponding Brice esp model.

4. Conclusions

1) The excess interstitial fraction in the ion deposition region decreases with increasing ion energy which favors the use of higher energy bombarding ions.
2) The use of the Brice electronic stopping power model gives a lower excess interstitial fraction than the LSS model in the damage and ion deposition peak.
3) For incident ion energies $\lesssim$ 5 MeV there exists no part of the ion range free from the presence of excess interstitials and is at the same time sufficiently far from the front surface to avoid surface phenomena.

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VOID SUPPRESSION IN NICKEL
DUE TO INJECTED INTERSTITIALS

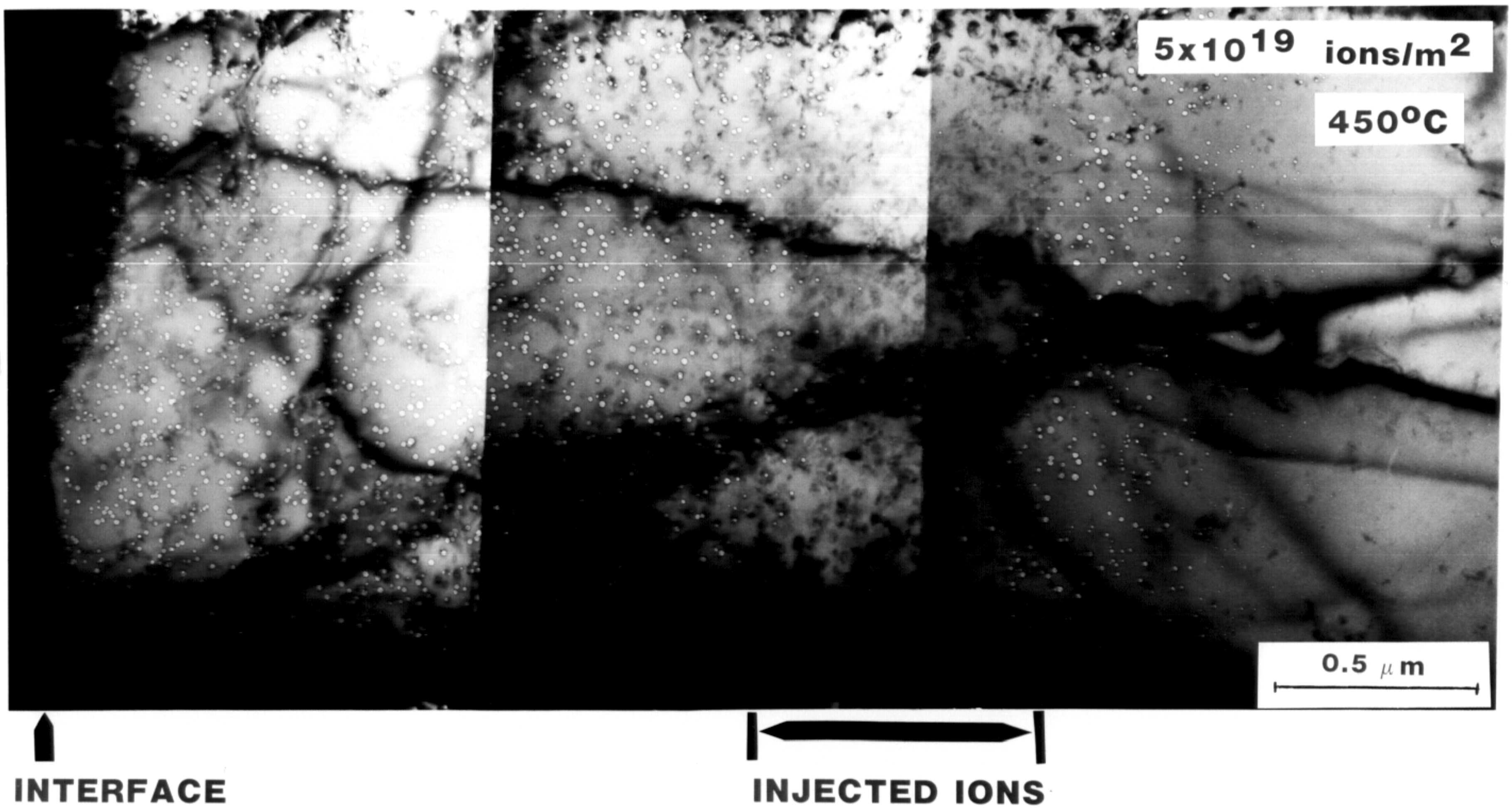

Fig. 1. Depth-dependent void microstructure of nickel irradiated with 14 MeV Ni ions to a peak damage level of 5 dpa (K = 0.8) at 450°C. Note the absence of voids in the implanted ion region.

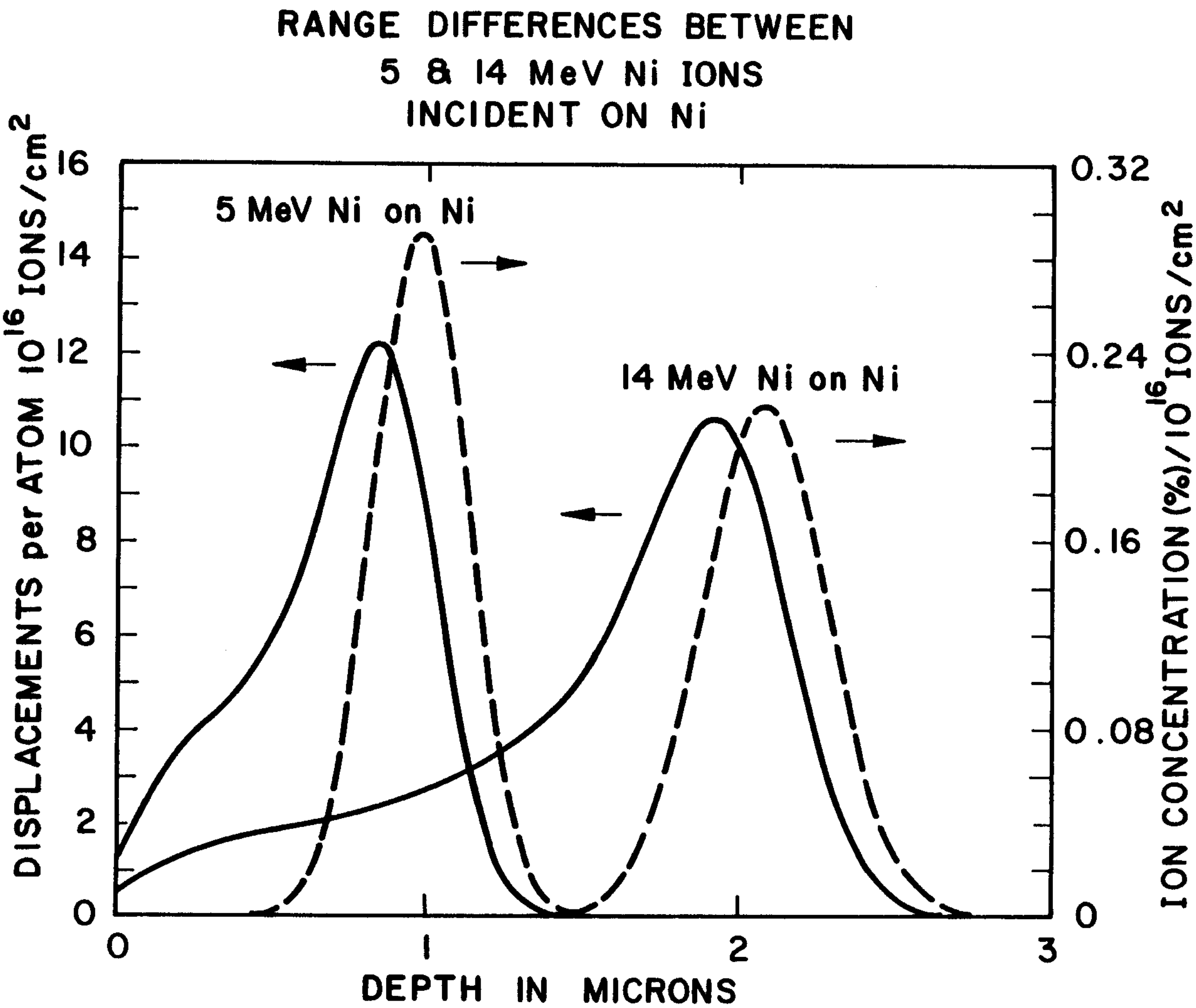

Fig. 2. Displacement damage and implanted ion concentration (atomic %) versus depth for 5 and 14 MeV Ni on Ni where the BRICE code calculation used the LSS esp, E_D = 40 eV and K = 0.8.

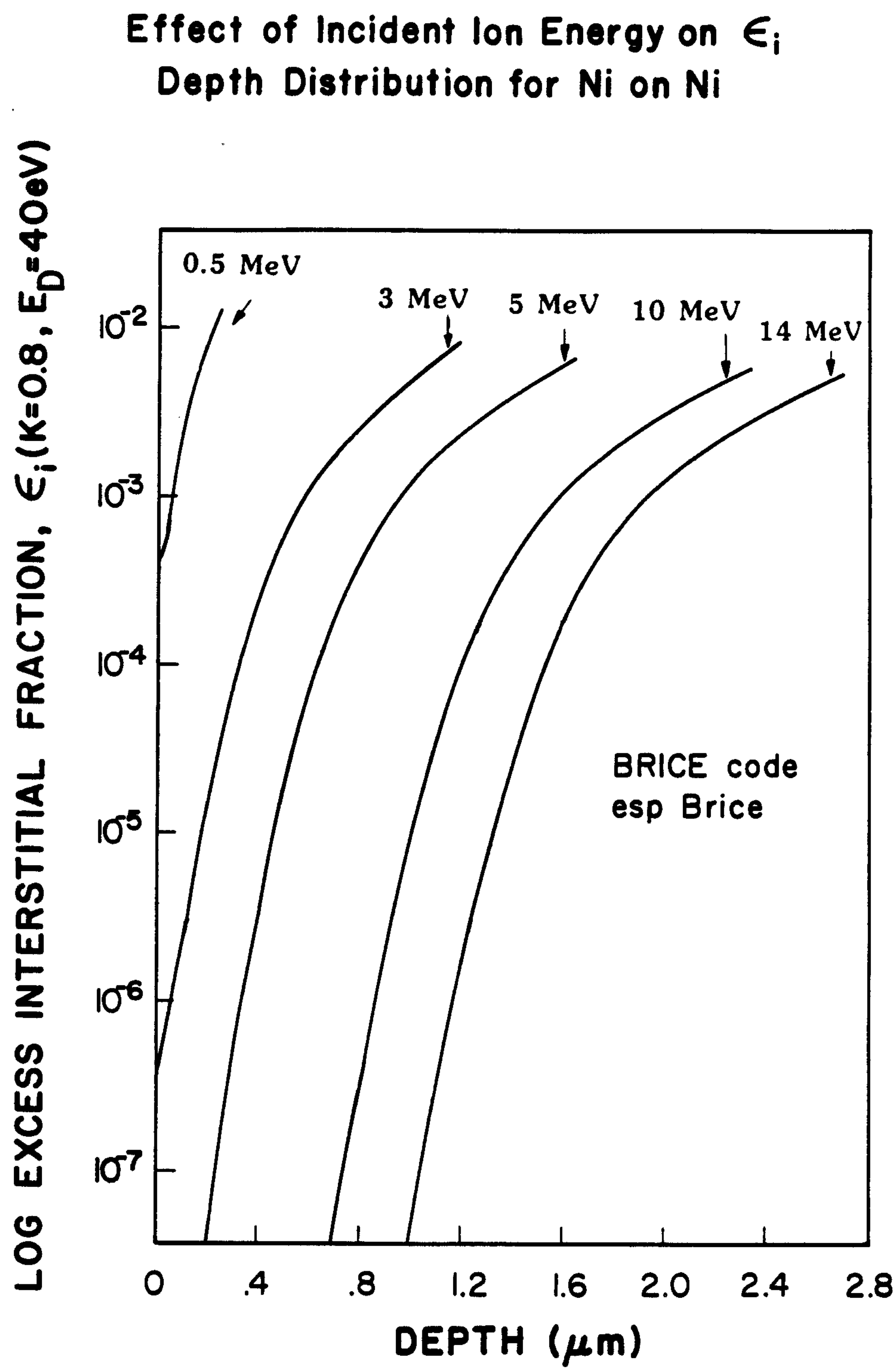

Fig. 3. Log ϵ_i versus depth for Ni on Ni at several incident ion energies.

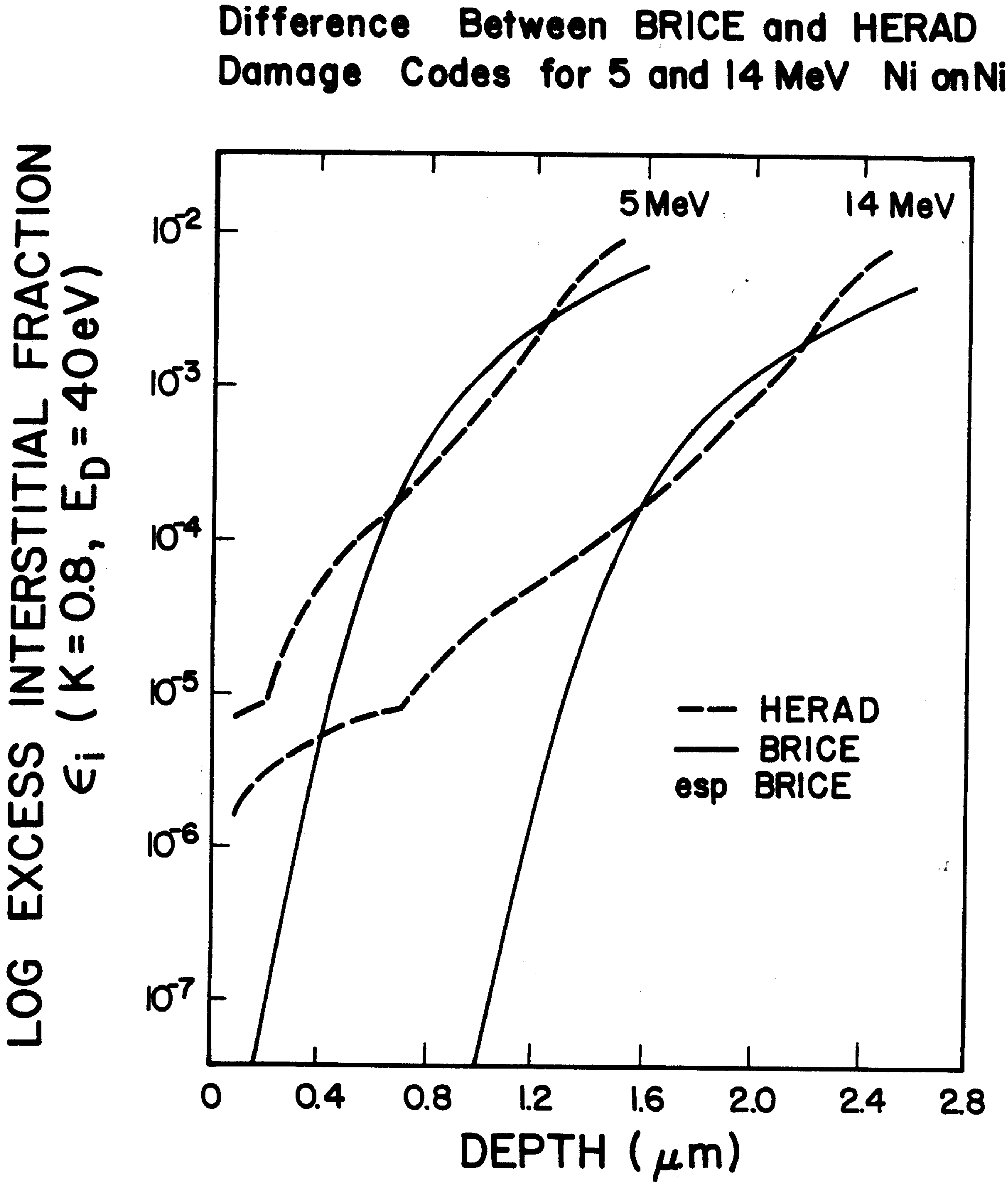

Fig. 4. Log ε_i versus depth for 5 and 14 MeV Ni on Ni where the BRICE and HERAD damage codes are compared.

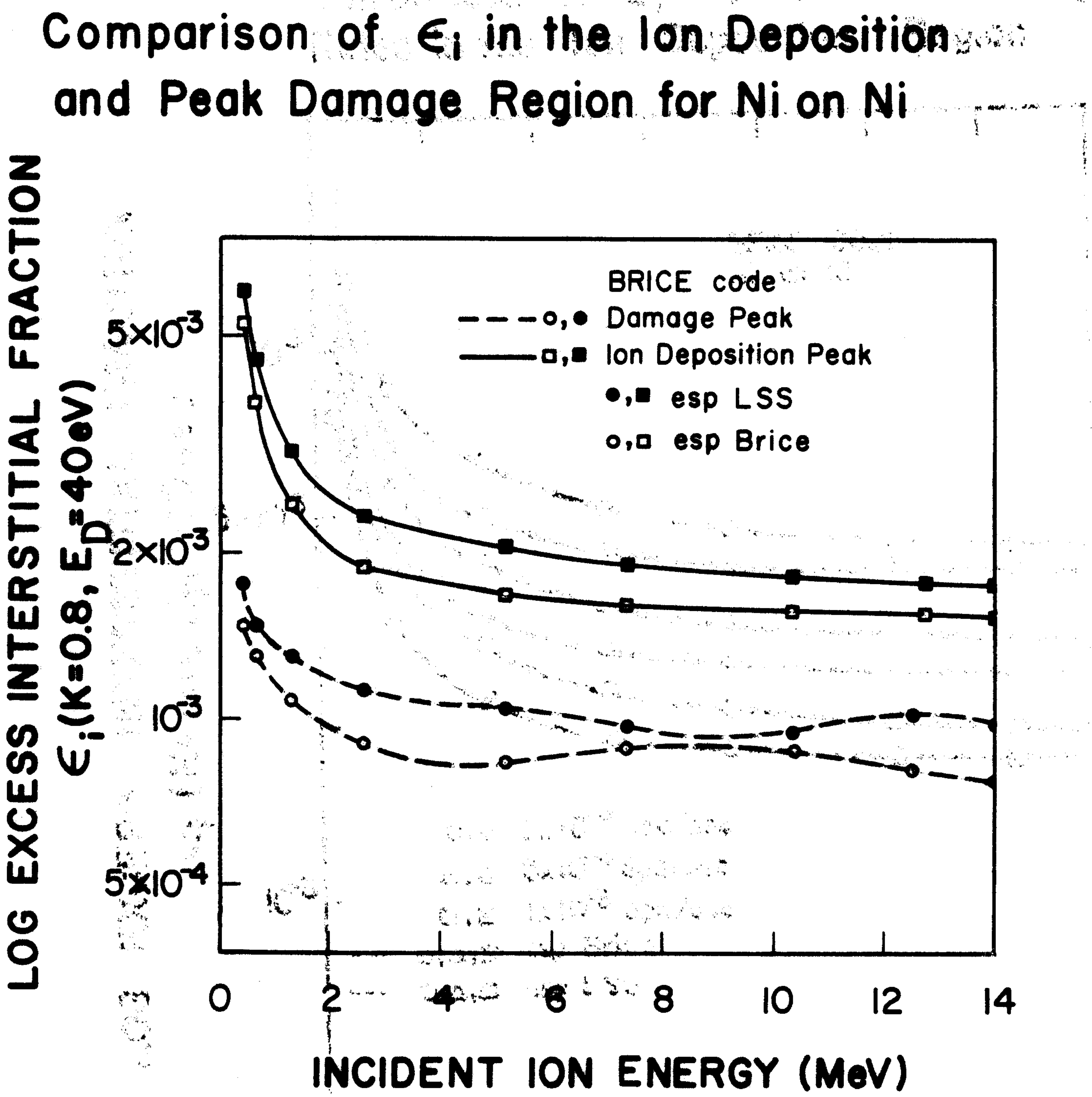

Fig. 5. Log ϵ_i versus incident ion energy for Ni on Ni at the damage peak and at the ion deposition peak.

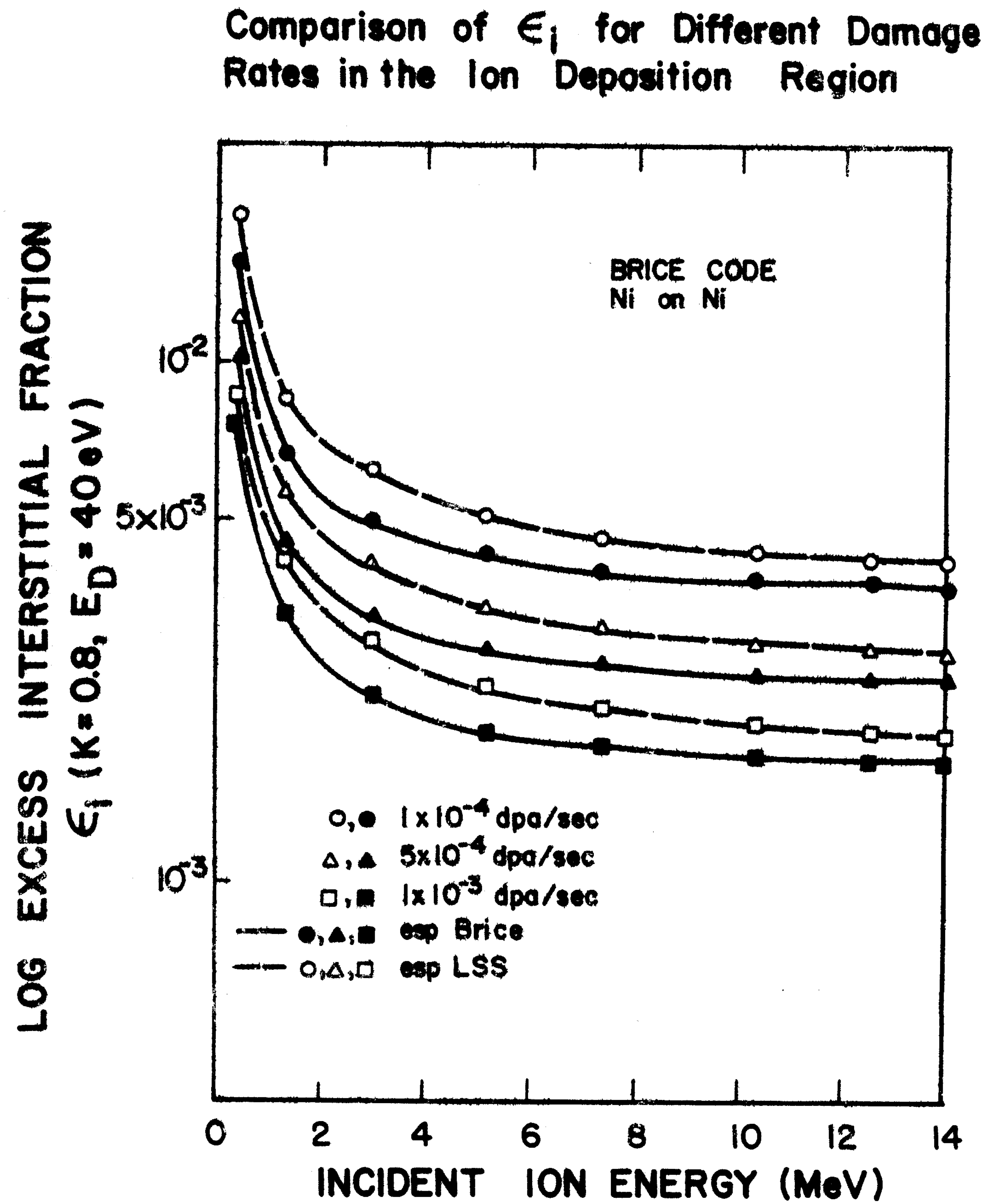

Fig. 6. Log ϵ_i versus incident ion energy for Ni on Ni at various constant damage rates on the backside of the ion deposition profile.