Grain size dependence of fracture toughness and crack-growth resistance of superelastic NiTi

Aslan Ahadi a,b, Qingping Sun a,*

a Department of Mechanical and Aerospace Engineering, The Hong Kong University of Science and Technology, Clear Water Bay, Hong Kong
b School of Civil Engineering, State Key Lab of Water Resources and Hydropower Engineering, Wuhan University, China

A R T I C L E   I N F O

Article history:
Received 12 August 2015
Received in revised form 10 October 2015
Accepted 22 October 2015
Available online xxx

Keywords:
Shape memory alloys
Martensitic phase transformation
Microstructure
Toughness
Intrinsic and extrinsic toughening mechanisms

A B S T R A C T

The grain size (GS) dependence of fracture toughness ($K_{IC}$) and static crack-growth resistance ($K_{CR}$) of superelastic NiTi with average GS from 10 to 1500 nm are investigated. The measurements of strain and temperature fields at the crack-tip region are synchronized with the force–displacement curves under mode-I crack opening tests. It is found that with GS reduction down to nanoscale, the $K_{IC}$ and the size of crack-tip phase transformation zone monotonically decrease and the $K_{CR}$ changes from a rising to a flat R-curve. The roles of intrinsic and extrinsic toughening mechanisms in the GS dependence of the fracture process are discussed.

© 2016 Elsevier Ltd. All rights reserved.

Due to a reversible martensitic phase transformation (PT), superelastic (SE) NiTi shape memory alloys (SMAs) are widely used in a variety of applications. However, the narrow temperature window of superelasticity [1], poor cyclic stability [2], and poor fatigue life [3], significantly limit the applications of SE NiTi. In recent years, significant steps have been taken to break the above limitations. Introducing defects via doping [4,5], addition of Nb nanowires to NiTi [6], and GS reduction down to nanocrystalline (nc) regime [7] are unique ways to achieve novel properties. It has been shown that with GS reduction to nc regime, SE NiTi indeed exhibits novel properties such as linear elastic, and numerical researches have been published [12,15–20].

The fracture toughness ($K_{IC}$) and crack-growth resistance ($K_{CR}$) are essential properties for successful application of metals and alloys [12]. It is well-known that with GS reduction to nc regime, metals exhibit lower $K_{IC}$ due to limited ductility [13]. Owing to a stress-induced martensitic PT, the most distinctive feature in the fracture process of SMAs is the formation of a kidney-like crack tip PT zone which can transform back at the crack wake [14]. In this regard, several experimental, analytical, and numerical researches have been published [12,15–20]. However, the GS effects on the fracture behavior and intrinsic/extrinsic toughening mechanisms of SE NiTi remain to be unveiled by systematic experiments. In this paper, we have studied the $K_{IC}$ and $K_{CR}$ behaviors of SE NiTi with average GS from 10 to 1500 nm. The strain and temperature fields at the crack-tip region and the crack wake are synchronized with the force–displacement curves by in-situ digital image correlation (DIC) and in-situ infrared (IR) thermography under the mode-I crack opening tests.

SE NiTi (Ti-50.9 at.% Ni) sheets with thickness of 1.7 mm were sandwiched in stainless steel sheets and were cold-rolled to 42% of thickness reduction. The as-rolled sheets were heat-treated at temperatures of 250 °C for 45 min, 520 °C for 2 min, 520 °C for 3 min, 520 °C for 6 min, and 600 °C for 45 min followed by quenching in water [7,8,11]. The average GS of the cold-rolled and heat-treated specimens was measured with TEM (see Fig. S1) and Williamson–Hall method. Dog-bone tensile specimens with thickness of 1 mm, width of 2 mm, and gauge length of 30 mm were cut along the rolling direction and tested to obtain stress–strain curves (see Fig. S2b) at a strain rate of $1 \times 10^{-4} \text{s}^{-1}$ using an MTS (UTM-RT/10) machine. Transformation temperatures were determined using differential scanning calorimeter (DSC-TA Q1000) with heating/cooling rate of 10 K/min.

Notched compact tension (CT) specimens with the dimension shown in Fig. S3a were EDM cut from the cold-rolled and heat-treated sheets. The highly polished CT specimens were pre-cracked (see Fig. S3b) with a MTS-858 table top machine at a frequency of 5 Hz with sinusoidal wave function under decreasing load amplitudes ($\Delta P_a < \ldots < \Delta P_1$) as suggested by ASTM E 561-10 ($\Delta P = P_{\text{max}} - P_{\text{min}}$). The load ratio of $R = P_{\text{max}} / P_{\text{min}} = 0.1$ was used (see inset in Fig. S3c). Different lengths of pre-cracks with $7.6 < a < 10.29$ mm ($0.4470 < a/W < 0.6053$ mm) were obtained. The crack length was monitored ex-situ using a Leica microscope. The strain and temperature fields at the vicinity of the crack tip...
were synchronized using in-situ DIC and IR thermography. The experimental setup for mode-I crack opening tests is shown in Fig. S3c. To create a fine and random pattern needed for strain field measurements using DIC, silicon micro-particles (with average size of 1 μm) were speckled to the specimen surface (Fig. S4). Ncorr open source 2D DIC was used for DIC calculations [21]. A fast multi-detector IR thermal camera (FLIR SC7700BB) equipped with a close-up lens with working distance of 30 cm and field of view of 9.6 × 7.2 mm² was used to record the temperature field at the crack-tip region. To achieve a uniform black body, one side of the specimens’ surface was covered with candle fume. The mode-I crack opening tests were performed at the loading rate of 5 mm/min in order to capture the temperature variations at the crack-tip region. The fractography studies were performed in a JEOL-JSM 6390 SEM.

The transformation temperatures measured from DSC (see Fig. S2a) are summarized in Table 1. It is seen that all the transformation temperatures ₐ₄ (austenite finish temperature), ₘ₄ (martensite start temperature), and ₘ₅ (martensite finish temperature) decrease with GS reduction. The effects of GS on the isothermal stress–strain tensile properties are shown in supplementary materials (Fig. S2b). It is seen that all the specimens are SE as they can recover large strains of 5%. With GS reduction to nc regime (GS < 60 nm) the stress–strain response changes from a plateau-type superelasticity to hardening SE response with reduced hysteresis loop area [9]. The critical transformation stress (储备), determined from the cross-cutting of the tangent lines in the elastic and PT regions (see Fig. S2b), increases from 381 MPa for GS = 1500 nm to 655 MPa for GS = 10 nm. The gradual increase of 储备 with GS reduction to nc indicates a gradual reduction in the ductility.

Table 1
The GS dependence of phase transformation properties of SE NiTi.

<table>
<thead>
<tr>
<th>Heat treatment</th>
<th>Average GS (nm)</th>
<th>ₐ₄/ₐ₅(°C)</th>
<th>ₘ₄/ₘ₅(°C)</th>
<th>ₐ₄(MPa)</th>
<th>ₐ₁(MPa)</th>
<th>ₐ₂(MPa)</th>
<th>ₐ₃(MPa)</th>
<th>ₐ₄(MPa)</th>
<th>ₐ₅(MPa)</th>
<th>ₐ₆(MPa)</th>
<th>ₐ₇(MPa)</th>
<th>ₐ₈(MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cold-rolled</td>
<td>10</td>
<td>N/A</td>
<td>N/A</td>
<td>655</td>
<td>N/A</td>
<td>502</td>
<td>1465</td>
<td>4.6</td>
<td>3.8</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
</tr>
<tr>
<td>250 °C − 45 min</td>
<td>18</td>
<td>N/A</td>
<td>N/A</td>
<td>502</td>
<td>1465</td>
<td>4.6</td>
<td>3.8</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
</tr>
<tr>
<td>520 °C − 2 min</td>
<td>42</td>
<td>−12.7/8</td>
<td>−60/−86</td>
<td>472</td>
<td>1452</td>
<td>49.8</td>
<td>6.9</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
</tr>
<tr>
<td>520 °C − 3 min</td>
<td>64</td>
<td>−12.5/6</td>
<td>−55/−78</td>
<td>461</td>
<td>1457</td>
<td>46.3</td>
<td>9.5</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
</tr>
<tr>
<td>520 °C − 6 min</td>
<td>80</td>
<td>−12.5/6</td>
<td>−55/−78</td>
<td>461</td>
<td>1457</td>
<td>46.3</td>
<td>9.5</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
</tr>
<tr>
<td>600 °C − 45 min</td>
<td>1500</td>
<td>−46/3</td>
<td>−30/−33</td>
<td>381</td>
<td>527</td>
<td>45</td>
<td>13.11</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
<td>5.0</td>
</tr>
</tbody>
</table>

When using Eq. (1) one has to note that this equation is only valid for plane strain condition where the size of the PT zone is much smaller than the specimen thickness. Looking at the size of PT zone in Fig. 2, it is seen that this condition is barely met in our experiments. Therefore, the values of ₖₜ and ₖₙ reported here are nominal and only serve as first approximations and are used for comparative purpose. More exact calculations such as using finite element method is needed to determine the K values. For each GS, the measured ₖₜ averaged over eight different values of a/W are shown in Fig. 1b. For GS = 1500 and 80 nm, the ₖₚ values are 46.3 MPa/m and 42.4 MPa/m, respectively. These values are consistent with the reported values for SE NiTi of the similar microstructure and stress–strain curves [16,23]. It is seen that when GS is reduced below 80 nm, the value of ₖₚ monotonically decreases and reaches 25.4 MPa/m for GS = 10 nm. Fig. 1c shows the GS dependence of crack-growth resistance ₖₚ (R-curve) as a function of crack extension (a − ad). For GS = 1500 and 80 nm, the ₖₚ keeps increasing (known as rising R-curve) with crack extension and eventually saturates. However, for GS = 18 and 10 nm, the R-curves become flat. In other words, more energy is dissipated during crack propagation for SE NiTi with GS = 1500 and 80 nm while for the GS = 18 and 10 nm the fracture behavior resembles those of brittle materials.

The fracture surfaces of the CT specimens are shown in Fig. 1d. The fracture surface of the specimen with GS = 1500 nm mainly shows dimples indicating a dominant ductile fracture. For GS = 80 nm the fracture surface is characterized by a mixture of dimples and cleavage planes. With further GS reduction, the tendency of the cleavage increases in the fracture process as it is shown for GS = 42 and 10 nm. Therefore, based on the consistent observations of (1) stress–strain behavior (2) crack-tip inelastic zone size (3) ₖₚ curves and (4) the SEM observations, it is suggested that the failure mode transitions from ductile to brittle fracture when GS is reduced down to nanoscale.

The effects of GS on the strain (储备) distribution around the crack-tip region taken at ₚₜ in shown in Fig. 2. It is seen that the strain field at the crack-tip vicinity is in the shape of two lobes. For GS = 1500 and 80 nm the lobes are inclined to an angle of about 48° and 51° from the crack line, respectively. The angle of inclination gradually increases to 63° for GS = 10 nm. Moreover, the DIC images indicate that with GS reduction the size of the lobe at ₚₜ immediately prior to the crack growth decreases. From the plot of储备 as a function of the distance from the crack tip, as shown in Fig. 2, one can determine the size of the PT zone along the x-axis ₐₖ→ₐₜ by referring to the stress–strain curves [16,32]. For GS = 1500 nm the 0.25 ≤ ₐₖ→ₐₜ ≤ 1.7 mm at the crack tip region and with GS reduction the ₐₖ→ₐₜ monotonically decreases to 0.23 ≤ ₐₖ→ₐₜ ≤ 0.81 mm for GS = 80 nm and 0.19 ≤ ₐₖ→ₐₜ ≤ 0.48 mm for GS = 64 nm. The measured

\[ f(a/W) = \left( \frac{P_{\text{max}}}{B\sqrt{W}} \right) \left( \frac{2 + a/W}{(1-a/W)^{3/2}} \right) f(a/W) \]

where ₚₜ is the applied maximum force, ₐ is the specimen thickness, ₐ is the specimen width measured from the load line, a is the physical crack length (see Fig. S3a), and f(a/W) can be expressed as (see ASTM E 561-08):

\[ f(a/W) = \left[ 0.886 + 4.64(a/W) - 13.32(a/W)^2 + 14.72(a/W)^3 - 5.6(a/W)^4 \right]. \]
values of $\epsilon_{\text{tr}}$ are consistent with the published data on nano-grained SE NiTi [16]. The GS dependence of $r_{A \rightarrow M}$ and stress distribution can be readily explained by the theoretical models of stress-induced PT at the crack tip region [18,24]. When the size of PT zone is small compared with the size of the specimen (small scale PT), the size and the shape of PT zone for each GS can be approximated as [25]:

$$r_{A \rightarrow M}(\theta) = \frac{1}{2\pi} \left[ \frac{K_{\text{app}}}{\sigma^2} \right]^2 \cos^2 \frac{\theta}{2} \left[ 1 + 3 \sin^2 \frac{\theta}{2} \right]$$

where $\sigma_{\text{tr}}$ is the critical transformation stress and depends on the GS and $K_{\text{app}}$ is the applied stress intensity factor at the crack tip. For a stationary crack $= K_{C}$, according to this equation, $r_{A \rightarrow M}$ scales with $(K_{\text{app}}/\sigma_{\text{tr}})^2$. Therefore, for a given $K_{\text{app}}$ a gradual reduction of $r_{A \rightarrow M}$ with GS reduction is expected since $\sigma_{\text{tr}}$ increases significantly with GS reduction. Another effect of GS reduction is that for a given $K_{\text{app}}$ the stress levels in the PT zone and plasticity zone are higher due to higher $\sigma_{\text{tr}}$ and $\sigma^2$ as schematically shown in Fig. S5. A direct consequence of such higher stress levels at the crack tip region is that the fracture behaves more like brittle materials and has features of brittle fracture such as a flat R-curve.

The synchronized IR thermographic images of the front surface of the specimens at $P_{\text{max}}$ (loading rate of 5 mm/min) are shown in Fig. 3a. It is seen that the crack-tip region is warmer than the rest of the specimen due to release of PT latent heat and heat transfer via conduction and convection [2,26,27]. The figure shows two important features. First, with GS reduction the size of the hot zone decreases which is consistent with the DIC measurements of $r_{A \rightarrow M}$ (Fig. 2a). Second, under the given loading rate of 5 mm/min, the magnitude of the temperature increase in the PT zone decreases with GS. For example, the magnitude of the temperature rise at the crack tip is 5.83 °C for GS = 1500 nm and it monotonically decreases to 1.02 °C for GS = 18 nm, and 0.03 °C for a GS = 10 nm. This is due to the reduction of latent heat with GS especially for GS = 18 nm [see Fig. S2b] [7–9]. Fig. 3b shows the thermographic images during crack propagation. Comparing Fig. 3b with 3a, one can note a remarkable increase in the size of the crack-tip hot zone for GS = 1500 and 80 nm. This is consistent with the rising R-curve characteristic of the two specimens. Furthermore, it is clearly seen that once the crack propagates it leaves two cold regions behind with temperatures lower than the room temperature (black arrows). For the GS = 1500 nm the minimum temperature in the crack wake is 15.5 °C which is 4.5 °C below the room temperature. This cooling effect is due to a rapid absorption of latent heat and provides a compelling evidence to the reverse PT when the crack passed by [26]. For the same reasons, the cooling in the wake becomes also weaker with GS reduction.

To elucidate the effects of GS on the controlling mechanisms of $K_{C}$ and $K_{P}$, a qualitative discussion of the intrinsic and extrinsic toughening mechanisms at the crack-tip region and crack wake [28] are given in Fig. 4a and b, respectively. For the intrinsic mechanisms, it is well-known that the most dominant toughening mechanism in SE NiTi is the formation of stress-induced martensite at the crack-tip region which act as a shielding mechanism. According to Baxevanis et al. [12, 15], the degree of the toughening due to the crack-tip PT depends mainly on the dimensionless parameter $\alpha = \epsilon_{\text{tr}}/\epsilon_{\text{M}}(\sigma_{\text{tr}}/E_{\text{A}})$ (ratio of dissipated to stored energy) where $\epsilon_{\text{tr}}$ is the transformation stress, $\epsilon_{\text{M}}$ is the transformation strain, and $E_{\text{A}}$ is the Young’s modulus of austenite. The higher value of $\alpha$ is beneficial for toughness enhancement [25]. As such, the gradual decrease of $\alpha$ (due to a gradual increase of $\epsilon_{\text{M}}$) is indeed the main reason for a gradual reduction of $K_{C}$ [29]. Another factor that needs to be considered is the GS dependence of the plastic yield’s stress of the martensite (\sigma^M). As can be seen in Table 1, the \sigma^M increases with GS reduction to nc. In the same way as that of the elastic–plastic

---

**Fig. 1.** (a) GS effects on the force–displacement curves during mode-I monotonic crack opening fracture tests, (b) GS effects on the $K = K_{C}$ (at $P_{\text{max}}$) for specimens of different initial crack lengths (with 0.4470 < a/W < 0.6053 mm), (c) GS effects on the crack-growth resistance ($K_{sg}$), and (d) top-view SEM images of the fracture surface of CT specimens.
materials, a higher yield's stress indicates a smaller size of the plastic deformation zone and higher stress levels (see Fig. S5) at the crack tip leading to less shielding effect and toughness enhancement. However, it is important to note that the effect of plastic deformation zone at the crack tip on the shielding and dissipation is believed to be small compared with the dissipation due to PT [12]. In addition, with GS reduction, the tortuous crack-path configuration (see Fig. 4a1) becomes straight (see Fig. 4a2). This causes a change in the mode of $K$ from mix of mode-I and mode-II in the former, to near mode-I in the latter. This leads to a decrease of the threshold (critical) driving force for crack propagation that in turn leads to reduction of $K_{IC}$ [30].

The effects of GS on the extrinsic mechanisms are schematically shown in Fig. 4b1 (for GS $\geq 80$ nm) and Fig. 4b2 (for nc NiTi). As it is seen in Fig. 4b, during crack propagation the crack wake undergoes a complete cycle of forward and reverse PT (see also Fig. 3b for the thermographic evidence of the reverse PT at the crack wake). The toughness enhancement $\Delta K = K_f - K_{IC}$ for steady-state crack propagation due to such forward/reverse PT or energy dissipation can be described by the following equation [31]:

$$K_f^2 = K_{IC}^2 + \left[\frac{2E}{(1-\nu^2)}\right]\int_0^H U^f(y)dy$$

(4)

where $E$ is the elastic modulus, $\nu$ is the Poisson's ratio, $H$ is the height of the crack wake, and $U^f (= f(\sigma - \varepsilon)$ is the specific mechanical energy dissipated in the wake and is represented by the $\sigma - \varepsilon$ hysteresis loop area. From Eq. (4) it is clearly seen that the gradual change of $K_f$ with GS reduction (from a rising R-curve to a flat R-curve) is not only caused by a reduction of $H$ due to the increase of $\sigma_f$, but also caused by a gradual reduction of the stress hysteresis ($U^f \rightarrow 0$) of the material [7].

In summary, the grain size (GS) dependence of fracture toughness ($K_{IC}$) and static crack-growth resistance ($K_f$) of SE NiTi were investigated by synchronized measurements of the force–displacement curves and the crack-tip strain and temperature fields. It is shown that with GS reduction to nanoscale, the $K_{IC}$ gradually decreases and $K_f$ transitions from rising to flat R-curve. The in-situ measurements showed a monotonic reduction in the size of the phase transformation zone at the crack-tip region. Using the controlling parameters of the fracture process of NiTi SMAs and their grain size dependencies, the intrinsic and extrinsic toughening mechanisms of the fracture behavior of the NiTi were analyzed and discussed. The results provide a better understanding of the fracture behavior of nanocrystalline superelastic NiTi which possesses exceptional thermomechanical properties.

The authors are grateful for the financial support of this work from the Hong Kong Research Grant Council (RGC) through the GRF grant (project no. 16214215) and the 973 Program of China (project no. 2014 CB046902). We also thank Maziar Jamishidi for DIC data analysis and Hamed Mokhtari for helping with thermographic experiments.

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scriptamat.2015.10.036.
References