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Acoustic emission and self-organized criticality associated with fracture processes during hydrogen precipitation in niobium

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Abstract

Acoustic emission activity was detected in niobium during hydrogen precipitation from solid solution. The observed emission is attributed to the nucleation and propagation of microcracks generated by the misfitting growing precipitate particles. Analysis of the acoustic signals reveals that the distribution of their amplitudes follows the Gutenberg–Ritcher law of earthquakes $N(A'>A)\sim A^{-c}$, $c=0.9$. The observed power law indicates that during the fracture processes caused by the precipitation, the Nb–H system goes into a self-organized critical state, similar to what occurs in the earth's crust during earthquakes.

1. Introduction

Recently, Bak *et al.* [1] have shown that in nature a large variety of dynamic composite systems organize themselves into a stationary critical state (SOC) characterized by spatial and temporal correlation functions following power laws [1–3].

The most relevant example of a SOC system is the earth's crust where earthquakes have a self similar distribution described by the Gutenberg–Ritcher law [4] characterized by a universal value of exponent, in the sense that it does not depend on the particular geographic area. Other examples of SOC behaviour are current fluctuations through resistors [1], the random steplike changes in the magnetization of ferromagnetic materials [5], and underground ultrasonic signals from rock fractures following primary activity of the Stromboli volcano [6].

The intense research activity presently conducted on SOC behaviour is based mostly on numerical simulations and simple physical models. The best known model, also very useful for pedagogical purposes, is the sandpile [1, 7], to which sand grains are continuously added, one grain at a time, until the pile reaches a critical stationary slope; in the critical state a single grain can provoke an avalanche of any size including a "catastrophic" event. Other cellular automaton models [3, 8, 9], inspired by the Burridge–Knopoff [10] model of earthquakes, are systems of blocks interconnected by springs and pulled on a frictional plane. The block system evolves to a critical state in which groups of blocks can slide simultaneously giving rise to a slip event of any size.

As the origin of earthquakes is associated with fracture processes in the earth's crust, the question arises whether fracture events in stressed materials also display SOC. To date, little attention has been devoted to this aspect; the only report of this kind is a reference by Chen *et al.* [3] to unpublished work showing a power-law distribution of fracture events in stressed Al and Nb bars.

It is well known that plastic deformation and fracture phenomena occur in metal–hydrogen systems during precipitation of hydrogen from solid solution (α-phase), owing to accommodation of the misfitting precipitate particles (β-phase) which generally have a density different from that of the metal matrix (12% volume increase in Nb–H [11]). It has been reported [12] that the nucleation and propagation of microcracks during hydride precipitation is accompanied by intense acoustic emission activity (AE) caused by a sudden release of elastic energy. In the present investigation the AE signals emitted during hydride precipitation in Nb–H were analysed to see whether this system exhibits SOC behaviour.

2. Experimental details

The samples were five discs (30–36 mm in diameter, 3–6 mm thick) of 99.9% pure polycrystalline Nb from

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different manufacturers, hereafter labelled Hr 2, Hr 4, K1, K2, Hy, having masses 17.8, 17, 29.5, 45, and 38.5 respectively. Hydrogen doping of the samples was conducted at 550 °C in a controlled pure H_2 atmosphere. The doping was preceded by a thermal treatment at 800 °C for 2 h in a vacuum of 5×10^{-4} Pa. The H contents were determined by the variation in mass of the samples, both after the H charge and after vacuum extraction subsequent to the AE experiments. The H concentrations of the specimens (ranging from 3.7 to 7.8 at.%) were chosen so that hydride precipitation, stimulated by decreasing temperature, occurred in the temperature range 330–220 K. The H precipitation temperature T_t was determined either by the solvus line of the Nb–H system [13] or by the sharp increase in the elastic energy dissipation and modulus occurring at the onset of the α–β transformation [14, 15].

The AE signals were detected during the first cooling after the H charge while the temperature (measured by an iron–constantan thermocouple attached directly to the specimen) was decreased at a constant rate of either 1 or 4 K min^{-1}. The AE set-up consisted of a PZT transducer coupled to the specimen by silicone grease, a low-noise preamplifier (40 dB gain), and a high-pass filter at 100 kHz whose output was connected to two parallel processing chains. The first chain consisted of a postamplifier (60 dB gain), a 100–300 kHz bandwidth filter (tuned to the maximum sensitivity of the PZT), and a counter (threshold level 1 V) giving the count rate $\dot{N}_r$ of all decaying oscillations (ringdown) of the PZT. The second chain was commenced with an amplitude detector (with adjustable threshold put at 30 dB) giving an output proportional to the log of the highest single peak of the whole decaying oscillation (burst); then the signals were sent to a multichannel analyser providing the total number of bursts N_e and their distribution in 100 channels according to their amplitude. The parameter $\dot{N}_r$ is more sensitive in revealing the onset of AE activity, while the N_e count has a physical meaning in as much as burst is correlated with an event occurring in the material. A burst signal occurring more than 10 ms after the level of the preceding burst had decayed below the threshold level was counted as a separate event.

3. Results and discussion

The total number of events N_e and the count rate of ringdowns $\dot{N}_r$, recorded in sample Hr2 during the first cooling (1 K min^{-1}) after the H charge (5.8 at.% H), are reported in Fig. 1. The N_e background of the H-free sample is displayed in the same figure. Acoustic signals started at a temperature somewhat lower than that of the onset of hydride precipitation T_t and had

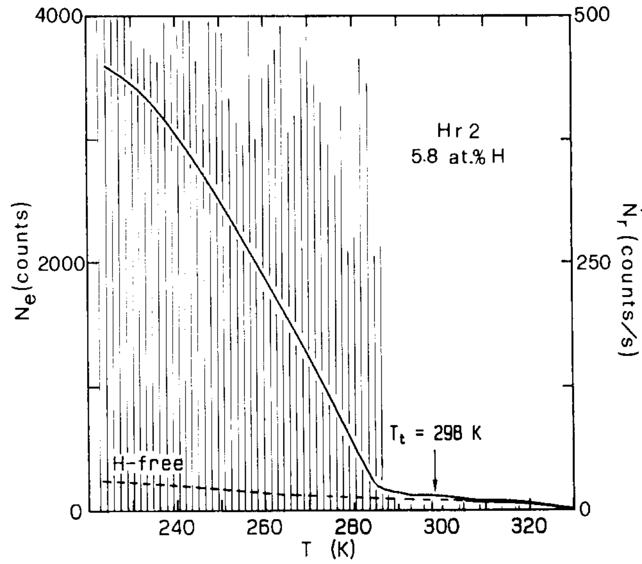

Fig. 1. Total count of bursts N_e (solid line) and ringdown count rate $\dot{N}_r$ (bar graphs) recorded from sample Hr2 during the first cooling (1 K min^{-1}) after the H charge. The arrow indicates the temperature T_t of the onset of hydride formation. The dootted line represents the burst count in the H-free sample.

the character of isolated intense burst with peak amplitudes up to 40 mV; similar behaviour was observed in all the tested samples. It has been demonstrated [12] that the observed emission is due to the nucleation and growth of microcracks and not to dislocation multiplication accompanying the hydride formation, which is characterized by signals of much lower amplitude. The temperature delay of the AE onset with respect to that of the hydride precipitation is explained by the fact that the microcrack nucleation and propagation, giving rise to the stress wave emission, occurs when the local stress field around the growing precipitates has reached a value such that elastic and plastic accommodation is no longer possible.

Figure 2 shows the amplitude distribution of the events recorded during the first cooling in all the tested samples at various H concentrations, for two different cooling rates, 1 and 4 K min^{-1}. The ordinates correspond to the number of events whose peak amplitude exceeds the level given by the corresponding abscissa. The amplitude is expressed in dB above 1 μV at the transducer output. The number of events has been reported after subtraction of the background consisting of counts recorded by each channel when a H-free sample was cooled under the same conditions. If reference to the first cooling is made [12], at a given cooling rate the total number of events generally increases with increasing H content and sample mass. High cooling rate may produce low AE activity or no activity at all; this is due to a finer dispersion of precipitates whose growth is diffusion controlled.

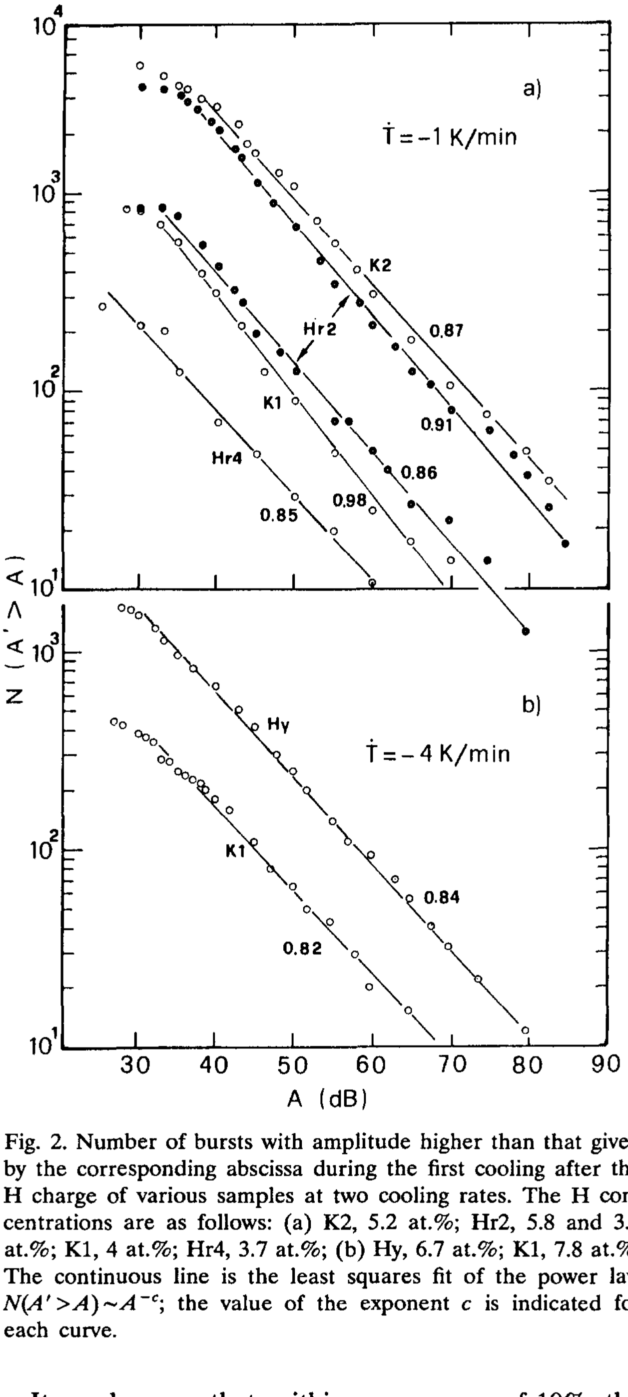

Fig. 2. Number of bursts with amplitude higher than that given by the corresponding abscissa during the first cooling after the H charge of various samples at two cooling rates. The H concentrations are as follows: (a) K2, 5.2 at.%; Hr2, 5.8 and 3.8 at.%; K1, 4 at.%; Hr4, 3.7 at.%; (b) Hy, 6.7 at.%; K1, 7.8 at.%. The continuous line is the least squares fit of the power law $N(A'>A)\sim A^{-c}$; the value of the exponent c is indicated for each curve.

It can be seen that, within an accuracy of 10%, the distribution functions of events for all samples at both cooling rates, follow the power law

$$N(A'>A)\sim A^{-c} \qquad c=0.9\pm0.1$$

over more than two orders of magnitude. This law is expected to be valid for signals with amplitudes higher than a threshold connected with some minimal event, *e.g.* the minimum energy for crack propagation. Here we limit its validity to signals higher than the experimental noise (30 dB corresponding to approximately 30 μV).

To compare our power law with those observed in SOC systems, we express the distribution function in terms of number of events with energy between E and $E+\mathrm{d}E$ remembering that A is proportional to $E^{1/2}$,

$$N(E)\mathrm{d}E\sim E^{-(1+c/2)}\mathrm{d}E \qquad c/2=0.45\pm0.05$$

The value of the exponent $c/2$ is surprisingly close to that of the energy distribution function of earthquakes estimated [3] from data of the Harvard earthquake catalog, $c/2=0.55$.

The occurrence of power-law distribution functions implies the absence of characteristic length and time scales, as in critical phenomena near a second-order phase transformation, in fractals, and in SOC systems. We exclude an explanation of our data in terms of a critical phenomenon because hydride precipitation is a first-order transformation, which does not imply fluctuations over all scales. Indeed, the shapes and dimensions of the precipitate particles are rather regular and depend on microstructure and cooling rate, because of the diffusion controlled nature of the process. Moreover, as each sample had a homogeneous grain texture, we can also exclude that the observed power-law may arise from a possible pre-existent fractal structure of grains and domains. Therefore, during the H precipitation the system goes into a SOC state, as far as the AE is concerned. The coincidence of the value of exponents of the power law presently observed and that of the Gutenberg–Ritcher law, referring to a phenomenon of remarkably different scale, should be considered as experimental evidence of what is suggested by numerical simulations (1, 2, 4, 7–9), *i.e.* fracture processes evolve into a SOC state.

In conclusion, the hydrogen precipitation in the Nb–H system produces a microseismic activity, revealed by recording the acoustic signals generated by the fracture processes occurring in the material. The analysis of the signals reveals that the distribution of their amplitudes follows a power law with an exponent very close to that of the Gutenberg–Ritcher law; this behaviour is indicative of a SOC state. The Nb–H system offers the possibility of observing SOC behaviour in a real physical system on scales of length and time much smaller than those of the dynamics of the earth's crust.

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