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Recovery stress and shape memory stability in Ni–Ti–Cu thin wires at high temperatures

The recovery stress evolution during thermal cycling (heating, isothermal holding, cooling) to temperatures significantly above the austenite finish temperature in Ni–Ti–Cu thin wires was evaluated in a dynamic mechanical analyser DMA TQ800. Ni–Ti–Cu wires were annealed at four different temperatures (350 °C, 450 °C, 550 °C and 650 °C). It was observed that the maximum recovery stress significantly decreases with increasing annealing temperature. It was also observed that the recovery stress during four isothermal holdings shows similar behaviour for all annealing temperatures, i.e. stress relaxation occurs after initial stress increase during the first isothermal holding and no stress relaxation is observed during isothermal holdings in subsequent thermal cycles. The maximum recovery stress after each thermal cycle decreases and shows a tendency to saturate with increasing number of thermal cycles. It is found that even when the recoverable strain is significantly decreased during thermal cycling, Ni–Ti–Cu wires are still able to generate very high recovery stresses.

Keywords: Shape memory alloys; Recovery stress; Ni–Ti–Cu; Stress relaxation

1. Introduction

From the application point of view NiTi and Ni–Ti–Cu are the most important shape memory alloys (SMAs) [1]. They can be prepared in a large variety of combinations (e.g. wires, rods, plates, springs and different complex structures from wires). SMAs have unique functional properties (e.g. one-way and two-way shape memory effect, recovery stress, actuator function). The unique functional properties of SMAs are basically nonlinear stress–strain–temperature hysteretic responses originating from a reversible martensitic transformation in the solid state. If an SMA is deformed in martensite, it will return to its original shape during subsequent heating. This process is known as one-way shape memory effect [2]. When the one-way shape memory effect is impeded, large recovery stresses are generated [2]. This is widely used in different applications e.g. couplings, fasteners, stone breakers etc. Since the recovery stress acts in the direction of the recoverable shape change, this can be used to perform work and the shape memory element acts as an actuator [1].

For different types of actuators it is desired to know the stability of recovery forces at a certain temperature and the ability to generate similar forces after certain time of usage. Another important question is how the recoverable strain will be affected by the isothermal holding at temperatures significantly higher than the austenite finish temperature (150 °C, 250 °C). This is for example crucial for smart hybrid composites, where prestrained SMA-wires are embedded into the matrix which is cured at high temperatures for quite a long time. In order to know the behaviour of the SMA wires after such a curing procedure it is desired to characterize the recovery stress forces at elevated temperatures and the influence of the curing procedure on the shape recovery characteristics of Ni–Ti–Cu wires.

Thermomechanical characteristics of Ni–Ti–Cu wires and the recovery stress behaviour were systematically studied in [3, 4]. Recovery stress evolution in Ni–Ti–Cu wires to 140 °C was studied and analysed by means of a phenomenological algorithm in [5]. Those articles deal with the evolution of recovery stresses to a maximum temperature of 140 °C. A limited number of articles study the influence of isothermal holding at temperatures significantly higher than the austenite finish (A_f) temperature on the functional behaviour of SMAs [6]. It is also known that the heat treatment significantly influences the functional properties of NiTi wire [7]. There are only few systematic studies on the effect of annealing temperature on recovery stress behaviour in NiTi wires [6, 8] but to the best of the authors knowledge there is no such a study for Ni–Ti–Cu wires.

In this article we study systematically the effect of annealing temperature on the recoverable strain and recovery stress evolution after constrained thermal cycling to temperatures significantly higher than A_f (150 °C) in Ni–Ti–Cu thin wires.

2. Experimental procedure

The experiments were performed on a commercial Ni-43 at.% Ti-7 at.% Cu wire provided by AMT (Belgium) (Table 1). Thermomechanical tests were carried out on a dynamic mechanical analyser (DMA), instrument TA Q800 using tension clamps. For all experiments INVAR clamps with low coefficient of thermal expansion were used.

The samples were loaded as follows (Fig. 1):
1. load to 4 % deformation at 25 °C,
2. unload to a force less than 0.02 N, which corresponds to the prestrain (residual strain) of 3.2 %,
3. fix the clamp and heat to 150 °C,
4. stay isothermal for 60 min,
5. cool down to 25 °C,
6. repeat (3), (4) and (5) for 3 times,
7. release the clamp,

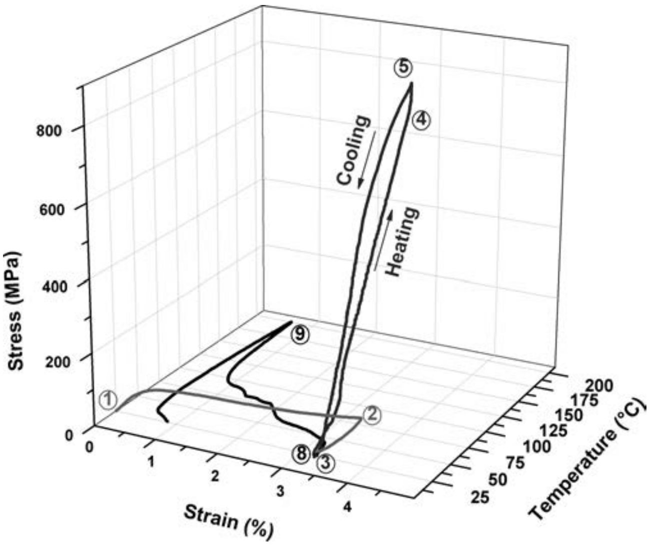


Fig. 1. Scheme of the experiment consisting of nine steps: (1) load to 4 % deformation at 25 °C, (2) unload to a force less than 0.02 N, which corresponds to the residual strain of 3.2 %, (3) fix the clamp and heat to 150 °C, (4) stay isothermal for 60 min, (5) Cool down to 25 °C, (6) Repeat (3), (4) and (5) for 3 times, (7) release the clamp, (8) heat to 200 °C under a constant small load of F = 0.02 N to measure the recoverable strain and (9) cool down to 25 °C under a constant small load of F = 0.02 N.

8. heat to 200 °C under a constant small load of F = 0.02 N to measure the recoverable strain and
9. cool down to 25 °C under a constant small load of F = 0.02 N.

Ni–Ti–Cu wires were annealed in an argon atmosphere at 350 °C, 450 °C, 550 °C and 650 °C for 10 min. After annealing the wires were left to cool to room temperature in an argon atmosphere for about 30 min. The differential scanning calorimetric (DSC) measurements from all tested wires were performed on a TA Q2000 instrument in the temperature range from –100 °C to 150 °C with heating and cooling rate of 10 K min^{-1}. The heating and cooling process was carried out in two cycles. The transformation temperatures obtained from the second cycle are given in Table 1.

Tensile tests at room temperature were performed on a DMA TA Q800 with a strain rate of 0.5 % min^{-1}. The gauge length of the tested wires was 8 mm.

3. Results and discussion

3.1. Transformation behaviour of Ni–Ti–Cu wires

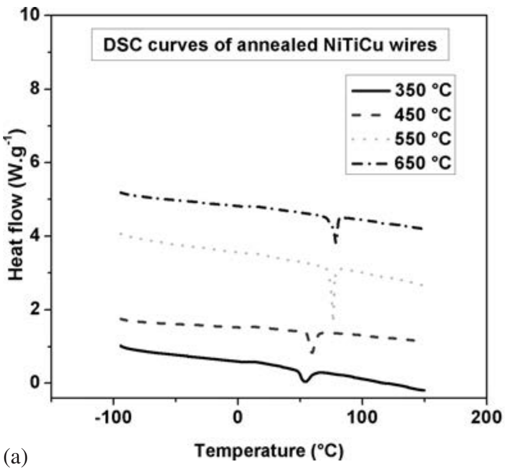
Figure 2 shows the DSC curves of Ni–Ti–Cu wires annealed at different temperatures. Transformation temperatures increase with increasing annealing temperature. The shape of the DSC peaks shows asymmetry. The asymmetry of the DSC peak increases with increasing annealing temperature. This can be related to the fractal growth of martensite.

3.2. Deformation behaviour of Ni–Ti–Cu wires

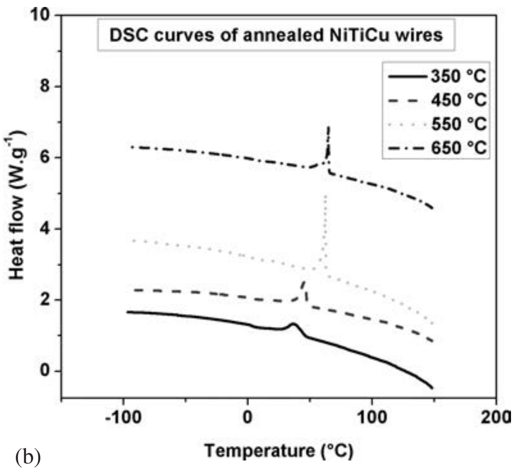
Figure 3 shows the stress–strain curves performed at room temperature on Ni–Ti–Cu wires annealed at different temperatures. The level of the stress plateau increases slightly with increasing annealing temperature and a quite abrupt increase is seen for the wire annealed at 650 °C. As the annealing temperature increases, the dislocation structure in

Table 1. Experimental materials.

Wire	Annealing temperature (°C)	Annealing time (min)	Diameter (mm)	Transformation temperatures (°C)			
				M_s	M_f	A_s	A_f
Ni–Ti–Cu	350	10	0.076	46.1	30.1	48.0	61.5
Ni–Ti–Cu	450	10	0.076	47.5	39.1	55.5	64.2
Ni–Ti–Cu	550	10	0.076	62.5	61.1	73.9	78.1
Ni–Ti–Cu	650	10	0.076	65.1	63.6	75.8	80.6



(a)



(b)

Fig. 2. DSC curves of Ni–Ti–Cu wires annealed at 350 °C, 450 °C and 650 °C for 10 min.

Table 2. Mechanical characteristics of Ni–Ti–Cu wire obtained from stress–strain curves at room temperature.

Annealing temperature and time	Yield strength (MPa)	Elongation (%)	Plateau stress (MPa)	Elasticity modulus (GPa)
350 °C–10 min	1118	10.8	87.3	18.7
450 °C–10 min	994	10.5	100.9	25.1
550 °C–10 min	716	51.6	107.9	22.9
650 °C–10 min	586	49.8	152.5	18.2

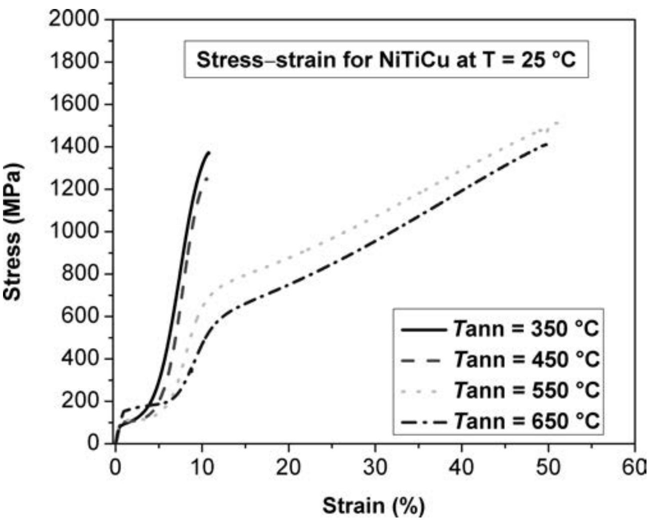


Fig. 3. Stress–strain curves of Ni–Ti–Cu wires at room temperature annealed at different temperatures (350 °C, 450 °C, 550 °C, 650 °C for 10 min).

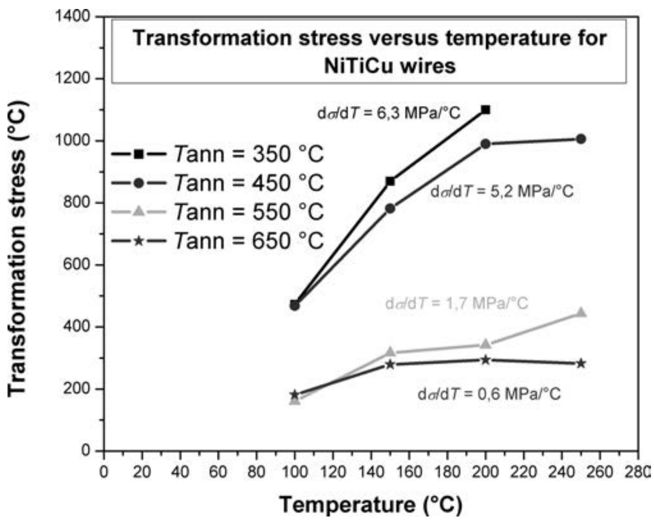


Fig. 4. Transformation stress as a function of testing temperature for Ni–Ti–Cu wires annealed at 350 °C, 450 °C, 550 °C, 650 °C for 10 min.

Ni–Ti–Cu wires rearranges and decreases, which leads to larger reorientation strains (Fig. 3). Mechanical properties extracted from stress–strain tests at room temperature on Ni–Ti–Cu wires are summarized in Table 2.

Figure 4 shows the transformation stress as a function of temperature for Ni–Ti–Cu wires annealed at different temperatures. The transformation stress increases linearly with temperature for the Ni–Ti–Cu wires annealed at 350 °C, 450 °C for 10 min. Wires annealed at 550 °C, 650 °C show a small increase in the transformation stresses with temperature. It is interesting to see that there is only a very small difference between the transformation stress at room temperature and at 100 °C for Ni–Ti–Cu wires annealed at 550 °C and 650 °C. The slope $d\sigma/dT$ changes from 5.95 MPa/°C for Ni–Ti–Cu wire annealed at 350 °C to $d\sigma/dT = 0.68$ MPa/°C for Ni–Ti–Cu wire annealed at 650 °C. These slopes were obtained as a linear fit of individual curves shown in Fig. 3. Usually for a material annealed at a certain temperature all factors in the Clausius–Clapeyron relationship, $d\sigma/dT = -\Delta H \cdot \rho/(\varepsilon_{tr} \cdot T_0)$, are well defined ($\Delta H$ is enthalpy of the transformation, ρ – density of Ni–Ti–Cu, and $T_0 = {}^1/_2(M_s + A_f)$ is the equilibrium temperature) except the transformation strain ε_{tr} [9]. The transformation strain can be either determined from the crystallography of the martensitic transformation or the actual strain that is measured in an individual experiment and is dependent on the stress state and the microstructural conditions of the matrix. The variation of ε_{tr} with stress or temperature leads to the deviation of the transformation stress–temperature dependence from linearity [10]. The $d\sigma/dT$ value is not a material constant for thermoelastic martensitic transformation, but it depends on the stress state, texture [11], thermomechanical history [2], annealing temperature [12] etc. Ni–Ti–Cu wires annealed at 550 °C and 650 °C show very small $d\sigma/dT$ values and there is only a slight increase in transformation stress with temperature (Fig. 4). The first annealing temperature is very close to the recrystallization temperature (600 °C) and the matrix contains a quite low density of rearranged dislocations [13]. The second annealing temperature (650 °C) is above the recrystallization temperature, where also grain growth occurs and the material has a very low resistance to the formation of dislocations. In this case the Ni–Ti–Cu wire will deform mostly by slip and as a result the irreversible deformation will occur with minor occurrence of stress–induced martensitic transformation. In this case the $d\sigma/dT$ values for Ni–Ti–Cu wires annealed at 550 °C and 650 °C mostly correspond to the yield tensile strength of Ni–Ti–Cu wire with minor contribution of transformation stress.

3.3. Recovery stress evolution in Ni–Ti–Cu wires after different annealing treatment

The recovery stress as a function of temperature for Ni–Ti–Cu wire thermally cycled to 150 °C for different annealing temperatures is shown in Fig. 5a–d. In total, four thermal cycles were applied to Ni–Ti–Cu wires, where one cycle consisted of heating to 150 °C, 60 min isothermal holding and cooling back to room temperature. The recovery stress evolution (stress–temperature) is non–linear

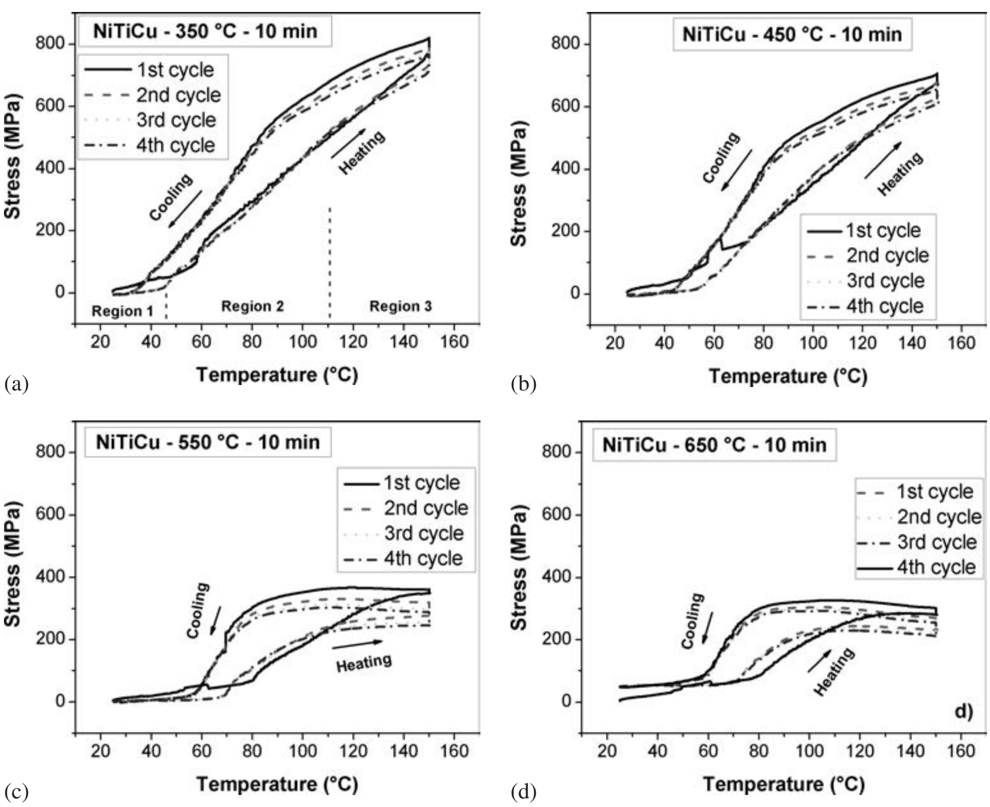


Fig. 5. Recovery stresses as a function of temperature for Ni–Ti–Cu wires annealed at: (a) 350 °C for 10 min, (b) 450 °C for 10 min, (c) 550 °C for 10 min, (d) 650 °C for 10 min.

and can be divided into three temperature regions with different slopes (Fig. 5a–d and Table 3). In the first temperature region the wire is below the A_s temperature and only a slight increase in the stress with temperature is observed, mainly on approaching the A_s temperature. The intersection between region 1 and region 2 is related to the A_s temperature of the individual wire. As the A_s temperature of Ni–Ti–Cu wire increases with increasing annealing temperature consequently the intersection between region 1 and 2 is shifted to higher temperatures. Region 2 corresponds to the abrupt increase in recovery stress and this is due to the reverse martensitic transformation to austenite. As the clamps are fixed and the Ni–Ti–Cu wire is trying to shrink back to the initial shape, large forces are generated during heating. The slope in this region is between 5–7 MPa/°C (Table 3). The slope in the third temperature region is lower than in region 2 for wires annealed at 350 °C and 450 °C (Fig. 5a and b), but a significant change in slope is observed for annealing temperatures 550 °C and 650 °C (Fig. 5c and d). Moreover, the slope of the recovery stress–temperature dependence becomes negative for the annealing temperature of 650 °C and a slight decrease in the recovery stress with increasing temperature is observed. This means that no reverse martensitic transformation occurs during the following continuous increase of the temperature.

Table 3. Slopes of regions 1, 2 and 3 from stress–temperature curves in Fig. 5.

Annealing temperature (°C)	Slope (MPa/°C)		
	1	2	3
350	0.8	7.4	5.2
450	0.6	7.7	4.2
550	0.2	5.3	0.8
650	0.1	6.0	−0.1

A maximum recovery stress of about 790 MPa was measured for Ni–Ti–Cu wire annealed at 350 °C, and it decreases with increasing annealing temperature up to about 280 MPa after annealing at 650 °C (Fig. 5a–d). As the initial prestrain of the Ni–Ti–Cu wires is the same then the parameter which mostly influences the maximum recovery stress value has to be related to the microstructural state of the wire before the thermomechanical test. This is in accordance with observations by Mallard et al. [7]. The measurement of recoverable strain after four thermal cycles to 150 °C confirms this statement (Fig. 8b). As the annealing temperature increases the measured recoverable strain decreases and is significantly lowered for Ni–Ti–Cu wires annealed at 550 °C and 650 °C. Thus it can be concluded that the plastic deformation is responsible for the recovery stress decrease in samples annealed at higher temperatures.

The temperature hysteresis is largest in the first thermal cycle for all annealing temperatures (Fig. 5). In the following cycles the hysteresis is smaller and does not change significantly. The beginning of the abrupt increase in the recovery stress which is associated to A_s temperature is shifted to higher temperatures. This might be caused by some martensite stabilization due to the former procedure.

3.4. Stress evolution during isothermal holding at 150 °C

The evolution of the recovery stresses during isothermal holding at 150 °C for different annealing temperatures is shown in Fig. 6. Recovery stress in all cases first increases and then stabilizes at approximately a constant stress value. The only difference is noticed in the first cycle for all an-

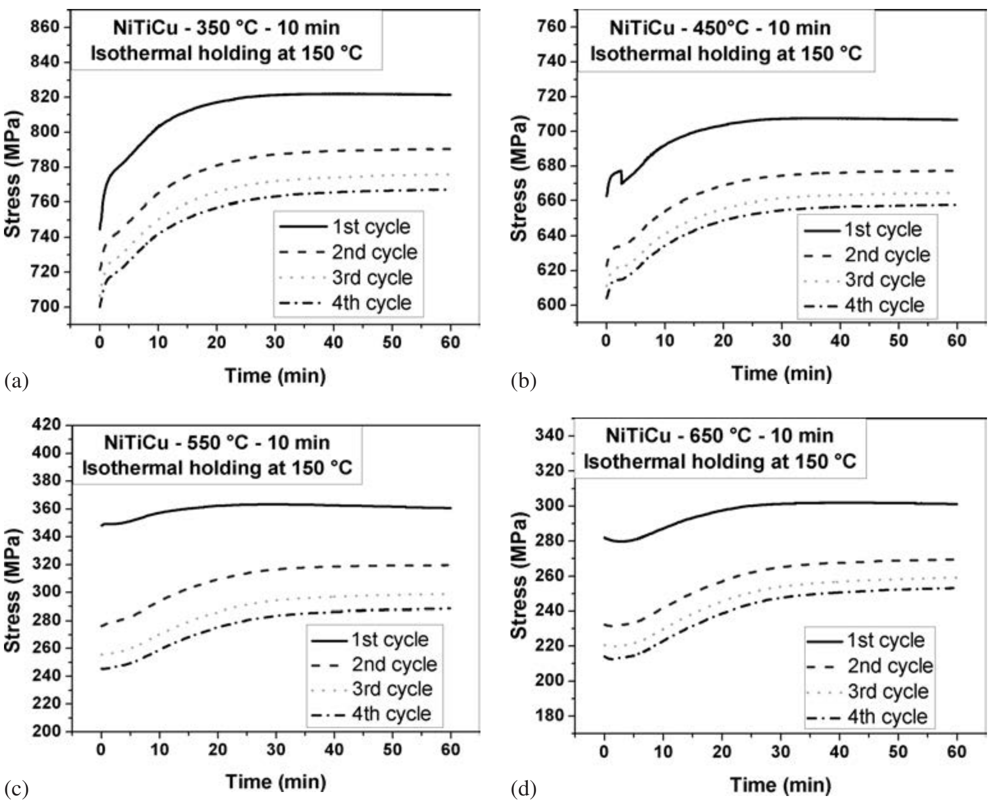


Fig. 6. Recovery stresses as a function of holding time at 150 °C for Ni–Ti–Cu wire annealed at: (a) 350 °C for 10 min, (b) 450 °C for 10 min, (c) 550 °C for 10 min, (d) 650 °C for 10 min.

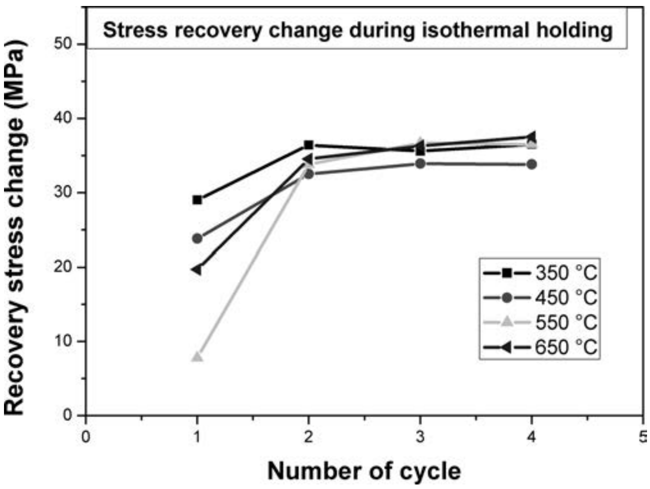


Fig. 7. Change of the recovery stress after one hour isothermal holding at 150 °C for Ni–Ti–Cu wire annealed at 350 °C, 450 °C, 550 °C and 650 °C.

nealing temperatures (Fig. 6a–d), where the recovery stress increases from the beginning and reaches a maximum value between 30 to 40 min depending on annealing temperature, and then slightly decreases.

Figure 7 shows the recovery stress change during 60 min isothermal holding for all thermal cycles and annealing temperatures. This change was calculated as the difference between the recovery stress at the time of about 6 min after reaching 150 °C and starting the isothermal holding and the recovery stress after 60 min. During the first 6 min the temperature was not stabilized at 150 °C. This is also the reason for the different evolution of recovery stresses during the first 6 min and afterwards (Fig. 6). Recovery stress change is different for the first thermal cycle for all annealing temperatures and is almost the same for the other thermal cycles (Fig. 7). The almost constant increase of recovery stress during isothermal holding at 150 °C for all other cycles could be related to the thermal dilatation of the clamps, which produce additional tensile stress in spite of the fact the clamps are made of INVAR. If it should be related to some isothermally activated recovery stress generation from Ni–Ti–Cu wire, then the wires annealed at 350 °C and 650 °C should give the largest and lowest increase of recovery stress, respectively. This was not observed. Thus the recovery stress evolution during the isothermal holding at 150 °C is stable after the first thermal cycle. The stress relaxation occurs during the first isothermal holding. Moreover, the maximum recovery stress decreases after each thermal cycle (Fig. 6a–d). For all annealing temperatures the largest loss of the recovery stress is after the first thermal cycle. In the following cycles it decreases and shows a tendency to saturate with increasing number of thermal cycles (Fig. 6). The decrease of the maximum recovery stress after each individual thermal cycle is a consequence of irreversible changes in the microstructure. This can be deduced from the measurement of recoverable strain after thermal cycling (Fig. 8a). Figure 8a shows the recoverable strain measured after the first thermal cycle and after four thermal cycles in Ni–Ti–Cu wire annealed at 350 °C. Initial 3.2 % prestrain decreased to about 2.44 % after the first thermal cycle. The difference in recoverable strain between the first and fourth cycle is about 0.05 %. This change is small but cannot be neglected when considering the decrease in the maximum recovery stress after each thermal cycle. The recovery stress value depends on the prestrain [4]. With decreasing recoverable strain in fact the prestrain decreases and consequently a lower value of recovery stress is obtained. It is interesting to

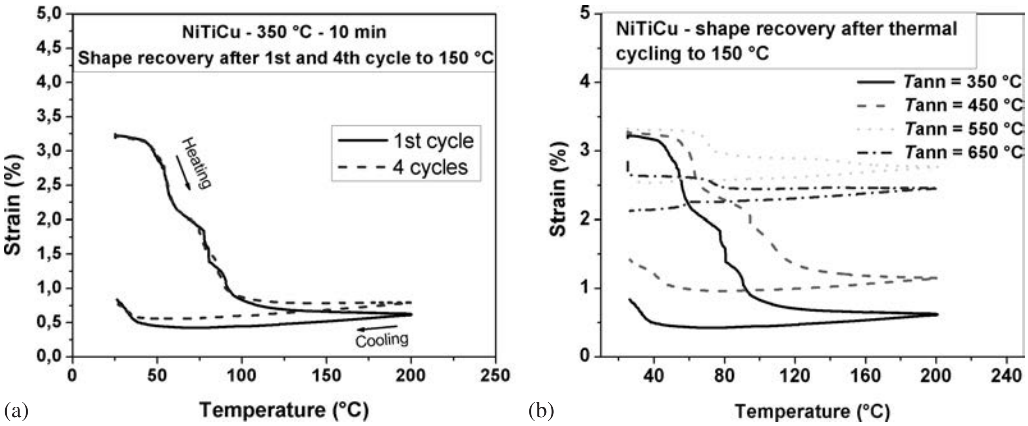


Fig. 8. (a) Recoverable strain after one and four thermal cycles to 150 °C. (b) Recoverable strain as a function of temperature for Ni–Ti–Cu wire annealed at 350 °C, 450 °C, 550 °C and 650 °C for 10 min.

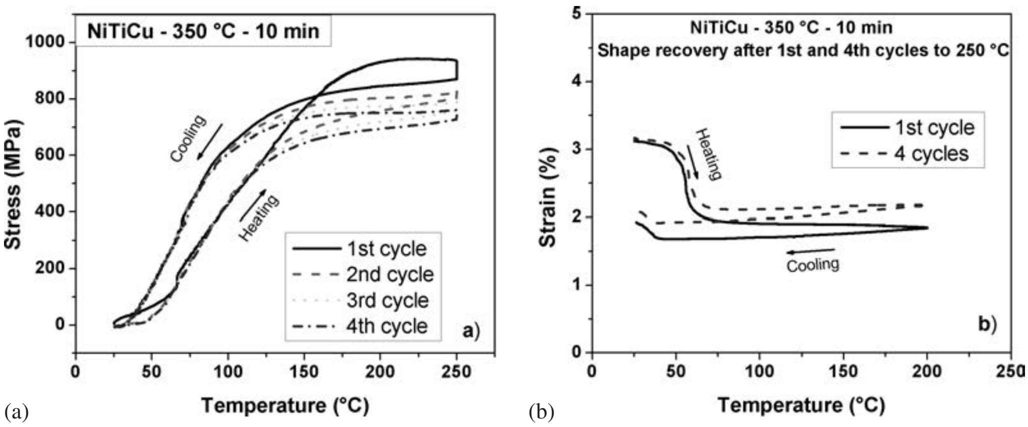


Fig. 9. (a) Recovery stress evolution during thermal cycling to 250 °C and (b) recoverable strain after one and four thermal cycles to 250 °C.

point out that the Ni–Ti–Cu wire thermally cycled to 250 °C with an initial prestrain of 3.2 % generates a recovery stress of 950 MPa (Fig. 9a). After the first thermal cycle the prestrain decreased to about 1.2 % and the generated recovery stress was 800 MPa. The loss of about 2 % of prestrain causes a recovery stress decrease of about 150 MPa (Fig. 9b). This clearly shows that there is no linear correlation between the prestrain and the recovery stress. It has to be related to the volume fraction of reversible martensitic variants which are present in the wire after the initial 4 % deformation (actual prestrain after unloading 3.2 %).

3.5. Shape recovery in Ni–Ti–Cu wires after thermal cycling to 150 °C

Shape recovery curves after four thermal cycles to 150 °C for Ni–Ti–Cu wire annealed at different temperatures are shown in Fig. 8b. The higher the annealing temperature the smaller is the recoverable strain obtained. Ni–Ti–Cu wires annealed at 550 °C and 650 °C show a small increase in recoverable strain during cooling, which starts around 56.3 °C and 59.8 °C, respectively (Fig. 8b).

With increasing annealing temperature the Ni–Ti–Cu wire starts to transform to austenite at higher temperatures (Fig. 8b), which is in agreement with transformation temperatures measured by DSC (Table 1). Shape recovery curves for wires annealed at 350 °C and 450 °C show two – stage behaviour, where the first part of the plateau transforms at the A_s temperature determined by DSC and the other part of the plateau is stabilized (higher A_s temperature). Wires annealed at 550 °C and 650 °C have only one small strain plateau (Fig. 8b). A two-way shape memory effect in the order of 0.36 % and 0.38 % is observed for wires annealed at 350 °C and 450 °C, respectively. Wires annealed at 550 °C and 650 °C show in contrast to the previous case a small increase of the recoverable strain (~0.13 %) during cooling (Fig. 8b).

The difference in the recoverable strain for different annealing temperatures (Fig. 8b) can be related to the different structural states of the wires. Wires annealed at 350 °C for 10 min have a very high dislocation density and no recovery or recrystallization occurred in those wires, as can be deduced from its stress–strain curve which shows a steep and short stress plateau together with the highest yield tensile strength (Fig. 3). These wires have a higher resistance to the introduction and formation of dislocations. With increasing annealing temperature the resistance to dislocation formation decreases and this might cause irreversible changes in the microstructure. This is clearly seen from the stress–strain curves from tensile tests (Fig. 3), where the yield tensile strength decreases with increasing annealing temperature. Another fact which also influences the movement of dislocations is the size of precipitates, which became incoherent with high annealing temperatures and loses the impeding effect on dislocation movement. The presence of residual martensite and strong dislocation activity, observed by electron microscopy was responsible for the loss of shape memory under thermal cycling under stress [14]. Residual martensite which is pinned by dislocations, limits the volume of austenite domains available for transformation. The recrystallized wire is very sensitive to the introduction and movement of dislocations and as a consequence after thermal cycling to 150 °C the lowest recoverable strain is observed (Fig. 8b).

4. Conclusions

Recovery stress measurements on Ni–Ti–Cu wires annealed at different temperatures (350 °C, 450 °C, 550 °C and 650 °C for 10 min) were performed during thermal cycling to 150 °C, together with the isothermal holding at this temperature. From the obtained results the following conclusions can be proposed:

- In order to obtain a high recovery stress and a large recoverable strain the Ni–Ti–Cu wire tested in this work should be annealed at 350 °C for 10 min.
- Recovery stress generated by Ni–Ti–Cu wire decreases with increasing annealing temperature. This is a consequence of the lower yield tensile strength in Ni–Ti–Cu wires annealed at higher temperatures.
- The recoverable strain after thermal cycling to high temperature decreases with increasing annealing temperature. This is caused by the lower resistance to dislocation formation with increasing annealing temperature.
- For Ni–Ti–Cu wires thermally cycled to 250 °C, a significant loss of recoverable strain is obtained, but the wires are still able to generate high recovery stresses.
- Due to significant loss of recoverable strain Ni–Ti–Cu wires annealed at 550 °C and 650 °C cannot be used as actuator wires but they are still able to generate quite large recovery stress (~300 MPa), which allows them to be used in certain smart hybrid composite applications.
- The ability of Ni–Ti–Cu wires annealed at 350 °C for 10 min to generate recovery stress is not significantly damaged by repeatedly exposing the wire to temperatures as high as 150 °C or even 250 °C for long time at high stress.

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