JMEPEG (2009) 18:818–823
DOI: 10.1007/s11665-009-9400-3

1059-9495/$19.00

Superelasticity of NiTi Ring-Shaped Springs Induced by Aging for Cranioplasty Applications

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(Submitted September 11, 2008; in revised form January 16, 2009)

This paper concerns the application of titanium-nickel rings in modeling the cranium. After being fixed to the osseous margins, the ring's expansion at the same time broadens and shortens the cranium vault. The rings formed from a straight superelastic wire, flattened to an ellipse, do not show the presence of a typical force plateau but rather a pseudoelastic loop during loading-unloading in the relationship between the force and the deflection. Based on the idea that superelasticity in more complex shape-springs may be induced by the precipitation hardening process, the further studies were carried out on alloys with higher nickel contents (51.06 at.% Ni). The rings that had been formed were welded and aged at an optimal temperature and time. The improved superelastic behavior during compression and unloading the rings was obtained by introducing small deformation by drawing the quenched wires before forming the rings and aging. Very positive clinical reshaping by long-term distraction with the superelastic ring-shaped springs was achieved in young children under one year and a less spectacular effect was observed in the group of older children.

Keywords biomaterials, heat treating, intermetallics

1. Introduction

An alternative method for cranial modeling by surgery is the distraction of the cranium vault bone with the aid of steel springs, proposed by Lauritzen et al. (Ref 1). High efficacy of the spring-mediated cranioplasty was confirmed by Guimares-Ferreira et al. (Ref 2). As it is well-known steel springs lose force during their expansion. This explains why the application of superelastic NiTi springs with their force plateau for the long-term distraction of the mandibular in pigs has been confirmed (Ref 3). Based on these results, further research into superelastic springs and rings for cranioplasty has been undertaken (Ref 4). The basic aim of bone elongation was to achieve a continuous and constant force in a wide range of deformations using superelastic springs and rings. The very plateau stress-strain curves exhibited by superelastic NiTi alloys can be used advantageously to sustain constant forces or minimize their variations over a large range of deformations. It concerns the loading and unloading plateau connected with stress hysteresis.

This article is an invited paper selected from presentations at Shape Memory and Superelastic Technologies 2008, held September 21-25, 2008, in Stresa, Italy, and has been expanded from the original presentation.

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This study presents the process of forming the superelastic rings from NiTi wire, which have been used for experimental elongation in the clinical research into modeling the cranial vault in children suffering from craniostosis.

2. Materials and Methods

The studies were carried out on three commercially available NiTi alloys in the form of straight wires. Their chemical composition and characteristics are presented in Table 1. The chemical analysis was carried out using the energy dispersive spectroscopy (EDS) method by the scanning electron microscope (SEM) JSM-6480. At the first stage of the studies, superelastic wires of 0.8, 1.0, and 1.2 mm diameter provided by SMATEC were used.

The rings were formed by bending a straight wire to a round form and then locating it in the holder which allows laser beam welding. To ensure a safe connection, both ends of the wire were overlap welded. The welding was carried out in a pure argon atmosphere using a Nd-YAG Dental Laser Welder type 2002 S with the power of 30 W. The characteristic temperatures and the course of the martensitic transformation were studied with the use of a differential scanning calorimetry (DSC) method and by the Perkin-Elmer DSC-7 instrument. The courses were registered at the cooling and heating speed of 10° per minute.

The aim of the second stage of the studies was to obtain superelastic behavior of rings during their deformation by flattening them to an ellipse. The alloys (nos. 2 and 3) marked NT-10 are destined to form complex shapes and induce superelasticity by aging. To obtain the best superelastic behavior, the optimization of the aging parameters was necessary. The aging was carried out in the temperature range 300-500 °C for different time holding. Before their implantation, the rings had undergone the process of steam passivity in an autoclave at 134 °C for 30 min.

Table 1 Chemical composition and characteristics of NiTi alloys used in the studies

Alloy	Ni, at.%	Ti, at.%	Al, at.%	Si, at.%	M_s, °C	A_f, °C	State of the wires	Delivering company
1	50.8 ± 0.27	49.0 ± 0.40	···	···	n.m. −15	n.m. +11	As received after quenching 800 °C/1 h	SMATEC
2	51.02 ± 0.36	48.71 ± 0.36	0.12	0.14	−80	−30	As received after quenching	AMT (NT-10)
					−65	−35	800 °C/1 h	
					−30	+15	After aging 500 °C/15 min	
3	51.06	48.9	···	···	n.m.	n.m.	As received after quenching 800 °C/1 h	SMATEC
					−32	+11	After aging 500 °C/30 min	

n.m., Not measurable

The structure of the obtained TiO_2 layer, about 4 nm thick, was amorphous, which was confirmed by a high-resolution electron microscopy (HREM) examination. The precipitation process was also controlled by a transmission electron microscopy (TEM) method in a JEM 3010.

The tensile tests of these wires were done on the Instron machine. The three-point bending tests in loading-unloading experiments were carried out on a specially computerized device equipped with a Hottinger force converter, a Pelton linear variable differential transformer, and a digital temperature indicator.

3. Results and Discussion

The idea of this work was to use rings instead of springs for cranioplasty, which by flattening to an ellipse would be connected to the skull and during long-term expanding would result in the elongation of the cranial bones perpendicularly to the sagittal direction and its contraction in a frontal plane, causing the craniofacial reshaping.

A flattened ring in the form of an ellipse is implanted onto the skull and the forces exerted on it are schematically shown in Fig. 1.

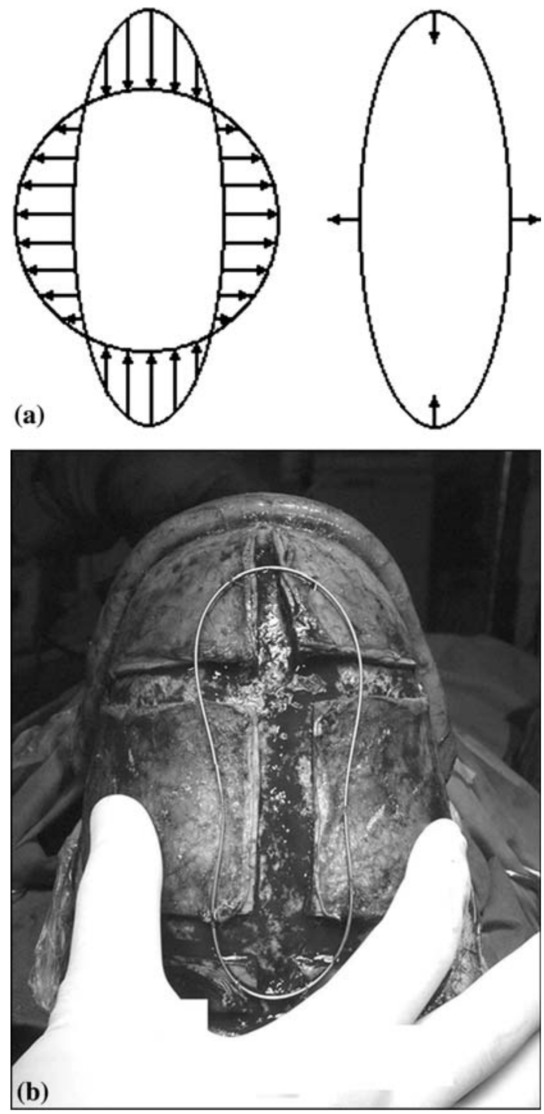

Fig. 1 Sketch of the forces exerted on the skull in a flattened ring (a) and its fixation on the cranium vault (b)

The problem was to achieve superelastic behavior of the rings, which means the presence of the broad force plateau on the force-deformation curve during force releasing. The rings formed from the superelastic wire of alloy 1 were welded with the use of a laser beam.

Their elastic characteristic during loading-unloading, which was obtained when the ring was flattened to an ellipse, does not show the presence of a typical force plateau but rather a linear relationship between the force and the deflection with a slope to the deflection axis as shown in Fig. 2.

In order to obtain typical superelastic properties of the ring ⇔ ellipse deflection, another method of superelastic induction for the rings was worked out. Based on the idea that superelasticity in more complex-shaped springs may be induced by the precipitating process, two alloys with higher nickel contents (alloys 2 and 3) were chosen.

The sequence of the martensitic transformation for the alloys 2 and 3 are shown on the DSC curves in Fig. 3.

The alloy 2 in as quenched state shows the direct B2 ⇔ B19′ and the reverse transformation, whereas after aging the transformation occurs through the R-phase, B2 ⇔ R ⇔ B19′. The preliminary slight deformation through drawing the wire of the parent B2-phase ($\varepsilon \sim 10\%$) and aging alloy 3 at 500 °C shows the same sequence through the R-phase. Both wires of alloys 2 and 3 after aging at 500 °C are at room temperature in the parent phase, as can be seen on the heating DSC curves in Fig. 3.

Aging of a straight wire of alloy 2 in the quenched state has shown the possibility of improving the elastic behavior of this wire, but still the amount of permanent strain is 2.5%. Considerable improvement of superelastic behavior has been

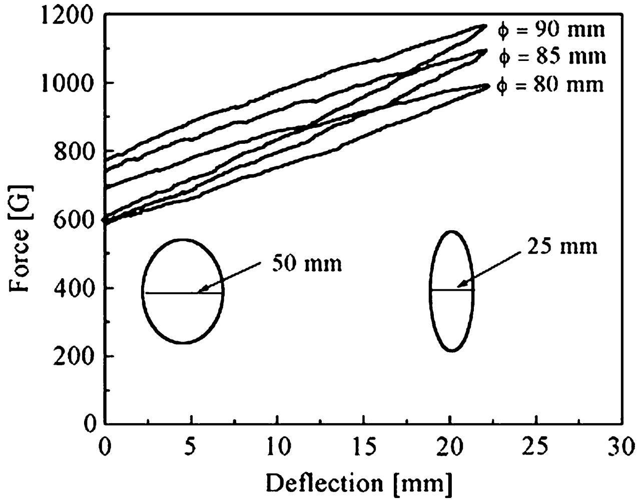

Fig. 2 Force vs. displacement for flattened rings formed from the straight superelastic wire (alloy no 1)

achieved by aging of the wires preliminary deformed by drawing. The three-point bending test for this state of alloy 3 after aging at 450 °C for 15 min shown in Fig. 4 exhibits a typical superelastic curve with a very low (about 0.1%) permanent strain.

The tree cycles of loading-unloading curves shown on this plot confirm the stability of the superelastic deformation.

The favorable affect of small deformation by drawing wires of alloy 3 on induced superelasticity is also presented in Fig. 5, which shows the effect of temperature aging on the hardening force-deflection curves by a three-point bending test. The hardening curves of the wires deformed by drawing show a more regular plateau and hysteresis force. As it can be seen, the higher temperature aging increases the force needed to start stress-induced martensite transformation, but the curve for the wire aged at 500 °C shows also the increase of the permanent strain up to 2.5%.

The rings formed were aged at the optimal temperature and time (500 °C/15′). The aging of rings results in hardening the parent phase by the precipitation of the coherent Ni_4Ti_3 phase. As a consequence, during deformation the parent phase exhibits superelastic behavior with a clear force plateau shown in Fig. 6 for rings of different diameters. As it can be seen, the level of force plateau lowers when the ring diameter increases.

The rings with the smallest diameter (D = 30 mm) show the highest force of the plateau, but simultaneously a large residual strain. This residual strain is caused by the dislocations which appear at the interface between the parent and the martensite phases due to the stress-induced martensite.

The smaller the ring's diameter the larger the pre-deformation during its formation and the higher the stress induced. It means that a relatively low deformation before the aging is favorable for the precipitation process. Identification of the precipitated particles was done using the TEM method. Figure 7 shows the disc shape coherent precipitates and their diffraction pattern.

The particles cause the matrix lattice distortions which achieved the maximum values perpendicular to the axis of the particles. In case of coherent Ni_4Ti_3 particles, this maximum appears in the < 111 > direction of the matrix and are equal to

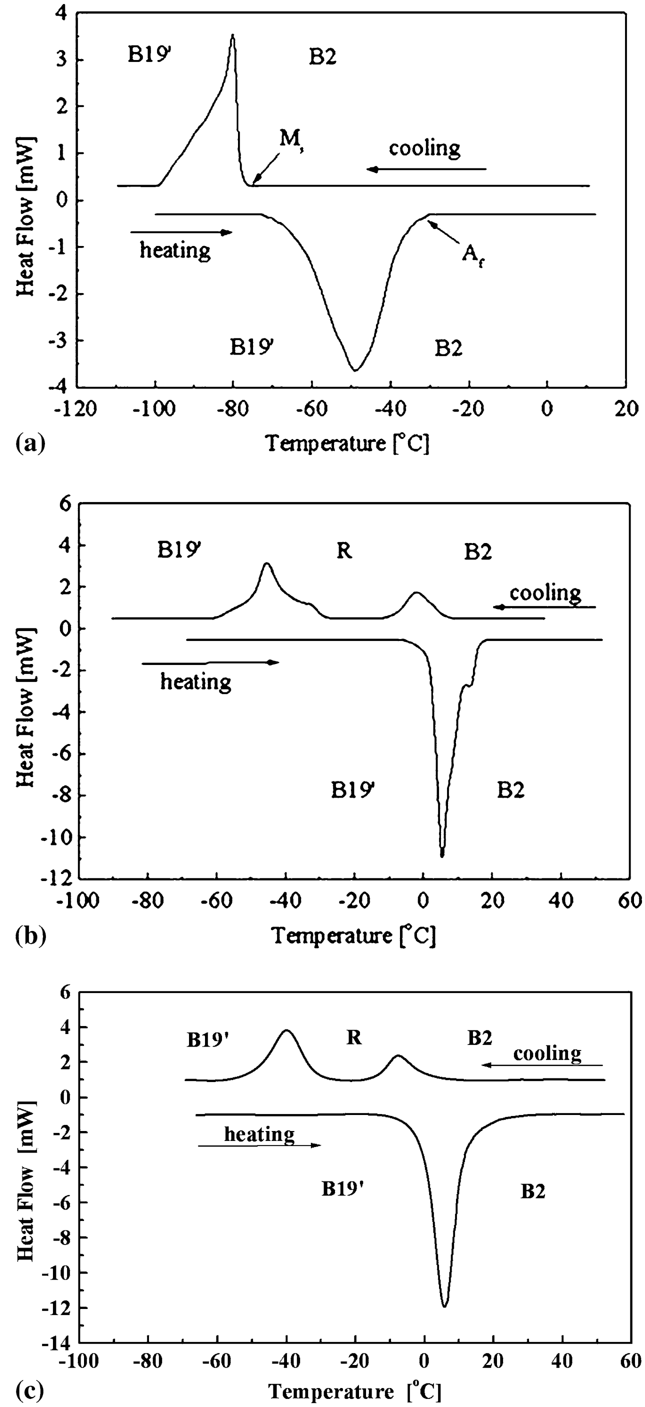

Fig. 3 DSC cooling and heating curves for alloy no 2 and 3 (a) for the delivery state of alloy 2, (b) after aging at 500 °C/15′, and (c) alloy no 3 deformed and aged at 500 °C/15′

the lattice misfit of both phases $\delta = (d_{111p} - d_{111B2})/d_{111B2}$ = 2.9%, they act as a tensile tension in this direction. Perpendicular to the < 111 > direction: [110] and [112] directions, the lattice misfit is 1.4% and acts as a compression stress. As a result, the stress distribution around the particle is inhomogeneous but exhibits symmetry in respect to < 111 > direction (Ref 5). The field stress distribution around the particles determined by Tirry and Schryvers has shown its lowering with a distance increase of the precipitates.

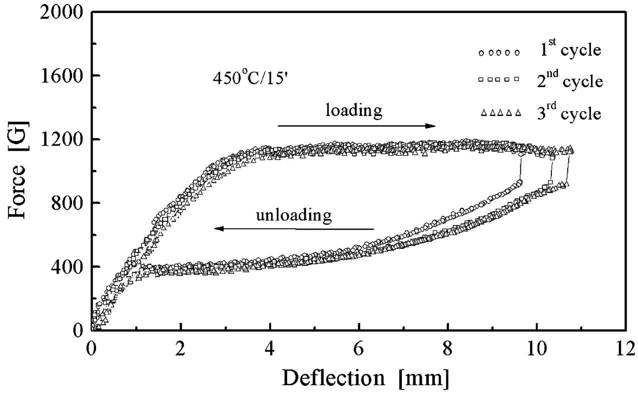

Fig. 4 Superelastic hysteresis of the three-point bending test for pre-strained wires of alloy 3 aged at 450 °C

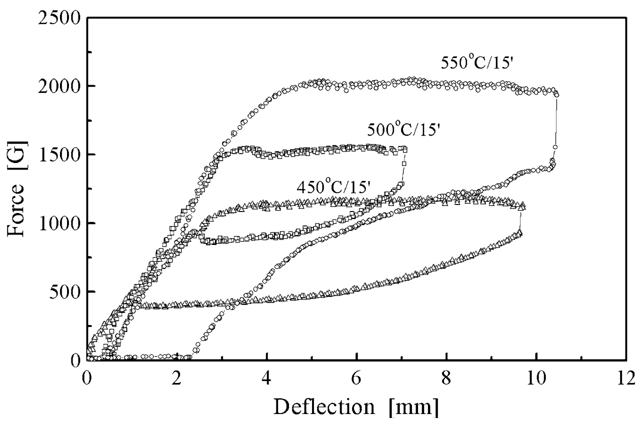

Fig. 5 Effect of temperature aging of alloy 3 on the superelastic force-deflection curves

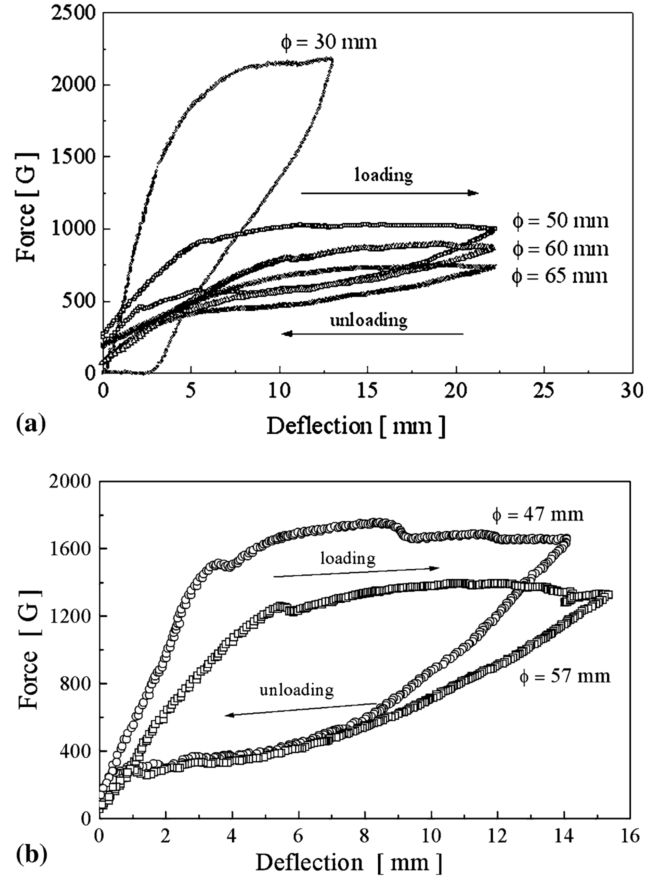

Fig. 6 Superelastic behavior of ring-shaped springs while deflecting to an elliptic shape and their reversion to the previous ring shape for (a) alloy 2 and (b) alloy 3

In accordance with the results obtained by Chumlyakov et al. (Ref 6), the superelastic effect is induced only under the following conditions: Ni_4Ti_3 particles are coherent with the B2 matrix and have the size of 50-100 nm. Coherent particles of Ni_4Ti_3 are the source of internal stresses, sites of the preferable martensite nucleation, and therefore assist lowering of the stress plateau. On the other hand, the particles do not undergo martensitic transformation and after the martensite is induced they become the source of the reverse stress fields and assist to store the elastic energy (Ref 7).

Clinical craniofacial reshaping by long-term distraction with the use of superelastic rings was carried out in the clinic of Plastic Surgery in Polanica (Ref 8). After the sparing excision of the cranium vault sutures in the shape of the letter "H," the compressed ring is given an oval shape in the sagittal axis and in this form it is fixed to the osseous margins. In four older children, the base parietal, frontal, and occipital bones were additionally resected approximately 2 cm. Ring expansion led to the expansion of the cranium vault bones perpendicularly in the sagittal direction, and to their compression in the anterior-posterior plane, which resulted in a desired cranium broadening and shortening. In these procedures, the rings with 90-100 mm diameters were used. The average operation time in case of the excision of the cranial sutures and rings placement was up to 100 min. The average hospitalization time was 7 days. After a 3-month period of distraction, the children were qualified for the removal of the rings. The ring was cut and removed through a small 0.5-cm incision on the welded site. The positive results of the operations carried out with the use of the superelastic rings can be seen in Fig. 8.

No early or late complications were observed during the implantation of the rings as well as during their removal. The change of cranium shape was approved as the criterion for the treatment results, since the objective measurement of the cranium capacity is still not very precise and the CT-3D tomography only in order to confirm clinical improvement appeared to be unadvisable. Better results were found in the cranium broadening than shortening, and mainly observed in younger patients under 1 year and less spectacular results in the group of older children. In every case, the shape of the head was subject to noticeable improvement. Classic procedures require extensive preparation and total reshaping of the calvaria. Large blood loss, long-term dissection, and the risk of infection or bone resorption pose a real threat that might be avoided in our method.

4. Conclusions

- The method of forming superelastic rings was worked out using the precipitation hardening of the rings previously

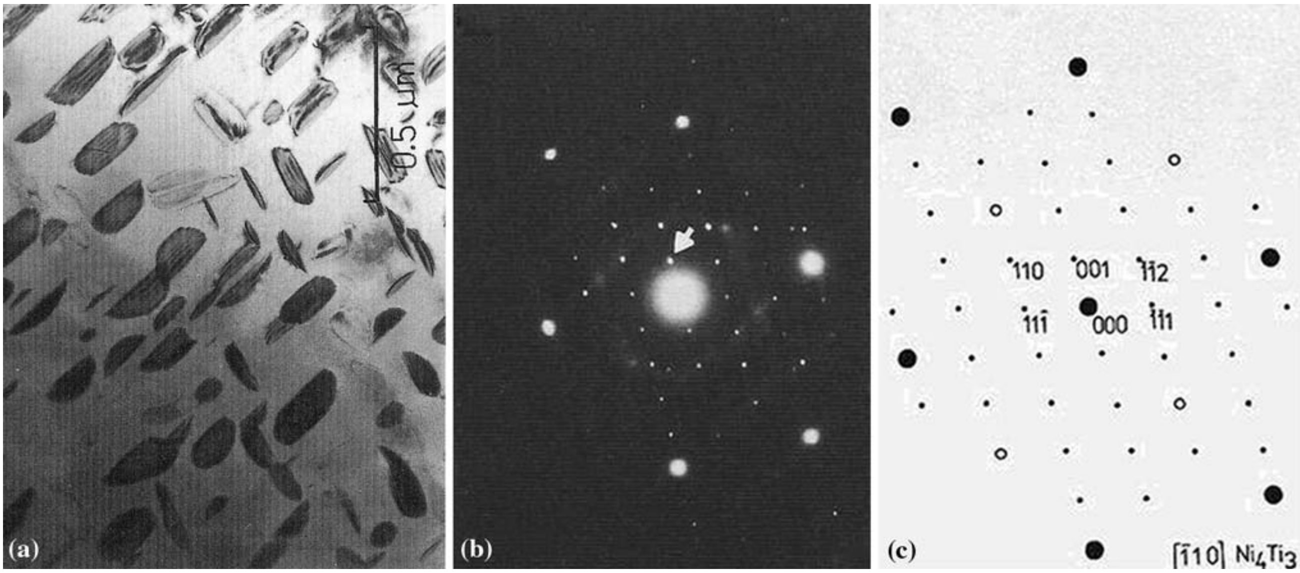

Fig. 7 Particles of Ni_4Ti_3 phase in the alloy aged at 500 °C for 1 h (a) bright-field image, (b) diffraction pattern from particle and (c) indexed electron diffraction pattern

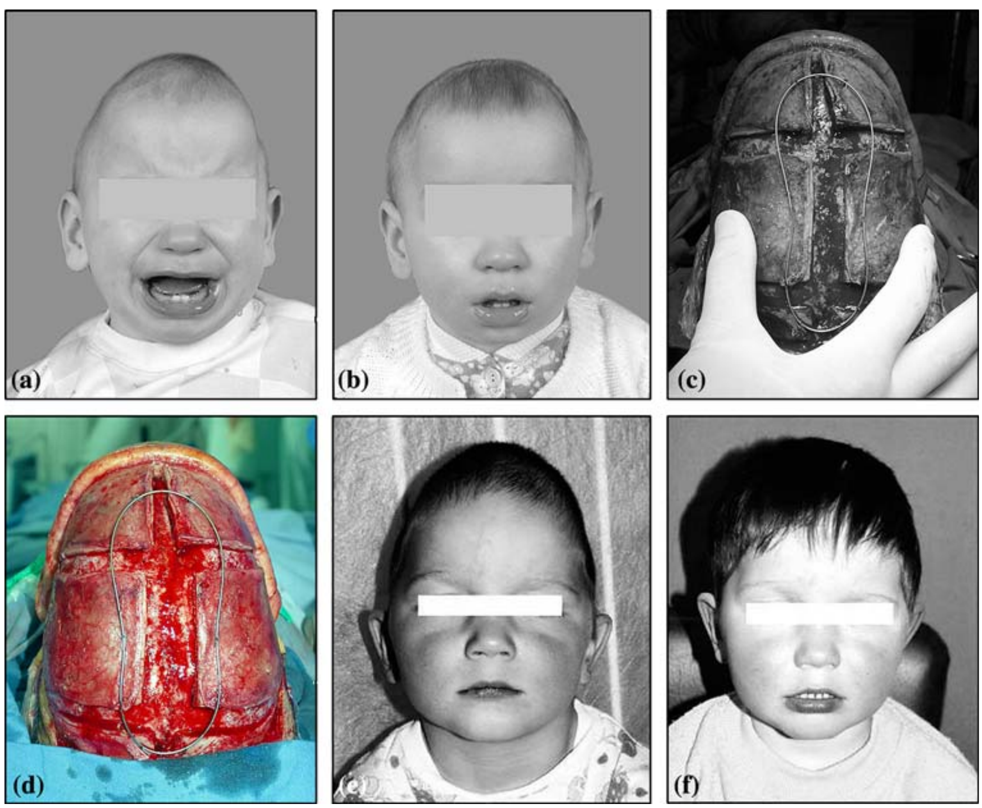

Fig. 8 Treatment of scaphocephaly with the use of superelastic ring-shaped springs

formed from wires of the NiTi alloy containing 51 at.% Ni.

- Superelastic rings deformed in the bending act with a constant force in the desired displacement range.
- Wires deformed by drawing and aged in the form of a ring improve the superelastic behavior of a ring-shaped spring.
- Clinical research confirmed the possibility of applying superelastic rings in cranioplasty.

Acknowledgment

Grateful acknowledgment is made for financial support by the Polish Ministry of Education and Science within the frame of the project PBR-4/rjp3/06.

References

1. C. Lauritzen, Y. Sugawara, O. Kocabalkan, and R. Olsson, Spring Mediated Dynamic Craniofacial Reshaping, *Scand. J. Plast. Reconstr. Hand Surg.*, 1998, **32**, p 331–338
2. J. Guimaraes-Ferreira, F. Gewalli, L. David, R. Olsson, H. Friede, and C.G.K. Lauritzen, Spring-Mediated Cranioplasty Compared with the Modified Pi-Plasty For Sagittal Synostosis, *Scand. J. Plast. Reconstr. Hand. Surg.*, 2003, **37**, p 208–215
3. Z.H. Lekston, J.T. Drugacz, and H. Morawiec, Application of Superelastic NiTi Wires for Mandibular Distraction, *Mat. Sci. Eng.*, 2004, **A378**, p 537–541
4. H.Z. Morawiec, Z.H. Lekston, K.F. Kobus, M.C. Węgrzyn, and J.T. Drugacz, Superelastic NiTi Springs for Corrective Skull Operations in Children with Craniosynostosis, *J. Mater. Sci: Mater. Med.*, 2007, **18**, p 1791–1798
5. W. Tirry and D. Schryvers, Quantitative Determination of Strain Fields Around Ni_4Ti_3 Precipitates in NiTi, *Acta Mater.*, 2005, **53**, p 1041–1049
6. Yu.I. Chumlyakov, I.V. Kireeva, E.I. Litviniva, and A.G. Lisyuk, Superelasticity During the Elastic Twinning, Slip and Martensitic Transformation, *Proceedings of the Second International Conference on Shape Memory and Superelastic Technologies*, 1st ed., A. Pelton, D. Hodgson, S. Russel, and T. Duerig, Eds., March 2–6, 1997 (Pacific Grove), International Organization on SMST, 1997, p 29–34
7. Yu.I. Chumlyakov, I.V. Kireeva, A.G. Lysyuk, and Y.L. Zuev, Superelasticity and Shape-Memory Effects in Ti-Ni Single Crystal, *Proceedings of the Second International Conference on Shape Memory and Superelastic Technologies*, 1st ed., A. Pelton, D. Hodgson, S. Russel, and T. Duerig, Eds., March 2–6, 1997 (Pacific Grove), International Organization on SMST, 1997, p 7-12
8. K.F. Kobus, M.C. Węgrzyn, Z.H. Lekston, H.Z. Morawiec, and J.T. Drugacz, Modeling of Scaphocephaly Using Superelastic Titanium-Nickel Rings: A Preliminary Report, *J. Craniofac. Surg.*, 2007, **18**, p 504–510