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MICROSTRUCTURE EVOLUTION AND PHASE TRANSFORMATIONS IN MICROALLOYED ARMOX 500T STEEL DURING A DILATATION PROCESS

RAZVOJ MIKROSTRUKTURE IN FAZNE TRANSFORMACIJE MIKROLEGIRANEGA JEKLA VRSTE ARMOX 500T MED PROCESOM ŠIRJENJA

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With this work we successfully developed two modified Armox 500T alloys using microalloying with different amounts of niobium (Nb) and boron (B) to obtain a finer grain size, which in return enhances the remnant properties. Furthermore, different heat-treatment cycles were designed and performed using a quenching dilatometer to study the combined effect of thermal cycling and microalloying with Nb and B on the microstructure and transformation temperatures including the start and finish temperature of the austenite transformation (Ac1, Ac3) and the martensite start and finish temperature (Ms, Mf) of the investigated alloys. Dilatometry results show that increasing the content of Nb from 0.07 *w*/% to 0.13 *w*/% and B from 0.0035 to 0.0046 *w*/% increases the temperature range between Ac1 and Ac3 by 55 °C, indicating a broader range for changing heat-treatment temperatures. In addition, the Ms temperature is reduced by 13 °C due to austenite refinement caused by the microalloying of Nb and B. The effect of the annealing temperature at a constant heating rate showed a significant impact on the austenite grain size and hardness. Furthermore, the kinetics of phase transformations were theoretically studied using Thermo-Calc, and the numerical predictions were confirmed experimentally with dilatometry results. Metallography investigations using a scanning electron microscope (SEM) and an optical microscope (OM) were conducted to evaluate the microstructure evolution of the developed alloys. Hardness tests were performed to evaluate the effect of the grain refinement of martensite lathes caused by microalloying with Nb, B, and heat-treatment thermal cycling. It is found that the hardness of the modified Armox alloys in this research was improved by 14 % in comparison with the conventional Armox 500T.

Keywords: Armox 500T steel, niobium microalloying, heat treatment, grain refinement

V članku avtorji opisujejo raziskavo s katero so uspešno razvili dve modificirani zlitini vrste Armox 500T. Pri tem so uporabili postopek mikrolegiranja različnih vsebnosti (Nb) in bora (B). Nadalje so oblikovali in izvedli različne postopke toplotne obdelave ter z uporabo kalilnega dilatometra študirali kombinirani učinek toplotnega cikla in mikrolegiranja z Nb in B na mikrostrukturo in temperature transformacij. Tako so določili začetek in konec temperature austenitne transformacije (Ac1, Ac3), kot tudi začetno in končno temperaturo tvorbe martenzita (Ms, Mf) preiskovanih zlitin oziroma jekel. Rezultati dilatometrije so pokazali, da povečanje vsebnosti Nb z 0,07 *w*/% na 0,13 *w*/% in B z 0,0035 *w*/% na 0,0046 *w*/% poveča temperaturno območje med Ac1 in Ac3 za 55 °C, kar pomeni širše območje temperatur toplotne obdelave. Dodatno se je temperatura Ms zmanjšala za 13 °C zaradi mikrolegiranja z Nb in B ter možnega udrobljenja austenita. Močno je na velikost austenitnih zrn in trdoto vplivala temperatura popuščanja pri konstantni hitrosti segrevanja. Nadalje so avtorji kinetiko faznih transformacij teoretično študirali s pomočjo programskega orodja Thermo-Calc in numerične napovedi eksperimentalno potrdili z rezultati dilatometrijskih preizkusov. Metalografske raziskave in ovrednotenje mikrostrukture razvitih zlitin so avtorji izvedli s pomočjo svetlobnega (LM) in vrstičnega elektronskega mikroskopa (SEM). Vpliv udrobljenja martenzitnih lamel zaradi mikrolegiranja z Nb in B in toplotne obdelave so določili z meritvami trdote. Ugotovili so, da se je trdota modificiranih zlitin vrste Armox izboljšala (povišala) za 14 % v primerjavi s konvencionalnim Armox 500T.

Ključne besede: jeklo vrste Armox 500T, mikrolegiranje z niobijem in borom, toplotna obdelava, udrobljenje kristalnih zrn.

1 INTRODUCTION

Armox 500T is high-strength armour steel whose mechanical properties can be controlled with chemical composition, heat treatment and deformation processes. It is used for protection as a shielding material in automotive vehicles and buildings. Moreover, this martensitic ARMOX steel is characterized by high performance and remarkable properties such as high tensile stress ranging from 1450 to 1750 MPa and nominal hardness of 500 HBW coupled with exceptional toughness values of 32 J at –40 °C.[1–3]

One of the fundamental characteristics influencing the steel's mechanical properties and increasing its hardenability during heat treatment is the austenite grain size. The high strength and toughness are hard to be obtained together except in grain refinement.[4–8] The austenite refinement of steels can be achieved through the formation of carbides and nitride precipitates that act as obstacles inhibiting the grain growth.[6] It has been re-

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Table 1: Chemical compositions in *w*/% of the modified Armox alloys

Alloy No.	C	Si	Mn	Cr	Mo	Ni	Nb	B	P	S	Fe
Armox-1	0.25	0.22	0.87	1.22	0.7	1.69	0.07	0.0035	0.013	0.008	Bal.
Armox-2	0.28	0.18	0.70	1.21	0.64	1.64	0.13	0.0046	0.013	0.008	Bal.

ported that percentages of microalloying with B and Nb should not exceed 0.15 % due to solubility and formation of secondary phases.[6,7] Gao et al.[8] found that Nb causes a grain refinement with subsequent heat treatments; fine spherical precipitates of niobium carbides are formed, which have a strengthening effect that causes an enhancement in the mechanical properties such as hardness, strength and impact toughness. These properties mainly exist in steels containing tempered martensite.[9] However, dynamic loading failure is due to retained austenite and coarse martensite, which decrease the mechanical properties.[7,9] Therefore, finer martensite with high hardness is desired, which can be achieved by Nb microalloying with heat treatment to retard the softening effect at higher tempering temperatures.

Carola et al.[10] studied the influence of the prior austenite grain size (PAGS) on the formed martensite. Moreover, they studied the effect of the austenitization conditions on low-carbon steel, determining the PAGS from which the martensite would be formed. It was found that grain refinement lowers the start temperature of martensite (Ms) and increases the rate of the initial stages of the transformation as a result of a more prominent grain boundary area that increases the density of nucleation sites.

In the literature, there are no investigations on the influence of microalloying of Nb accompanied by thermal cycling on the microstructure evolution of Armox 500T alloys. Therefore, this work aims at studying the combined effect of Nb and B microalloying and heat treatment on the grain refinement and hardness of Armox 500T. Additionally, the kinetics of phase transformation is studied theoretically using Thermo-Calc whose outcome is confirmed by dilatometry results.

2 EXPERIMENTAL PART

2.1 Melting and casting

Two different steel compositions were used in this current investigation. The melting process was performed in a medium-frequency 300-kg induction furnace with a quartzite lining. The main raw and alloy additions, used to achieve the final chemical compositions shown in **Table 1**, were high-purity steel scrap, carburizer, Fe-Si (85.0 *w*/% Si), Fe-Cr (70.0 *w*/% Cr), Fe-Mo (70.0 *w*/% Mo), Fe-B (60.0 *w*/% B), Fe-Nb (70.0 *w*/% Nb), pure Mn and Pure Ni. It had been reported that the alloying of molten steel with FeNb requires specific control parameters to avoid segregation inside the molten steel as well as increase its recovery in the solidified castings.[11] In addition, FeNb and FeB exhibit high affinity to the carbon element at high temperatures and can easily form strong Nb and B carbides, which can segregate at the grain boundaries. Accordingly, FeNb and FeB were charged into the final step of the melting process of steel in the form of fine particles (below 50 mm). After completed melting and alloying additions, the liquid steel temperature was measured using a digital handheld pyrometer and then poured at 1650 °C. The molten steel was poured into green sand moulds, and the cast shape was a keel block with dimensions of (200 × 150 × 25) mm. The green sand moulds were then left for free cooling to room temperature. **Table 1** shows the chemical compositions of the modified Armox alloys in this research. The apparatus used for the analysis of the chemical composition was an optical emission spectrometer (OES), Oxford Foundry-Master Pro (Hitachi Ltd, Tokyo Japan).

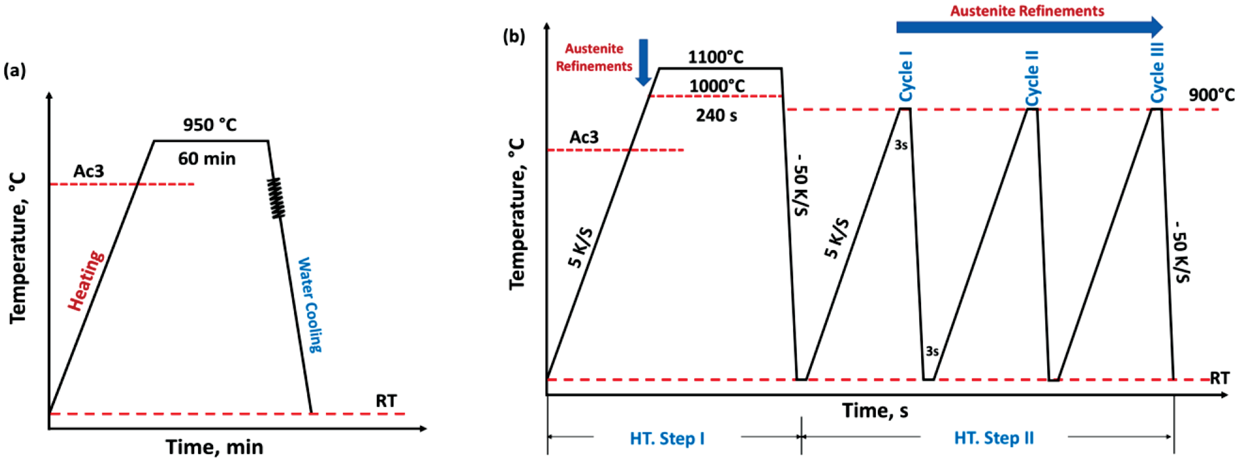


Figure 1: Hot forging and heat treatment process conducted on the designed Armox alloys: (a) hot forging process, (b) austenite-refinement heat treatment cycles

2.2 Thermodynamic calculations

Thermodynamic calculations were conducted to simulate and determine the critical transformation temperatures as well as predict the phase compositions under equilibrium that help design the heat treatment and forging processes. These thermodynamic calculations were conducted using the ThermoCalc (TC) software with the TCFE11 database (Thermo-Calc Software AB, Solna, Sweden).

2.3 Hot forging process

The as-cast samples were annealed in a heat-resistance furnace to 950 °C and held at this temperature for 60 mins. The annealed samples were then multi-pass freehand hot forged to round bars with a diameter of 25 mm. The free hot forging process was performed using a 250-kg capacity hammer machine. The forged bars were then water-cooled to room temperature. A schematic drawing of the hot forging process is shown in **Figure 1a**.

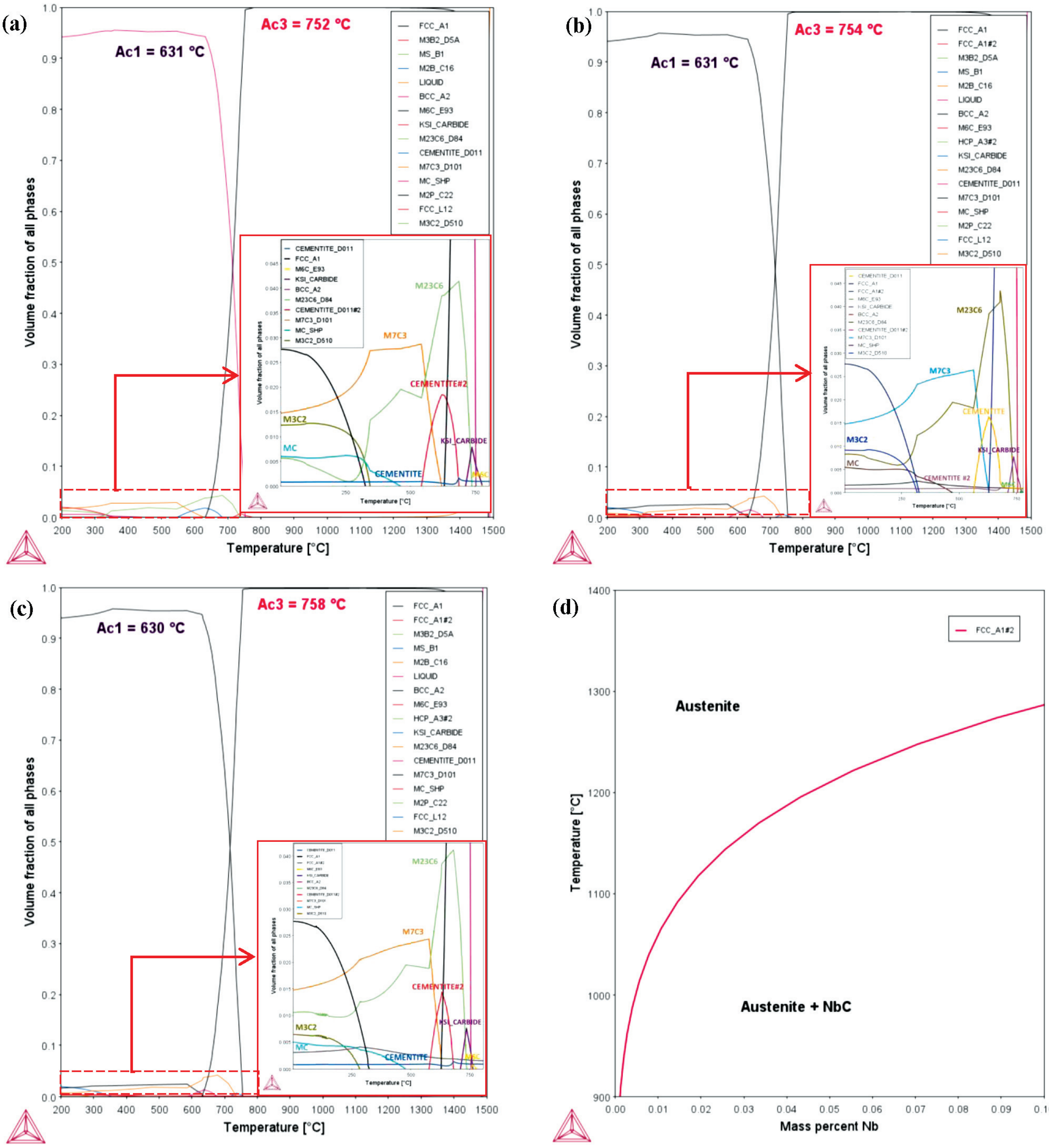


Figure 2: Equilibrium phase fraction as a function of temperature showing the critical temperatures of: a) Armox 500T-R, b) Armox 1-0.07Nb, c) Armox 2-0.1 Nb, d) equilibrium phase diagram showing Nb solubility in austenite for Armox 500T

Phase definitions: **FCC_A1** (Fe, Si, Mn, P, S, Cr, Ni, Mo, Nb)$_1$(VA, C, B)$_1$; **BCC_A2** (Fe, Si, Mn, P, S, Cr, Ni, Mo, Nb)$_1$(VA, C, B)1; **CEMENTITE_D011** (Fe, Si, Mn, Cr, Ni, Mo, Nb)$_3$(C, B)1; **M6C_E93** (Fe, Ni)$_2$(Mo, Nb)$_2$(Fe, Si, Cr, Ni, Mo)$_2$(C)1; **KSI-CARBIDE** (Fe, Cr, Mo)$_3$(C)1; **M23C6_D84** (Fe, Mn, Cr, Ni)$_{20}$(Fe, Mn, Cr, Ni, Mo)$_3$(C, B)6; **M7C3_D101** (Fe, Si, Mn, Cr, Ni, Mo, Nb)$_7$ (C, B)3; **MC_SHP** (Mo)$_1$(C)1, **M3C2_D510** (Cr, Mo)$_3$(C)$_2$

2.4 Heat treatment cycles

Several heat treatment strategies have been applied to the investigated alloys to confirm the decisive role of the austenite grain size on the final martensite morphology. The first step of these heat treatment cycles started with heating the samples above the temperature of Ac_3 at two austenitizing temperatures, 1000 °C and 1100 °C, and then held for 240 s. The main objective of selecting several austenitization temperatures is to vary the prior austenite grain size. The second heat treatment step included three consecutive thermal and rapid quenching processes (see **Figure 1b**). It was reported that applying several rapid heating and quenching processes increases the tendency of austenite nucleation and ultimately results in a significant refinement in the final microstructure.[10,12] All heat treatment processes were carried out using a quenching dilatometer, LINSEIS L78/RITA (Linseis Messgeraete GmbH, Germany). Cylindrical specimens with a 3-mm diameter and 10-mm length were machined from the forged bars. The change in the length was recorded against the temperature and time during the computer-controlled test cycle. The heat treatment cycles were conducted under a vacuum of 5×10^{-2} Pa using a high-frequency induction-heating generator, while high-purity helium gas was used for the cooling stages.

2.5 Metallography characterizations

All the samples were prepared for a metallography analysis including mounting, grinding and polishing. A 3-% nital solution was used for etching the samples. The microstructure characterization was performed using a digital inverted metallurgical microscope (Olympus, GX71). Scanning electron microscopy was also conducted on deep-etched samples using a JEOL JSM-6010LV.

2.6 Hardness measurements

Brinell hardness tests were done using a ZwickRoell ZHU 250 universal hardness testing machine using a load of 100 N and a dwell time of 20 s.

3 RESULTS AND DISCUSSION

3.1 ThermoCalc calculations

Figures 2a to **2c** show ThermoCalc diagrams of the predicted phases and the effect of different Nb contents on the critical transformation temperature of Armox 500T. Theoretically, under equilibrium, the starting and finishing austenitization temperatures Ac1 and Ac3 for the Armox-500T without any Nb additions are 631 °C and 752 °C, respectively (see **Figure 2a**). According to **Figures 2b** and **2c**, increasing the Nb additions to 0.13 *w*/% has no thermodynamic influence on the austenite Ac1, but it causes a slight increase in the austenite end temperatures Ac3 under equilibrium. Furthermore, under-equilibrium phases such as M_3C_2, MC, M_7C_3, $M_{23}C_6$ and KSI carbides were predicted for the investigated alloys. Nevertheless, **Figure 2d** indicates that niobium carbides (NbC) might precipitate and segregate at high temperatures. The same conclusion was also observed and reported by a previous study.[13] However, it was reported that the precipitation of NbC can reduce and refine the grain size of austenite during the deformation and heat treatment processes by impeding the grain boundary movement; ultimately, this might lead to a more acceptable martensite morphology of the investigated steel alloys in this work.[14,15]

3.2 Phase transformations

Dilatation curves for the heating and cooling stages of the investigated steel samples are shown in **Figure 3**.

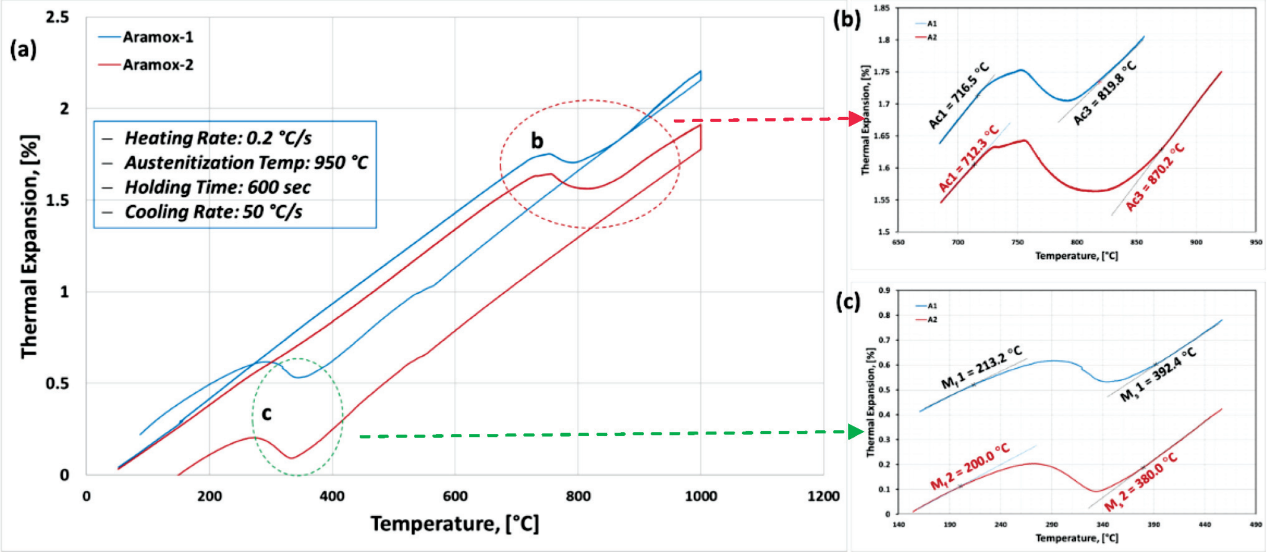


Figure 3: Dilatation chart during heating and cooling of the modified Armox alloys

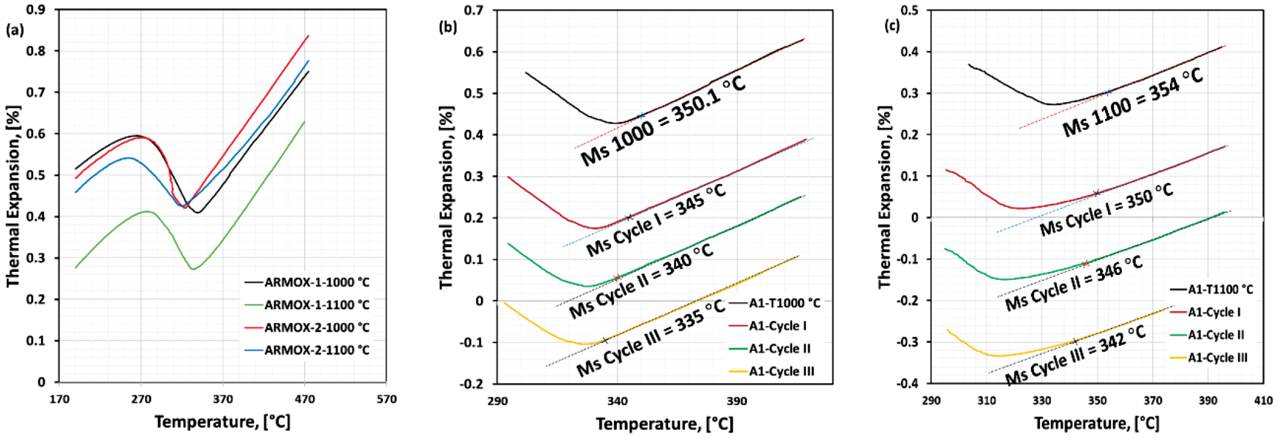


Figure 4: Dilatometry charts for the cooling stages of the heat treatment process described in **Figure 1b**: a) at step I for the investigated Armox alloys, b) at step II for Armox-1 heat treated at 1000 °C, and c) at step II for Armox-1 heat treated at 1100 °C.

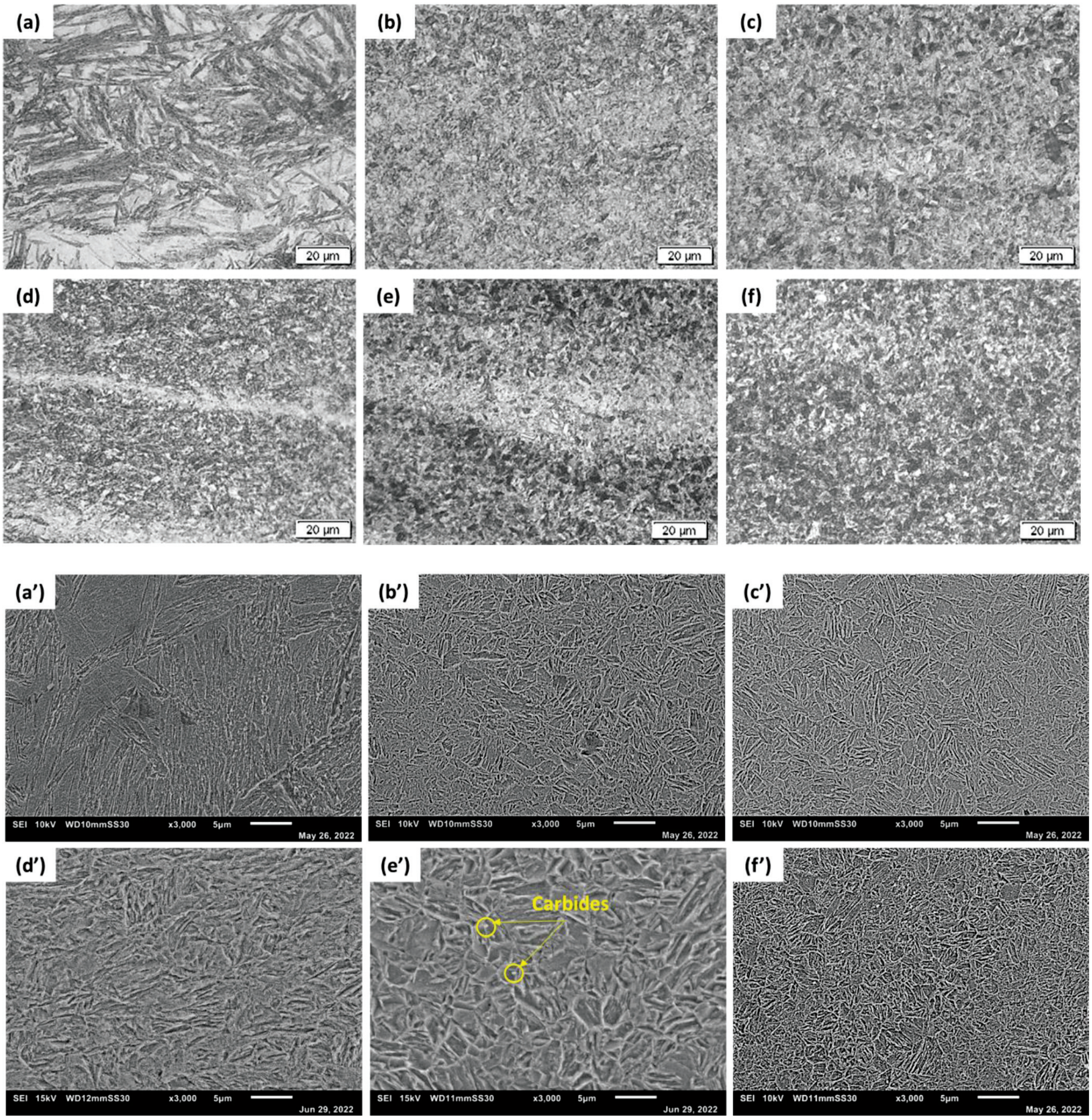


Figure 5: Optical and scanning electron micrographs of the investigated Armox alloys: (a, a') Armox-1 after forging, (b, b') Armox-1 after the heat treatment cycles at 1000 °C, (c, c') Armox-1 after the heat treatment cycles at 1100 °C, (d, d') Armox-2 after forging, (e, e') Armox-2 after the heat treatment cycles at 1000 °C, (f, f') Armox-2 after the heat treatment cycles at 1100 °C

The investigated samples were heated to 950 °C at 0.2 K/s, held at that temperature for 600 s and then rapidly cooled at 50 K/s to room temperature. The critical transformation temperatures, including the start and finish temperature of the austenite transformation (Ac_1, Ac_3) and the martensite start and finish temperature (Ms, Mf), were calculated. It was found that by increasing the Nb and B content to 0.13 and 0.0048 *w*/%, respectively, for sample Armox-2, the austenite finish temperature Ac_3 increased by 50 °C, which resulted in widening the range between Ac_1 and Ac_3 by 55 °C. Additionally, the martensite start and finish temperatures shifted down by 13 °C.

3.3 Martensite start temperature

Figure 4 displays parts of dilatation curves for cooling obtained from the heat treatment cycles described in **Figure 1b** for the two investigated steel samples. A minor shift observed in **Figure 4a** dilatation curves for the two proposed Armox alloys during cooling at two austenitization temperatures (1000 °C and 1100 °C), based on the relation between the thermal expansion and temperature can be attributed to both the differences in the alloy compositions and the formation of carbides. It can be noticed that the martensite transformation continuously shifts to lower temperatures with step II of thermal cycling I, II and III (see **Figures 4b** and **4c**). Celada-Casero et al.[10] and J. Hidalgo et al.[12] reported that the thermal expansion related to the martensite transformation shifts to lower temperatures with a decreasing austenite prior grain size. This might explain the austenite refinement in both modified Armox steel samples, even at higher annealing temperatures (see **Figure 5**).

3.4 Microstructure and grain-size characterization

Figure 5 depicts optical and scanning electron micrographs of the investigated samples in different conditions. A fully martensitic structure can be noticed in all the samples. However, the martensite lathe sizes differ with the increasing Nb and B content; for instance, after hot forging, Armox-2 shows a more refined martensitic structure than Armox-1. Moreover, after the austenite refinement cycles described in **Figure 1b**, both samples, Armox-1 and Armox-2, showed a significantly finer martensitic structure than after forging. Additionally, increasing the annealing temperature from 1000 °C to 1100 °C resulted in a slight difference in the martensitic morphology. Furthermore, Armox-2 showed more carbides and precipitates than Armox-1, as predicted in the thermodynamic calculations from **Figures 2b** and **2c**.

Figure 6 shows grain-size calculations. It was found that the average grain length in hot forged Armox-1 was 6.64 µm, which decreased after heat treatment austenite refinement cycles at 1000 and 1100°C to be 2.25 and 2.64 µm, respectively. Moreover, the increase in the Nb and B content caused a decrease in the grain size by nearly 50 % in hot-forged Armox-2, being 3.55 µm. However, after the heat treatment cycles at 1000 and 1100°C, the grain size was 2.54 and 1.94 µm, respectively, nearly the same as for Armox-1.

3.5 Hardness measurements

Figure 7 shows the Brinell hardness values of the investigated alloys measured at different stages of the heat treatment cycle. It can be noticed that the highest hardness values were recorded for hot forged samples, 563 and 577 HBW for Armox-1 and Armox-2, respectively.

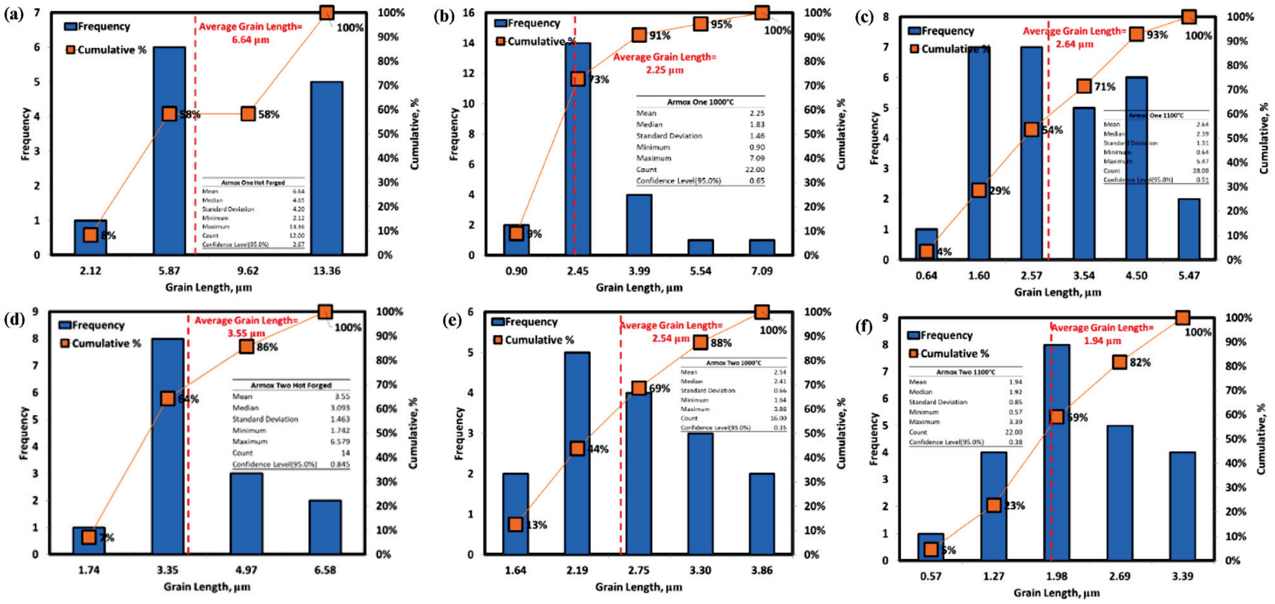
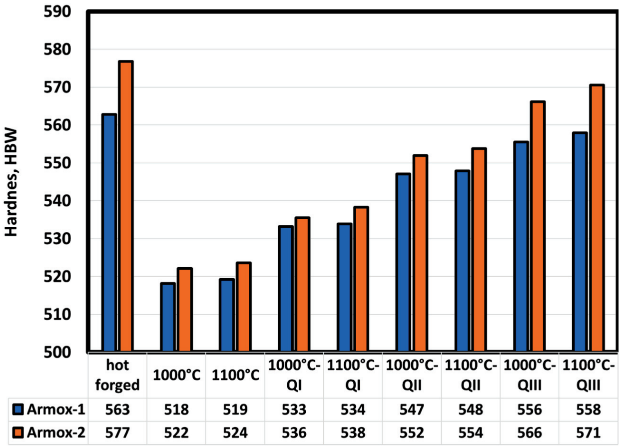


Figure 6: Grain-size measurements of the investigated Armox alloys: a) Armox-1 after forging, b) Armox-1 after the heat treatment cycles at 1000 °C, c) Armox-1 after the heat treatment cycles at 1100 °C, d) Armox-2 after forging, e) Armox-2 after the heat treatment cycles at 1000 °C, f) Armox-2 after the heat treatment cycles at 1100 °C



	hot forged	1000°C	1100°C	1000°C-QI	1100°C-QI	1000°C-QII	1100°C-QII	1000°C-QIII	1100°C-QIII
🟦 Armox-1	563	518	519	533	534	547	548	556	558
🟧 Armox-2	577	522	524	536	538	552	554	566	571

Figure 7: Brinell hardness measurements of the investigated Armox alloys

Gradual increases in the hardness were observed until reaching stage Q-III of the thermal cycling. Moreover, Armox-2 exhibited higher hardness values than Armox-1 due to the carbide formation and grain refinement, resulting from increasing the Nb and B *w*/%. The hardness values confirmed the positive impact of Nb, B and heat treatment on the refinement of the produced martensitic structure. These results confirm the assumption that a more refined structure leads to increased hardness values.

4 CONCLUSIONS

This research successfully developed two Armox steel alloys modified with two different amounts of Nb and B. The following points summarize the main findings of this research:

- Thermodynamic calculations predicted the formation of different carbide phases and precipitates such as M_3C_2, MC, M_7C_3, $M_{23}C_6$, and KSI carbides. Additionally, NbC could precipitate and segregate at high temperatures and refine the grain size of the austenite during the deformation and heat treatment processes.
- The dilatometry measurements revealed that increasing the Nb and B content increased the austenite finish temperature Ac_3 by 50 °C, which resulted in widening the range between Ac1 and Ac_3 by 55 °C. Additionally, the martensite start and finish temperatures shifted down by 13 °C.
- Heat treatment cycles that were performed to obtain a more refined structure, revealed that the martensite transformation temperature constantly shifted by 5 °C to lower temperatures in step II, which included thermal cycles I, II and III for the two investigated alloys.
- Optical and SEM micrographs of the Armox-1 heat-treated samples showed fine grain sizes of 2.25 µm and 2.64 µm at 1000 and 1100 °C, respectively. The heat-treated Armox-2 samples achieved 2.54 µm and 1.94 µm grain sizes at 1000 °C and 1100 °C, respectively.
- The Brinell hardness values confirmed a positive impact of Nb, B and heat treatment on the refinement of the produced martensitic structure. The hardness of the modified Armox alloys in this research was improved by 14 % compared to the conventional Armox 500T.
- The results obtained with this research may have promising applications in the protection of ATMs, VIP vehicles, helmets and personnel bulletproof vests.

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