Laser beam hardening of carbon and low alloyed steels: discussion of increased quantity of retained austenite

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It was shown that mainly three effects are responsible for the increased quantity of retained austenite in carbon and low alloyed steels after laser transformation hardening. At low peak temperatures a high dislocation density appears in the initial austenite phase during the reversed polymorphic transformation which is preserved till the martensite transformation begins and retards this reaction. At high peak temperatures this is more complete carbides dissolution than after standard furnace hardening. Both effects are overlapped by a third one. This is the carbon up-hill diffusion resulting in surface enrichment of carbon and a decrease of the martensite start temperature. © *1999 Kluwer Academic Publishers*

1. Introduction
Various assumptions are suggested to explain the increased content of retained γ-phase after laser hardening (LH). In the early studies the effect was explained by partial suppression of the martensite transformation because of the high cooling rate. However, according to [1] this is a result of the bainite transformation. Today, the most generally accepted explanation is that laser heating leads to higher temperatures than during conventional hardening, that the carbon is not homogeneously distributed due to the short austenizing time and that grain refinement occurs. However, some of our results [2–6] do not fit within those explanations, initiating the present discussion.

2. Experimental
For the experiments plain carbon steels (between 0 and 1.2 wt % C), low alloyed steel (1 wt % C-1.5 wt % Cr) and stainless austenitic steel (0.04 wt % C-18 wt % Cr-10 wt % Ni) were used. The laser treatment was carried out by a pulsed Nd:GLASS-laser in three temperature regimes: above the melting point (LSM), above Ac_3, near the melting temperature (complete laser hardening: LH_c) and between Ac_1 and Ac_3 (incomplete laser hardening, i.e. with incomplete austenization: LH_i). Prior to laser processing, the samples were subjected to furnace hardening.

The quantity of the γ-phase was measured by X-ray diffraction. The grain size was determined metallographically. Nital etch was used. To improve the resolving of the grain boundaries, the samples were tempered at $400\,^\circ$C for 2 h prior to etching. This temperature is far below the recrystallization temperature and therefore the tempering should not influence the state of the high-angle boundaries. The experimental processing is described in more detail in references [2–6]. Demo ver-

sion of the software used for computer simulation of the laser beam induced heating and cooling cycles: http://star.sstu.samara.ru/~physics/fedosov/thermo_d.zip

3. Discussion
It is possible to assume *a priori*, that the increased content of retained γ-phase in a structure of carbon and low alloyed steels, subjected to LH, could be a consequence of the following effects:

- a high cooling rate;
- a higher peak temperature compared to furnace hardening under standard conditions;
- a short austenizing time;
- grain refinement;
- greater strain hardening of initial austenite before the martensite transformation starts;
- influence of plastic deformation and stresses;
- nitrogen pick-up from the atmosphere during irradiation;
- up-hill diffusion of carbon.

Each of these is considered in detail below.

3.1. Cooling rate
An increased amount of the retained γ-phase as a result of LH, contradicts the known dependence, that this phenomenon is observed with decreased, but not increased quenching rates within the regions of bainite and martensite transformation [7, p. 24]. However, the suggestion has been made [1, 8] that during laser processing the cooling rate is high only at high temperatures, but when the temperature decreases, it drops fast and becomes even lower than typical rates for conventional quenching. This leads to bainite transformation

and, as result, in an increased retained austenite (rA) content.

It is clear that the cooling rate decreases when the temperature approaches equilibrium. But what is its real value at the temperature intervals of the bainite and martensite transformation? We simulated heating and cooling rates as a function of the current temperature and the peak temperature as a variable parameter [5]. When the peak temperature on the surface was 1200 °C (solidus temperature of 1.0% C-1.5% Cr-Fe steel) and the duration of the laser pulse was 1 ms, the following cooling rates at certain temperature levels were calculated using the thermophysical data of carbon and low-alloyed steels:

$$700\,°C–3.7 \times 10^5 \text{ K/s}$$
$$400\,°C–6.6 \times 10^4 \text{ K/s}$$
$$200\,°C–8.2 \times 10^3 \text{ K/s}$$
$$100\,°C–1.0 \times 10^3 \text{ K/s}$$

These temperatures correspond with the temperature intervals of the polymorphic, bainite and martensite transformations of high-carbon steels. One can see, these cooling rates exceed those typical for water quenching (130–270 K/s).

How can such high cooling rates result in increased of rA values? This can only be explained by assuming a decreased M_S temperature or altered kinetics of the martensite reaction: the cooling occurs faster than does transformation and at low temperatures the latter would slow down. In fact, this means a transition of the athermal kinetics of martensite transformation to isothermal kinetics. Evidently, the latter is impossible in carbon steels [9, 10]. Moreover, LH is usually carried out at room temperature when isothermal stabilisation is not observed, even in higher alloyed steels [10, p. 61].

In the literature there is data on cold processing of irradiated materials. However, such treatment carried out after laser processing (after cooling to ambient temperature) and therefore, naturally, it can in no way influence on kinetic slow down of the transformation. On the contrary, it decreases of γ-phase content owing to its transformation to a martensite when low temperatures.

It was theoretically shown [11], that an increase of the cooling rate does not decrease, but increase M_S. However, this change is insignificant. So for a 1.1% carbon steel the M_S increases by 1 K with a cooling rate of 10^3 K/s and by 7.5 K with a cooling rate of 10^8 K/s. Obviously, such small changes of the martensite start temperature cannot have a significant influence on rA amount.

This calculation was proved experimentally [12]. Thin sheets (thickness 0.2 mm) of a 1.2% C-Fe steel were quenched in a special high-rate sprayer refrigerating unit after vacuum heating. The measured M_S temperature appeared to be independent of the cooling rate within a range of $10–60 \times 10^3$ K/s. Moreover, the content of rA even decreased slightly with increasing cooling rate.

The experiments on 0.45% C steel [16] showed a discontinuous change in M_S. Within the range of $1–12 \times 10^3$ K/s, $M_S = 360\,°C$. Thin layers of rA, formed by partial bainite reaction, were detected. At higher cooling rates the M_S decreased to 300 °C and the rA disappeared. Comparing the cooling rate where the decrease in M_S occurs with those typical for LH, it can be concluded that this phenomenon is one possible explanation for the difference in the amount of rA observed in some references [2, 13].

From the above discussion it can be concluded, that the high cooling rates, typical for LH, cannot explain the increased content of retained γ-phase. The indirect influence of the cooling rate through changes of the structure and substructure will be considered below.

3.2. Austenization temperature

Hyper-eutectoid steels usually are quenched from intercritical temperatures (Ac_1–Ac_3). With increasing austenization temperature, the amount of dissolved carbides and the carbon concentration in the austenite also increases, which causes a decrease of M_S and an increase of rA content [14, p. 217, Fig. 152; p. 179, Fig. 256]. When the austenization temperature exceeds the Ac_3, the carbon concentration in the austenite reaches its maximum and the retained γ-phase content decreases again due to grain growth [7, p. 18].

When LH, the surface is usually heated up to higher temperatures then during conventional quenching. Therefore it is need to compare rA amount after hardening from similar temperature regimes.

In our experiments between 28 and 29% of rA were measured after water quenching of 1.0 wt % C-1.5 wt % Cr-Fe steel from a temperature of 930 °C (holding time 40 min, thickness of the samples 8 mm). An increase of the austenization temperature (always provided that the holding time is sufficient for complete dissolution of the carbides) led to no further increase of rA. In samples of the same steel after LH_c between 40 and 42% rA were measured [2, 6].

Similar results were obtained for a 1.2% C-Fe steel. The rA content was between 47 and 48% after LH_c [2], while water quenching from high temperatures led only to 38% rA ([12] and Fig. 1). These results show that a high austenization temperature is not the only cause of the increased rA content after the LH_c.

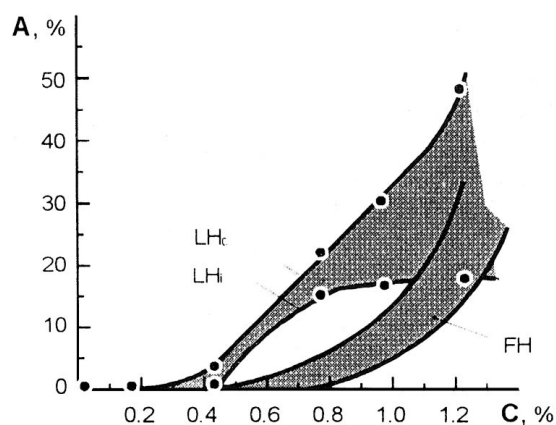The above discussion refers to LH_c with austenization temperatures near the solid-liquid phase transition.

Figure 1 Effect of carbon on the retained austenite content in steel after laser hardening (LH_c, complete; LH_i, incomplete laser hardening) [2]; FH, after conventional furnace hardening, generalised data [31].

As Fig. 1 shows, an increasing *rA* content is also observed when the peak temperature is between Ac_1–Ac_3 (LH_i). It is clear, that different mechanisms, not correlated with the austenization temperature or with the degree of dissolved carbides, are responsible for this effect.

3.3. Inhomogeneous distribution of carbon

The increased amount of *rA* after LH is attributed by many workers to an insufficient homogenisation of the carbon concentration in the γ-phase due to the short time of austenization [1, 15–19]. In regions with a high carbon content, the start temperature of the martensite transformation is decreased resulting in a higher amount of *rA*. However in [1, 15–18] the samples were irradiated in an annealed or high-tempered condition—which necessitates the dissolution of carbides during LH. While in our work, a homogeneous carbon distribution structure was ensured prior to laser processing by furnace hardening. The homogeneous carbon concentration also explains why variations of the pulse duration showed an insignificant influence on the *rA* content in our experiments [2, 6].

3.4. Grain size

The effect of the grain size on M_S is well known [20]. Can an increased *rA* proportion after LH be connected to a decrease in the grain size?

Sadovskiy *et al.* showed recovery of the grain size after LH [20–24]. However, other workers found contradicting results [8, 18, 25–27]. Therefore, we carried out metallographic analysis of specimens where we measured the *rA* content.

According our metallography, LH_c leads to a deeper etching of the grain boundaries but has no significant effect on the grain size (section of 1.0% C-1.5% Cr-Fe steel are shown in Fig. 2; the similar patterns were observed in other samples).
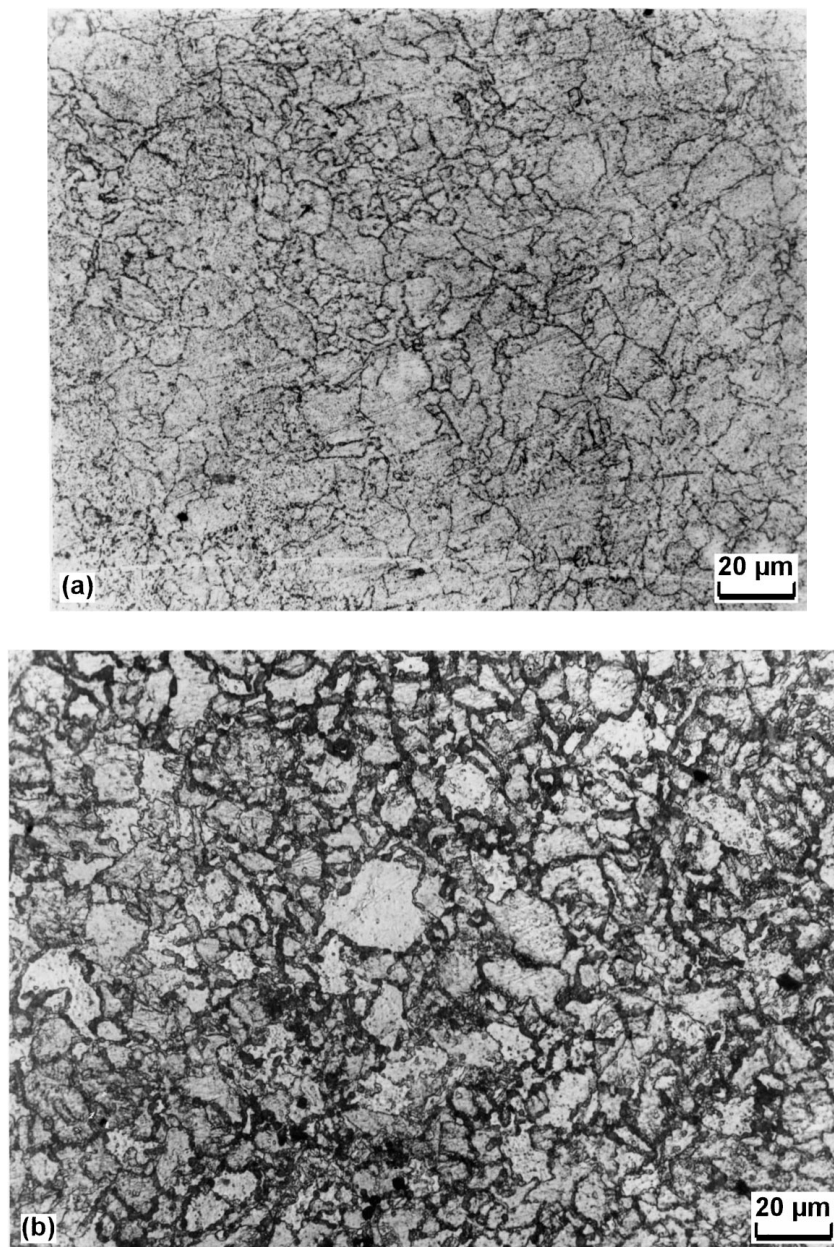

Figure 2 Microstructure of 1.0% C-1.5% Cr-Fe steel after laser processing and 400 °C tempering during 2 h: (a) in nonprocessed and (b) in processed zones. ×500.

3.5. Strain hardening of austenite

The energy necessary for start of the martensite transformation is:

$$\Delta F_{\alpha\gamma} \geq F_N + F_\tau$$

This corresponds to supercooling (ΔT), where:

$$\Delta T = T_0 - M_S = (F_N + F_\tau)/\Delta S_{\alpha\gamma} \qquad (1)$$

where $\Delta S_{\alpha\gamma}$ is entropy of the transformation, T_0 the equilibrium temperature, F_N the surface energy of the nucleation of the martensite crystals, and F_τ is the energy of deformation (the difference in specific heat of the phases can be neglected) with

$$F_\tau = 0.5(V\tau\varphi_{\gamma\alpha}) \qquad (2)$$

where V is molar volume, $\varphi_{\gamma\alpha}$ the shear angle for the $\gamma \to \alpha$ transformation, and τ the shear stress [20].

Thus, any hardening of the initial austenite must reduce M_S (e.g. [28]). It should be noted that the above discussed influence of the grain refinement on M_S is also connected with a strengthening effect of the high-angle boundaries.

Hardening of the initial austenite can occur due to an increase in dislocation density as a result of thermally induced deformation (high temperature gradient) and inherited of dislocations formed during the $\alpha \to \gamma$ transformation [10, p. 62]. The latter is reasonable, considering the short time of a laser processing.

There are other statements on the influence of deformation on M_S. According [29] only the grain refinement lowers M_S, while an increased dislocation density causes its rise. Other references with similar statements are quoted in Ref. [20].

This discrepancy is related to the complexity of plastic deformation on the martensite transformation, which cannot be simply derived from the above equations. The effect depends strongly on the degree of plastic flow: a high deformation hampers the transformation whereas a low deformation activates it [10, p. 61]. This is due to the energy of formation of martensite crystals [30].

For structural broadening of X-ray lines of retained γ-phase, we can write:

$$\beta = \beta_l + \beta_M \qquad (3)$$

where β_M is a structural broadening which appears as a result of phase hardening during a martensite transformation and β_l is a structural broadening before the beginning of the martensite transformation.

It is clear from Fig. 3, that for both furnance hardening and LH$_c$, β depends basically on the content of the γ-phase, i.e. on the degree of the martensite transformation. Taking into account, that the martensite transformation influences β_M but not β_l, it can be concluded that β_l is constant for this case. In other words, there is no increased dislocation density in the initial austenite before the martensite transformation starts during LH$_c$ in comparison with the analogous parameter during furnace quenching.

Conducting LH$_i$ leads to a different effect. Now structural broadening of the γ-phase lines increases signif-
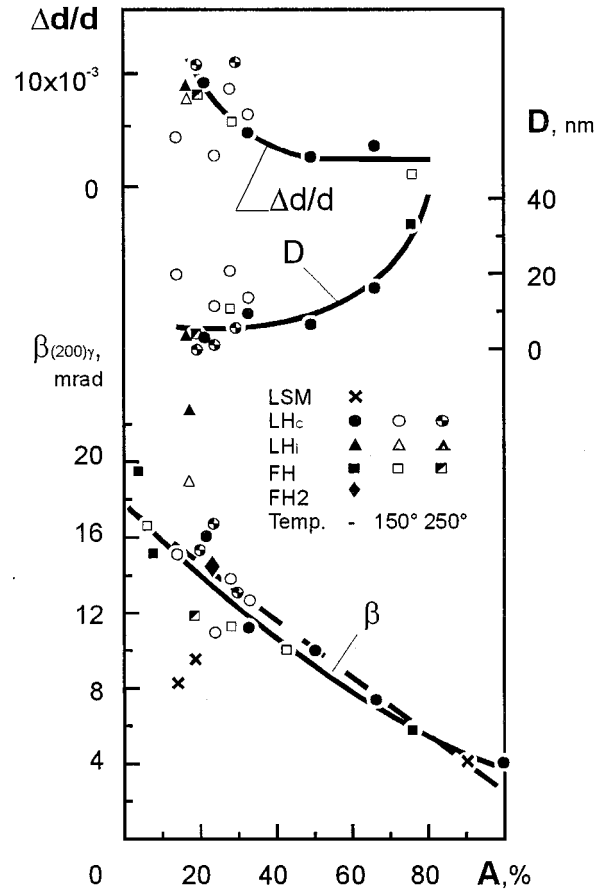

Figure 3 Dependence of the structural broadening of lines $(200)_\gamma$, the size of mosaic blocks and microdistortions of the crystal lattice of retained austenite on its content in steel (the solid line, triple term approximation; dotted one, linear approximation). Symbols in the graph: LSM, laser surface melting; LH$_c$, complete and LH$_i$, incomplete laser hardening; FH, furnace hardening; FH2, double furnace hardening; Temp., tempering temperature [3].

icantly (Fig. 3), which can be attributed to an increase of β_l only. The most probable explanation of this fact is that dislocations, formed by phase-hardening during the $\alpha \to \gamma$ transformation, are preserved in the austenite because there is insufficient time for their annealing at low heating temperatures. Thus there is an increase in the density of dislocations in the austenite before the martensite transformation starts [3]. This will restrain the martensite transformation and, consequently, increase the rA content, which is in fact observed for such peak temperatures (see curve LH$_i$ in Fig. 1).

The corresponding change of the M_S can be calculated using Equations 1 and 2 and data of Fig. 3. Assuming that at the initial γ-phase $\Delta\tau$ is proportional to β, taking into account that the surface energy of the semicoherent boundary of α- and γ-phases is about 100–200 mJ/m^2 for iron based alloys [20], that for medium- and high-carbon steels ΔT is of the order of 200 K, the depression of the M_S in a 0.8% C-Fe steel will be 70–80 K. This must be followed by 10–15% increase of rA content [10, 31]. Fig. 1 shows that the difference in rA content for LH$_i$ and furnance hardening has the same magnitude.

An increase of the peak temperature and an extension of the laser exposure time will increase the recovery of dislocations formed during $\alpha \to \gamma$ transformation and hence will reduce their density before the martensite

transformation starts. This must cause a reduction in *rA* content.

The same changes of the beam parameters will increase the dissolution of carbides, which leads to a decrease of M_S. Thus, the two mechanisms have an opposite effect which weakens the influence of LH conditions on the *rA* content.

It is known that *plastic deformation* at temperatures below M_S causes an acceleration of the martensite transformation. Does it occur in the zone of laser hardening? Or, at least, is the effect stronger than during conventional quenching? The latter can be supposed because of the high temperature gradients typical for LH.

So, some slip lines were found by TEM analysis in austenite crystals of irradiated dual (alpha-gamma) Fe-Ni alloy [32]. However, the average value of such deformation is very small. Furthermore, the observed slip can be the effect of the recrystallization in neighbouring alpha regions, not a thermoshock effect. In this case, its magnitude during LH is no more than during conventional quenching.

Indeed, X-ray tests of the nontransformable austenitic steel exposed to laser processing have shown an absence of plastic hardening [3]. A similar conclusion can be made from the above discussed correlation between structural broadening of γ-phase lines as a function of the degree of martensite transformation (Fig. 3). This indicates that thermal stresses are too low for essential plastic deformation.

The changes in the kinetics of the martensite transformation are possible because of a *stress effect* (even if the stresses are too low for plastic deformation to occur). This can be explained, at least partially, with the Le Chatelier principle: tensile stresses favour a transformation accompanied by an increase in the volume, while compressive stresses hamper such transformation. It is known, that the martensite reaction is accompanied by increasing in volume.

According [7, p. 50] the M_S decreases about 7–8 K per 100 MPa hydrostatic (compressive) stress. The increase in M_S caused by tensile stresses is of the same order. When the temperature is far from M_S, this effect disappears.

The value and sign of residual stresses developing during LH depends on the ratio of the thermo- and the phase-components. It is evident that near the M_S the contribution of the volume expansion due to martensite formation is negligible. Thus the stresses will be tensile due to a restricted shrinkage of the heated volume [33]. However, these stresses are not very high (25–43 MPa for ferritic steel [34]). In this way, developing stresses favour the martensite transformation, but the effect is not very significant (M_S will be raised by 3–4 K).

3.6. Nitriding during LH in air

If nitriding occurs during LH in air, as it is indicated in [35, 36], it should lead to an increase in the *rA* content, since nitrogen stabilises austenite. However, in our experiments [2–5] laser treatment was conducted in a helium environment. And we have not observed significant difference in the *rA* content with laser processing
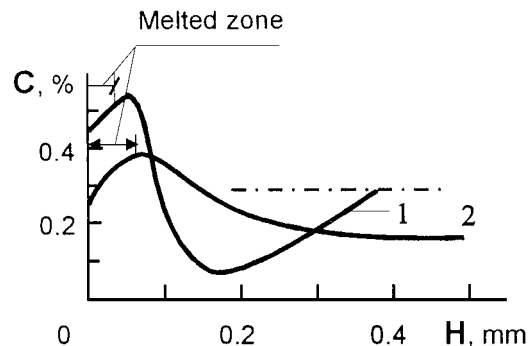

Figure 4 Distribution of carbon concentration (*C*) with a depth (*H*) after laser irradiation: 1. 0.3% C-24% Ni-Fe alloy [37]; 2. low carbon steel (the stroke-point line, initial content of carbon) [38].

in air [6]. Perhaps, in the latter case nitriding occurs together with decarburizing. And they compensate influence of each other on the *rA*.

3.7. Up-hill diffusion of carbon to the surface

Data on the enrichment of the surface layers of a steel with carbon as a result of laser processing was given in work [37]. It was shown, that this effect occurs because of up-hill diffusion of carbon atoms to layers undergoing polymorphic transformation. The determination of the carbon concentration in a solid solution in that work was carried out by X-ray analysis, which is not a very reliable method to measure the carbon concentration. However, this data confirms previous results, where a method of layer-by-layer spectral analysis in microvolumes was applied [38, p. 84]. SEM and EDX analysis were used in work [39]. The appropriate plots are shown in Fig. 4. It can be seen that the carbon concentration was increased about 0.2% after LH in the solid state. This effect should increase the *rA* content in the range of 3–7% [7, 31, p. 231, Fig. 200, p. 232, Fig. 201].

It can also be seen, that for conventional hardening *rA* appears in steels with 0.4–0.6 wt % carbon, whereas after LH it is occurs at carbon level of 0.2–0.4 wt % C (Fig. 1). The difference of 0.2 wt % is exactly the value of up-hill diffusion enrichment and so might be an explanation for the observations noted above.

The reduction of the *rA* content after LSM in comparison with its content after LH is also supported by the above stated explanations. Melting completely removes work hardening of the austenite arising from the $\alpha \rightarrow \gamma$ transformation. A dendritic structure is formed [2, Fig. 3] which means an increase of space between high-angle boundaries. In addition, melted layers are less enriched with carbon than solid state hardened ones (see Fig. 4).

4. Conclusions

Summarising the above discussion we can draw the following conclusions concerning the explanation of the increased *rA* content after laser hardening of furnance hardened carbon and low-alloyed steels:

1. At relatively low peak temperatures (low beam energy), the increase rA content is due to the effect of an increased dislocation density which originated in the initial austenite during reverse polymorphic transformation and which is not recovered when the martensite transformation starts because of a short interaction time.

2. At high peak temperatures near the solid-liquid interface phase region (high beam energy), the increase rA content is due to the higher amount of carbide dissolution compared to furnance hardening under standard conditions.

3. An increase of the peak temperature and/or an increase of the pulse duration weakens the first mechanism and favours the second one. This weakens the influence of the laser processing parameters on amount of rA.

4. The effects of dislocation density and carbide dissolution are superimposed by a third mechanism: this is the up-hill diffusion of carbon resulting in an carbon enrichment in the near surface layers which causes an decrease of M_S.

Acknowledgements

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