Effect of Filter Functional Coating on Detrimental Nonmetallic Inclusions in 42CrMo4 Steel and Its Resulting Mechanical Properties

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Carbon-bonded ceramic foam filters with different functional coatings are immersed in a 42CrMo4 steel melt within a steel casting simulator. The solidified steel is analyzed with respect to the size distribution and the chemical composition of the remaining nonmetallic inclusions (NMI). Cyclic loading and quasi-static tests are performed to determine the fatigue limit, the strength, deformability, and toughness of the steel after filter immersion. The immersion of filter with calcium hexaluminate (CA6) coating significantly reduces the population of small (4–20 μm) NMIs. This leads to an increased deformability and, thus, ability for energy dissipation during deformation. However, the maximum size of NMIs is increased from 100 to 150 μm, which results in fatigue limit reduction, despite the decrease in NMIs total density. The majority of inclusions are found to be pure alumina. Large (up to 150 μm) plate-like alumina inclusions introduce most of the detrimental effects on cyclic strength, whereas significant effect on quasi-static strength is not found.

1. Introduction

Nonmetallic inclusions (NMI) detrimentally affect the mechanical behavior of steel. Among other grades, high-strength steels are more sensitive to the presence of NMIs in terms of crack initiation. The stress-raising effect acts under cyclic as well as quasi-static and dynamic loading conditions.

According to Kamal and Rahman, at least half of all mechanical failures are caused by fatigue. Most of metallic parts undergo cyclic loading during their utilization. These parts can be assumed as conditionally safe below the classical fatigue limit of 10⁷. However, the number of components that exceed 10⁷ cycles is growing along with the engineering development. This includes engines, rotors, turbines, automobile parts, train wheels, bridges, etc. In the absence of surface crack initiation, components can exceed 10⁷ cycles and undergo very high cycle fatigue (VHCF) life. In the VHCF regime, fatigue crack initiation occurs on the internal defects, among which NMIs are in first place acting as internal notches.

Analogously to cyclic loading, NMIs act as internal notches also during quasi-static loading. Consequently, voids are nucleated by decohesion or by fracture of the inclusions. These voids grow and eventually coalesce after a certain amount of deformation. The size, density, and spatial distribution of NMIs affect these damage processes. Larger inclusions promote the void nucleation as the critical strain is smaller. Furthermore, a smaller distance between the NMIs and, thus, a higher volume fraction of inclusions, reduces the deformability up to void coalescence.

Thereby, the reduction of the NMI content is crucial for obtaining the high mechanical properties of steel and other alloys. This can be achieved by different refining methods, among which metal melt filtration by ceramic foam filters is a very promising one. Steel refining by ceramic foam filters can be divided into two principal mechanisms: 1) active filtration—when a fluid flows through the filter which collects the inclusions by interception, gravitation, or inertia; 2) reactive filtration—when a filter surface or coating reacts chemically with the steel melt.

The latter promotes inclusions’ deposition and CO production in case of carbon-bonded refractories. CO bubbles purge the steel melt causing agglomeration of inclusions and their flotation to the slag layer.

Both filtration mechanisms can work together. However, to study them one should differentiate between active and reactive filtrations. To study the reaction of filter surface with the steel melt, one can introduce a filter inside the melt for a limited...
time. Such a procedure can significantly reduce the NMI population in steel after a filter immersion time of only 10 s. The changes in NMI population after reactive filter immersion are to be studied by the methods of particle analysis. Optical microscopic (OM) analysis of sections can reveal the NMI size distribution and is relatively fast in processing a large amount of statistical data. Scanning electron microscopy (SEM)-based analysis of sections is slower than OM, yet can reveal the distribution of chemical composition between NMIs by energy dispersive X-ray (EDX) mapping. Chemical extraction (deep etching) of NMIs is the most time-consuming process; however, it is the only precise method of revealing NMIs with complex 3D shapes, such as dendrites, interconnected agglomerates, shells, etc. Fracture surface analysis reveals the role of NMIs in crack propagation and their influence on the mechanical properties.

It is known that the largest NMI within a cyclically loaded volume initiates a fatigue crack, which can be used in particle analysis for defining the maximum inclusion size. This is achievable by cyclic loading in the VHCF regime by ultrasonic fatigue testing (USFT) and by additional protection of the surface from crack initiation (for example, by nitriding). Cooperative NMI analysis by section imaging, chemical extraction and fractography covers the whole spectrum of size and chemical composition distribution.

The aim of the present work is to study the influence of different reactive filter coatings on NMI population and mechanical properties of treated 42CrMo4 steel with the methods described above.

2. Experimental Section

2.1. Reactive Filter Treatment of 42CrMo4 Steel

Prismatic \((125 \times 20 \times 20 \text{ mm}^3)\) ceramic foam filters based on carbon-bonded alumina were produced by the replica technique with the composition and procedure presented by Emmel and Aneziris. In addition, functional coatings were applied on the filters by cold spray coating and the filters were heat treated again under reducing atmosphere. Coating abbreviations in Table 1 denote alumina \((\text{Al}_2\text{O}_3, \text{“+A” coating})\), alumina-based slurry \((\text{AC5, “+AC5” coating})\), carbon nanotubes and alumina nanosheets \((\text{CNT and ANS, “+Nano” coating})\) and calcium hex-aluminate \((\text{CA6, “+CA6” coating})\). Detailed description on the coatings design, production, and properties are given elsewhere.

A metal casting simulator (Systec, Germany) was applied to test the \(\text{Al}_2\text{O}_3\)-C filters with different surface functionalization. Before melting, the casting simulator was evacuated to a pressure of 2 mbar and filled with Ar 4.6 (purity = 99.996%). Thus, the whole remelting procedure until the melt solidification was conducted under the atmosphere of pure argon. Next, approximately 40 kg of commercially available 42CrMo4 steel \((\text{AISI 4142})\) was melted in a special crucible with a 220 mm diameter. It consisted of hydratable alumina-bonded alumina/ alumina-magnesia-spinel material, without any silica \((\text{SiO}_2)\), calcia \((\text{CaO})\), or further additions (to prevent unwanted reactions of the steel melt with the crucible), presintered at 1600 °C for 2 h. The total dissolved oxygen \([\text{O}]\) content and the temperature \(T\) of the steel melt were measured with a combined \(\text{pO}_2/T\)-sensor system \((\text{Heraeus Electro-Nite, Germany})\) at different stages of each test. Before the immersion of the filter, defined alumina impurities were created in the steel melt as follows. Once the desired temperature of 1650 °C was reached, 0.5 wt% \((\text{related to the steel mass})\) of an iron oxide mixture were added. Accordingly, the dissolved oxygen rose from 20–30 ppm up to 60–70 ppm. At this point, endogenous alumina inclusions were generated by adding 0.05 wt% \((\text{again related to the steel mass})\) of pure aluminum metal to the melt. Only about 10 ppm \([\text{O}]\) was detected after this step. Next, ceramic foam filter was dipped 60 mm deep into the center of the crucible with the melt and rotated along its own central axis with 30 rpm for 10 s. The reaction between the filter and the melt was enhanced by induction currents with the turbulent melt flow up to 4 m/s. Detailed analysis of the flow conditions are given by Asad et al.

After the 10 s reaction, the filter was removed and the melt was cooled down. Each filter type was immersed once in an individual steel melt giving four different batches, each treated with the different filter. One steel batch was only oxidized and deoxidized without any filter immersion and taken as a reference one. After solidification, each batch was hot isostatically pressed (HIP) to remove closed shrinkage porosity. Specimens for fatigue, tensile, and fracture toughness tests as well as polished sections were manufactured from the HIPed steel blocks. Each type of specimen/section was distributed uniformly within the steel block volume to avoid a possible influence of local heterogeneities.

2.2. Thermochemical Treatment

After turning and grinding (average particle size \(\approx 10 \mu\text{m}\), fatigue specimens were austenitized and quenched without any filter immersion and taken as a reference one. After solidification, each batch was hot isostatically pressed (HIP) to remove closed shrinkage porosity. Specimens for fatigue, tensile, and fracture toughness tests as well as polished sections were manufactured from the HIPed steel blocks. Each type of specimen/section was distributed uniformly within the steel block volume to avoid a possible influence of local heterogeneities.
preferentially occurs at the surface.\cite{4} Thus, not the largest inclusion would initiate a crack. Therefore, specimens underwent plasma nitriding to protect the surface from unwanted crack initiation, so that only the NMIs size would play a role in internal fatigue crack initiation. The two-step nitriding procedure (1 h at 420 °C and 2 h at 570 °C) was developed for the given material and geometry of specimens for the optimal distribution of residual stresses in the surface and low tempering of the core.\cite{24} Residual stresses and hardness were controlled on the section of a fatigue specimen from each steel batch. Vickers hardness test was implemented linearly along the diameter of the cross section at the gauge (ø 4 mm) and the side (ø 14 mm) parts of a specimen on the mirror-finished surface. The hardness profile along the gauge section was determined by the means of Vickers microhardness HV0.1 measurements. The core hardness was determined by HV10. Residual stresses were measured by X-ray diffraction (XRD) using Cr Kα radiation (wavelength $\lambda = 0.23$ nm) and the (211) reflection. The residual stress profile along the gauge section was measured at different depths below the surface with the help of electropolishing.\cite{24}

The specimens for quasi-static testing were austenitized after machining at 840 °C for 20 min in vacuum and quenched by a high-pressure gas stream of He. This quenching procedure corresponds to quenching in oil. Finally, the specimens were tempered at 560 °C for 1 h in N2 atmosphere.

2.3. Fatigue Testing

Fatigue tests for all steel batches (Table 1) were performed using the USFT equipment by BOKU (Vienna, Austria). The geometry of the cylinder-shaped specimens with the gauge diameter and length of 4 and 10 mm, respectively, was calculated to have a resonant frequency of $\approx 19.5$ kHz by the methods of computer simulation. The details on the USFT specimen geometry and stress–strain distribution are given in the study by Kreweth et al.\cite{25} USFT was carried out under symmetrical compression–tension load ($R = -1$) at room temperature in pulse-pause mode under constant compressed air cooling.\cite{24} The temperature of specimen’s gauges was controlled by in situ thermography measurement with infrared (IR) camera and kept at 20 ± 2 °C.\cite{25} Fatigue failure of specimen was defined as the amount of cycles $N_f$ at which the strain amplitude remained below 85% of the set level for more than 100 ms. Fatigue cracks were opened manually by initiating a brittle final fracture after cooling in liquid nitrogen. Specimens that reached 106 cycles without a failure were considered as the runouts. It is worth mentioning that these runout specimens were not taken for cycling at higher loading amplitudes to avoid uncertainties on the real fatigue life estimation.

2.4. Quasi-Static Testing

The strength and deformation properties are investigated for all batches defined in Table 1. The tensile tests at quasi-static loading rates ($4 \times 10^{-4}$ s$^{-1}$) were performed at different temperatures (20, −40 °C). The specimen geometry B5 × 25 according to DIN 50125 was applied.\cite{26} Yield strength, YS, and ultimate tensile strength, UTS, were determined. The failed specimens were analyzed in terms of the reduction of area, RA, and the curvature of necking. From the true stress $\sigma_t$, which was corrected for the stress state according to Bridgman,\cite{27} and the true strain $\varepsilon_t$, the dissipated energy up to fracture $E_{diss}$ was calculated as follow

$$E_{diss} = \int_0^{\varepsilon_t} \sigma_t \mathrm{d}\varepsilon_t + \frac{(\varepsilon_f - \varepsilon_{tu}) (\sigma_{tu} + \sigma_u)}{2}$$  \hspace{1cm} (1)

In Equation (1), the indices $u$ and $f$ refer to the point of highest force, i.e., the UTS, and to the point of fracture, respectively. The dissipated energy was then normalized by UTS at the respective temperature (NDE). Details on the calculation were described recently.\cite{28}

In addition to the strength and deformation properties, the fracture toughness of the steels was studied at temperatures of 20 and −40 °C. The batches “+ACS5” (only at 20 °C), “+A,” “+Nano,” and “+CA6” were analyzed. The stable crack growth was described by the J integral as a function of stable crack extension ($\Delta a$) according to ISO 12135.\cite{29} The engineering point of crack initiation, $J_{o,2BL}$, as well as the point of physical crack initiation, $J_{o}$, were measured.

2.5. Fractographic Analysis

Fracture surfaces of all cyclically and quasi-statically tested specimens were analyzed with respect to NMI promoting the fracture process. This was performed using field-emission SEM by Tescan (Czech Republic) in secondary electron (SE) and backscattered electron (BSE) contrast. In addition, the chemical composition of every fatigue crack-initiating NMI was analyzed by energy dispersive X-ray spectroscopy (EDS). In addition, the fracture surfaces of the fracture mechanics specimens were characterized with respect to the stretch zone width SZW. This analysis was performed in accordance with ISO 12135.\cite{29} Experimental details can be found elsewhere.\cite{30} With this information, the point of physical crack initiation $J_{o}$ was determined.

2.6. NonMetallic Inclusions Analysis

Metallographic sections were cut from the steel blocks near to each fatigue specimen for a better correlation of size distribution analysis by sectioning and fatigue testing. From each metallographic section, the area of mirror-finished 100 mm$^2$ was scanned. The total size of analyzed section area was 5000 mm$^2$ by OM and 1000 mm$^2$ by SEM systems.

OM investigations were performed on GX51 microscope with built-in camera XC-10 by Olympus. With the help of a special software “Analysis 5 Particle Inspector” (hereinafter referred to as API), size, orientation, Feret diameters, and location of each inclusion were analyzed automatically. Only NMIs with the maximal Feret diameter $Feret_{max} > 2 \mu m$ were counted. Inclusion detection was based on the light contrast difference between an inclusion and the matrix. Inclusions with $Feret_{max} > 4 \mu m$ were manually checked for errors (e.g., dust, scratches). As the inclusion analysis revealed no significant amount of aggregated (interconnected) inclusion networks, NMIs were
considered as single ones. Cluster analysis of NMIs in 42CrMo4 is given in the study by Seleznev et al.\cite{31}

SEM investigation was performed with the help of automated particle analysis system FEI-ASPEX PSEM eXpress (hereinafter referred to as ASPEX). Inclusion detection was based on the BSE contrast difference between an inclusion and the matrix. The chemical composition of NMIs with $F_{\text{eret}} > 1 \mu m$ was determined by EDS analysis, enabling further chemical classification of inclusions.

In addition to the metallographic analysis of sections, the chemical extraction of NMIs was performed to reveal possible complex 3D morphology. The details of the chemical extraction of steel are described, e.g., in the work by Gleinig et al.\cite{16}

3. Results

3.1. General Material Properties

As it can be seen on orientation maps (Figure 1a,d), the microstructure of quenched and annealed specimens is a tempered martensite, which consists of martensitic needles and some traces of austenite ($\leq 1\%$, not visible on the graphs). The microstructure of a typical quasi-static specimen (Figure 1a) is similar to the one of fatigue specimens (Figure 1d). Due to some minor differences in the annealing procedures (Section 2.2), hardness values slightly differ: 330–340 HV and 310–320 HV in specimens for quasi-static and fatigue testing, respectively.

A significant difference can be found between the NMI morphology of 42CrMo4 steel obtained by filter immersion test in casting simulator (Figure 1b) and commercially available industrial trial casting (Figure 1c).\cite{32} The steel from the casting simulator, which is the object of analysis of the present paper, contains a significant amount of plate-like inclusions even without a filter immersion. Cutting of such plate-shaped NMIs gives linear shapes on metallography sections (Figure 1b). For this reason, NMIs sizes in metallography were estimated as $F_{\text{eret}}$ instead of square root of area to avoid underestimation of their real sizes in 3D.

The typical profile of microhardness HV 0.1 values along the fatigue specimens gauge section (Figure 1e, black line) shows an excellent agreement with the core hardness HV10 values, which lie in the range of 310–320 HV for all fatigue specimens within the investigated steel batches. The residual stress $\sigma_\tau$ profile (Figure 1e, blue line) measured up to the depth of 0.7 mm shows the significant values of compressive stress up to 500 MPa near the surface, which is sufficient for protecting the surface from crack initiation.

3.2. Morphology, Chemical Composition, and Size Distribution of Nonmetallic Inclusions

The chemical extraction of NMIs from the investigated 42CrMo4 steel after treatment in the steel casting simulator reveals different inclusions morphologies (Figure 2, upper row, obtained with SEM imaging) and chemical compositions (Figure 2, lower row, obtained by EDS). Practically, all of the found inclusions are oxides. Manganese sulfides were found only as a part of multiphase NMIs (Figure 2d) due to the dissolution of pure MnS.
inclusions in extraction acid. Homogeneous NMIs consisted of relatively small (≤10 μm) polyhedral particles in the case of both complex (Si + Ca, Al + Si) and single (Si) oxides (Figure 2a). Plate-like NMIs observed on cross sections (Figure 1b) were found to consist only of aluminum oxide (Figure 2a). Multiphase NMIs with globular (Figure 2b) and polyhedral (Figure 2c,d) shapes were found to consist of the oxide phases of different elements (Al, Si, Ti, Si, Mg) and the rare presence of MnS phase (Figure 2d).

It was observed that most of the extracted NMIs are less than 10 μm in diameter, which is below the conditional limit of fatigue crack initiation.[2] Plate-shaped alumina inclusions are the only potential crack initiators due to their large size. Quasi-static mechanical properties are also influenced by the NMI population. However, contrary to fatigue strength, not only the largest NMI size, but also the inclusion's density has to be considered.

Quantitative size and chemical composition distributions were calculated with API and ASPEX systems, respectively. In agreement with the extraction results, around 100% of NMIs detected by EDS from cross sections are oxides, whereas the traces of MnS are negligible. The oxides can be divided into two general groups of pure alumina (Al₂O₃) and other mixed oxides (Figure 3a). The total inclusion density (in terms of number of inclusions per unit area) was found to change after filter immersion. Coatings “+A” and “+ACS” increase the NMI density, whereas coatings “+Nano” and “+CA6” reduce it. Herewith, all the filter coatings reduce the population of alumina NMIs and increase the population of other oxides (Figure 3b). In terms of reduction of total oxide (11–8 mm⁻²) and alumina (11–2 mm⁻²) NMIs population, hexaluminate coating (“+CA6”) shows the most promising results.

NMIs size distribution after API sectional analysis is shown in terms of number of inclusions per mm² versus logarithm of Feret_max (Figure 3c). The reference steel batch (“Ref,” black line) shows the distribution of inclusions without any filter immersion, according to Section 2.1. Here, inclusions up to the size of 10 μm remain on the same level of areal density of around 1 mm⁻². With an increase in the NMIs size, the areal density drops continuously down to 0.01 mm⁻² for 40 μm in size. Immersion of filters with “ACS” and “+Nano” coatings does not change this trend significantly (red and orange lines, respectively). The size distribution of “+A” steel batch (blue line) follows the reference one down to the Feret_max of 6 μm and increases from 1 to 10 mm⁻² for inclusions smaller than 6 μm. The most interesting change in NMI size distribution was observed after “+CA6” immersion test (green line). The population of relatively small inclusions with size range of 4–20 μm is reduced significantly, by almost an order of magnitude (from 1 to nearly 0.1 mm⁻²) for 6–8 μm sizes. At the same time, the size distribution in the range of large NMIs (the distribution “tail”) shows almost double increase in Feret_max (from 50 to around 100 μm) at 0.01 mm⁻² areal density (inset on Figure 3c). This has a negative influence on the fatigue limit which is shown further in Section 3.4.

3.3. Tensile Strength, Deformability, and Energy Dissipation

Figure 4 shows the effect of filter functional coating on strength, deformability, and energy dissipation at temperatures of 20 and −40 °C. It was observed that YS and UTS slightly depend on the filter functional coating. The lowest strength was found for the batch with the AC5-coated filter. At 20 °C, the immersion of the filter with “+Nano” coating led to the highest strength. At −40 °C, the highest strength was achieved when no filter was used. However, the “+CA6” batch has comparable properties.
At both temperatures, the deformability in terms of reduction of area, RA, was highest for the steel treated with the CA6-coated filter. Both strength and deformability parameters correlate with the ability to dissipate energy. As the strength varies slightly between the different steel batches, the deformability is mainly related to the dissipated energy up to fracture. The effect of the strength increase with decreasing temperature was minimized by normalizing the dissipated energy by UTS. It seems that the highest deformability in the “+CA6” batch in comparison to the others is due to the lowest total NMI population (Figure 3). The possible explanations of this effect are discussed in Section 4.

3.4. Fatigue Limit

The results of the fatigue tests of 42CrMo4 steel batches are presented in Figure 5. Each point on the stress versus number of cycles to failure (S–N) diagram (Figure 5a) indicates one of the three possibilities: fatigue failure without (fully colored circles) or with (half-colored circles) fine granular area (FGA).
Figure 5. Results of USFT under symmetrical conditions ($R = -1$). Stress versus cycles to failure graph (S–N diagram) illustrates a set of points (a), corresponding to specimens fatigue failures (circles) or run outs (triangles). Full- and half-colored circles (a) denote the absence or presence of FGA on the fracture surface, respectively. The sizes of fatigue crack-initiating inclusions are measured from every fatigue fracture surface (b) and used as the maximum size inclusions for the calculation of the fatigue limit $\sigma_{wC}$ (c) according to Matsumoto approach. Data points coloring (a–c) represent the steel batches according to the inset (a) and the legend in Table 1.

3.5. Quasi-Static Fracture Toughness

The difference of fracture toughness between the treated steel batches was estimated in terms of $J_{\text{i}}$ integral related to the material resistance to stable crack growth. The physical $J_i$ and engineering $J_{0.2\text{BL}}$ points of crack initiation were measured at temperatures of 20 and $-40^\circ$C (left and right parts of Figure 6, respectively). As it can be seen, the measurements of reference state and “+ACS” at $-40^\circ$C are not given due to a lack of testing material. Nevertheless, it is enough to reveal the most important correlation between fracture toughness and total NMI population density, with the maximum for “+A” and minimum for “+CA6” (Figure 3a).

The fracture toughness of the steel batches tested at 20°C shows almost the same level. Conversely, at $-40^\circ$C, the batch treated with the CA6-coated filter showed the highest mean engineering resistance $J_{0.2\text{BL}}$ to crack initiation, although the scatter was relatively high (Figure 6, error bars). The batch treated with “+Nano” filter coating exhibits higher values of fracture toughness (both $J_{0.2\text{BL}}$ and $J_i$) at low temperature in comparison to “+A.”

Although at room temperature, the fracture toughness shows no significant difference between tested steel batches, the low
temperature fracture tests reveal some difference in mean $J_{0.2BL}$ which is higher for “+CA6” and lower for “+A.” A similar trend can be seen for the deformability of “+A” and “+CA6” at both testing temperatures (Figure 4b). The difference of values of these two quasi-static mechanical properties within the observed steel batches can be related to the difference in their total NMI population density: the lower the inclusions’ amount, the higher the deformability and fracture toughness.

3.6. Fractography

Figure 7 shows images of crack surfaces after tensile (upper row), fatigue (middle row), and fracture toughness (lower row) tests. For a clear distinction between the given mechanical tests, the photographic images of the corresponding specimens are introduced (Figure 7, left column). The indication of the investigated crack surfaces is given by red circles. All presented fractography results are the examples of typical fracture behavior at room temperature observed within the test series. For all loading conditions, the most noticeable feature was the presence of plate-like NMIs (Figure 7, right column). EDS-based chemical analysis showed that all of the observed plate-shaped NMIs consist of alumina (insets on Figure 7d,h). Unlike in section analysis (Figure 1b), plate-shaped NMIs were fully opened by a propagating crack even being tilted to the main crack plain.

The tensile tests resulted in a dimpled fracture surface at room temperature (Figure 7a-c), which indicates a ductile character of crack propagation. Voids initiated from NMIs include large alumina plates (Figure 7d). However, in contrast to fatigue, the size of these plates seems to play a minor role for deformability, as the highest values of RA and NDE are obtained from “+CA6” batch (Figure 4b) even at $-40 \, ^\circ C$, where the crack surface is less ductile. This could be due to the smallest NMI population density in “+CA6”, as voids initiate from all the NMIs regardless of their size.

Fracture surfaces after USFT show morphology typical for internal crack initiation due to nitriding. From the point of initiation, a crack was growing in a penny-shaped manner, forming the so-called fisheye (FiE, marked with a circle in Figure 7f). After a FiE reached the nitrided layer, negative residual stresses blocked its further propagation to the surface and a crack continued to grow internally until the loading stop condition, described in Section 2.3. The FiE is covered with radial ridges that converge to the center (Figure 7g). A closer look at the crack initiation center reveals plate-shaped alumina (Figure 7h), surrounded by FGA if fatigue life was above 100 cycles. SEM analysis showed that all fatigue cracks were initiated by plate-shaped alumina. The size of these plates varies in the range of 50–150 $\mu m$, whereas the thickness is not greater than 1 $\mu m$. Quantitative fractography measurements by confocal laser scanning microscopy showed that plate inclusions could be treated as regular NMIs in terms of their influence on fatigue. This means that their stress intensity factor can be calculated from their projection area on the stress plane, which is achievable directly from the SEM images. The distance of crack-initiating NMIs from the specimen surface was found to be in the range of 0.5–1.7 mm depth, whereas the peak distribution was at 0.8–0.9 mm depth. Therefore, influence of the residual stresses on cracks initiation was negligible. Thus, a drop of fatigue toughness in “+CA6” in comparison to the other steel batches (Figure 5a) is due to a bigger size of the largest NMIs (Figure 5b).

Prior to the fracture toughness tests, a pre-crack (PC) was introduced according to ISO 12135 (Figure 7i, j). During the test, the initially sharp PC blunted and formed the stress zone. At a critical point, the crack extended in a stable manner by ductile fracture. This critical point is known as the physical crack initiation. Analogously to the tensile tests, voids nucleated around NMIs (Figure 7k). In some cases, large plate-like NMIs were found to form agglomerates, forcing a crack path to deflect and include their plane (Figure 7k). Despite the promotion of void formation, large NMI plates (Figure 7l) and their agglomerates did not decrease the fracture toughness. $J$ integral parameters of “+CA6” batch are either on the same level or higher (Figure 6) than the parameters for other batches with smaller NMI sizes.

4. Discussion

The combination of sections analysis with the mechanical tests under fatigue and quasi-static loading conditions allowed a detailed evaluation of the NMI population features and their influence on mechanical properties. The most pronounced difference in properties from the reference state “+Ref” was observed in the steel after the immersion of the filter with hexaluminate coating “+CA6.” Fatigue tests revealed that the “+CA6” coating led to the formation of the largest NMIs and, consequently, to the lowest fatigue life (Figure 5a) and fatigue limit (Figure 5c) calculated by the Murakami formula (Equation (1)). On the other hand, the same “+CA6” coating led to the highest deformability in terms of RA and NDE (Figure 4), which correlates with the lowest NMI population density both in total (Figure 3a) and in the range of small sizes (4–20 $\mu m$, Figure 3c). The improvement in deformability during necking due to NMIs’ density reduction could be explained in terms of mean spacing between inclusions. Less NMIs initiate
less voids, which then coalesce slower due to their lower number and increase in the mean distance between them.

This dual effect on cyclic and static properties means that aiming to reduce only the total amount of inclusions could be insufficient for improving the material properties by refining techniques. Even the section analysis could not be enough for a good evaluation of the NMIs with a complex shape (Figure 1). Thus, one of the most reliable ways to determine the location and shape of the biggest NMIs is fatigue testing. For the observed steel batches, the maximum NMI size found by section analysis is two times smaller than the actual one, found by fatigue testing (Figure 3c, 5b). Such a discrepancy is typically reported for other metals as well.\textsuperscript{[35]}

The observed fatigue life reduction is caused only by plate-shaped alumina inclusions. Yet, no correlation between the size of alumina plates and quasi-static mechanical properties (toughness, deformability, fracture toughness) was found. Other oxides do not influence the fatigue even though their amount is increased after filters’ immersion (Figure 3b). However, the appearance of plate alumina inclusions was not due to filter immersion, but to the treatment in the steel casting simulator prior to testing.\textsuperscript{[13]} Industrial casting deals with the different morphologies of alumina, which appear due to deoxidation and reoxidation in steel.\textsuperscript{[17]} The formation of alumina plates (especially of 100 μm scale) is reported quite rare and seems to be possible only in special circumstances within certain local concentrations of Al and O ($\leq 0.05\%$ and $\leq 0.005\%$, respectively).\textsuperscript{[36]} These local concentrations are in a good agreement with the total concentrations in the investigated steel batches (Table 1). However, no correlation was

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**Figure 7.** Results of analysis of fracture surfaces after a–d) tensile, e–h) fatigue, and i–l) fracture toughness tests. The magnification of images increases from left to right, showing fracture from the macro- to microscale: view of full specimens with the fractured part highlighted by a, e, i) red circle, b, f, j) fracture full view, c, g, k) fracture microstructure, and d, h, l) fracture-causing inclusions. The chemical composition of inclusions at the insets (d, h) is obtained by EDS. The abbreviations on fracture views denote pre-crack (PC), stable crack extension (SCE, j, k), and fisheye (FiE, f).
found between the morphology and size of the detected alumina plates and the Al concentration, which varies by almost one order of magnitude ($<0.001–0.007\%$). The formation of plate-shaped alumina is a separate question, which lies beyond the focus of this work. Yet, it is fair to conclude that a control of the NMIs morphology is necessary, because even the reduction of the total impurities content (Al and O contents for alumina NMIs) does not guarantee an improvement of mechanical properties.

The main purpose of the filter immersion tests in the casting simulator was to study the reactions of a steel melt with the filter coating. As we could see for the filter with “+CA6” coating, after the immersion the following parameters were reduced: 1) the total number of NMIs to 70% of the reference value; 2) the number of alumina NMIs to 20% of the reference value; 3) the number of other oxides to 7% of the reference value; 4) the number of relatively small inclusions (4–20 $\mu$m); 5) the number of relatively big inclusions ($\geq 30\,\mu$m).

The exact numbers in brackets are given for the “+CA6” immersion test, whereas similar effects are known to arise in other types of steel refining by reactive materials. The main mechanisms during active/reactive filtering are shown in the schematic diagram in Figure 8a and are as follows: 1) formation of CO gas bubbles and Al (suboxides) due to carbothermic reaction between C and $\text{Al}_2\text{O}_3$ from the filter coating in the presence of the steel melt. CO bubbles float up, which gives an effect similar to Ar purging. These bubbles can entrap inclusions and transport them from the steel melt to the slag. CO purging also induces NMIs collision which can promote agglomeration. Some evidence of agglomeration is known even for the nanometer-sized NMIs (Figure 8d). However, the correlation between NMIs agglomeration and reactive refinement is not clear yet and is a subject for further research.

2) In situ deposition of a secondary oxide layer between the steel melt and the reactive coating. In case of alumina-based coating, the secondary layer also consists of Al and O which is taken from the steel melt (corundum, Figure 8b). When this layer is dense, it isolates the filter from the steel melt and blocks any further CO production. 3) The newly formed secondary oxide layer is highly active: it adsorbs NMIs from the steel melt on its surface. 4) Deep bed filtration (active cleaning) takes place when the steel melt flows through the filter. NMIs, which are large enough to stick to the filter wall, will be collected, whereas small NMIs will pass through.

The filtering of molten steel by ceramic filters is well known. However, active filtration is efficient in removing only large inclusions, leaving smaller ones in the steel melt. As was shown on the example of “+CA6” coating, reactive filters are efficient in the reduction of relatively small inclusions. Thus, the implementation of an active + reactive filter system for two-stage steel cleaning is believed to remove most of the NMIs. An investigation on the active + reactive filter system will be presented in future work.

5. Conclusions

In the present paper, metal melt filters with different reactive coatings were studied. The effect of different functional filter coatings on the remaining detrimental NMIs was investigated. The inclusion distribution with respect to size and chemical composition was determined by means of optical microscopy and...
scanning electron microscopy. Furthermore, the mechanical properties of the treated steel were evaluated by ultrasonic fatigue tests, quasi-static tensile tests, and quasi-static fracture toughness tests. The main results can be summarized as follows: 1) The application of the CA6-coated filter led to the most significant changes in NMI population in comparison to the reference state by reducing: The total number of NMIs to 70% of the reference value; the number of alumina NMIs to 20% of the reference value; the number of other oxides to 7% of the reference value; the number of relatively small inclusions (4–20 μm); the number of relatively big inclusions (≥30 μm); the maximum inclusions size range (from 30–100 μm to 90–150 μm). 2) The chemical analysis of NMIs revealed mostly pure alumina and other oxides (e.g., calcia, silica, and mullite). All tested filter coatings reduced the total amount of alumina and increased the total amount of other oxides. 3) Plate-like alumina inclusions were the most detrimental in terms of fatigue strength and were responsible in the initiation of 100% of fatigue cracks. Treatment with the CA6-coated filter resulted in the lowest fatigue limit due to the largest NMIs. 4) Detrimental effect of plate-like alumina on quasi-static loading was not found. 5) The total NMIs density seems to affect the tensile deformability and energy dissipation. Thus, the relatively low number of NMIs after treatment with the CA6-coated filter resulted in higher deformability and higher ability to dissipate energy. 6) The resistance to the onset of stable crack growth was not significantly affected by the type of the filter coating. 7) The implementation of an active + reactive filter system for two-stage steel cleaning is believed to be an effective method for removal of both small and large NMIs from the steel.

Acknowledgements

The authors thank the German Research Foundation (DFG) for the financial support of the investigations within the Collaborative Research Center 920, subprojects A01, C01, C04, and C05. The authors acknowledge the support of Dr. Steffen Dudczig for performing the filter immersion tests, Ms. Birgit Witschel for quantitative optical microscopic analysis, and Mr. Johannes Gleinig for inclusions analysis.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

AISI 4142, ceramic foam filters, fracture toughness, nonmetallic inclusions, very high cycle fatigue

Received: May 10, 2019
Revised: July 5, 2019
Published online: August 14, 2019

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