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## Correlation between local chemical information, microstructure and recrystallisation: how Sn addition influences texture development in single-phase Fe alloys

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#### Abstract

Sn addition is known to affect the recrystallisation texture development of  $\alpha$ -Fe. To date, even though the texture effect of Sn has been experimentally verified, although indirectly, the underlying mechanism has not been studied. In the present study, we focus on the static recrystallisation behaviour of Fe-Si-Sn alloys to elucidate the texture preferences assessing for the effect of Sn solute addition. The experimental approach is based on sequential electron back-scatter diffraction (EBSD) texture analysis complemented with a site-specific chemical analysis of grain boundaries by atom probe tomography (APT). It is directly observed through our experiments that boundary segregation phenomena, which occur at the onset and later stages of recrystallisation, provide crucial understanding of texture development. Especially in the presence of solute, the orientation dependence of the energy stored by deformation largely determines the course of the subsequent recrystallisation. In the Fe-Si-Sn alloy, recrystallisation at the highest stored energy grains is hindered. As a consequence of segregation, {001}(110) orientations recrystallise at later stages of the process, forming predominantly grains with orientations of the {h11}(1/h 1 2) fibre. These grains are suggested to form following an oriented nucleation mechanism, while their growth could be interpreted with Ibe and Lücke's theory of selective growth of 26.5°(110) boundaries in bcc metals.

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*Keywords:* recrystallisation, grain boundary segregation, solute addition, texture.

#### 1. Introduction

During the thermomechanical processing of metals desired properties in the final product can be obtained via controlling recrystallisation and the related annealing phenomena [1]. The process of recrystallisation involves the softening of plastically deformed metal by restoring its ductility. Recrystallisation is a structural transformation that progresses heterogeneously and typically its kinetics can be separated in two main stages: nucleation and growth [2].

The term nucleation in recrystallisation typically refers to <sup>26</sup> the incubation and growth stages of a subgrain already present <sup>27</sup> in the deformed microstructure [1]. Considering nucleation, the <sup>28</sup> subgrain model theory was introduced by Cahn [3], Beck [4] <sup>29</sup> and more recently Humphreys [5] in studies that shed light on <sup>30</sup> the nuclei selectivity of recrystallisation. According to these, <sup>31</sup>

for a subgrain to become a successful nucleus of recrystallisation, there are two main criteria that must be fulfilled; the nucleus needs to acquire a critical size and a boundary mobility advantage [6]. In some cases the size threshold may be reached during annealing by the process of rotation and coalescence of two or more subgrains as proposed by Hu [7]. Nevertheless, it is also essential to have or to attain a misorientation with its environment to achieve the necessary mobility [6]. Several theories have been developed on the basis of relative grain boundary mobility. Some boundaries with low  $\Sigma$  coincidence site lattice (CSL), are known to have higher mobility than grain boundaries (GBs) with higher  $\Sigma$  or general GBs. Impurities that remain in solid solution are known to drastically alter grain boundary mobility and can impact remarkably grain boundary migration-controlled phenomena, such as recrystallisation and phase transformations [8, 9, 10]. Solute segregation at crystal interfaces largely determines the migration rates of the different boundaries. Aust and Rutter have measured the effect of small addition of Sn on the migration rate in pure Pb and observed that special boundaries are less affected by solute, although overall all boundaries underwent a reduced migration rate [11]. It has been suggested that low  $\Sigma$  GB mobility becomes predominant over the one of general GBs [12], as solute

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segregation enhances the mobility contrast in the microstructure <sup>96</sup>
 [13, 14, 15].

Another important aspect of recrystallisation is the orienta- 98 41 tion dependence of stored energy [16]. In general by increasing 99 42 the cold rolling strain in bcc alloys, the orientations present in100 43 the cold-rolled microstructure rotate towards the  $\{111\}\langle uvw\rangle_{,101}$ 44  $\gamma$ -fibre component and  $\{hkl\}\langle 110\rangle$  ( $\alpha$ -fibre) component. In this 102 45 case,  $\{111\}\langle uvw \rangle$  ( $\gamma$ -fibre grains) are able to accommodate rel-103 46 atively higher amount of strain and have a higher Taylor factor<sub>104</sub> 47 than other grain orientations. As the driving force for recrys-105 48 tallisation is proportional to the stored energy of deformation, 106 49 parent grain orientations are expected to influence the recrys-107 50 tallisation preferences. An established orientation dependence108 51 of the stored energy in subgrain coarsening was shown elabo-109 52 rately by Dillamore [16] and later by Hutchinson [17] in low<sub>110</sub> 53 carbon steels. The study of Hutchinson [17] suggested that nu-111 54 cleation proceeds in an orientation-dependent manner. It has112 55 also been shown [18] that nucleation in heavily rolled (75-85%)113 56 steels takes place primarily in orientations of high Taylor factor<sub>114</sub> 57 (i.e. high stored energy). In the study of recrystallisation it is115 58 therefore essential to evaluate the effect of parent grain structure116 59 on the recrystallisation process. 117 60

Recrystallisation texture strongly depends on the thermo-118 61 mechanical process parameters i.e. annealing conditions and<sub>119</sub> 62 deformation mode, while its formation and orientation depen-120 63 dence to the deformed structure has attracted significant re-121 64 search interest throughout the years [19]. Two major theories<sub>122</sub> 65 have prevailed to explain the origin of the recrystallisation tex-123 66 ture: (i) by its nucleation preferences; oriented nucleation, (ii)<sub>124</sub> 67 the observed texture is claimed to be a result of the preferred<sub>125</sub> 68 growth of some grains; oriented growth. Nevertheless, to date, 126 69 both theories are widely used to explain recrystallisation obser-70 vations and in practice both can contribute to texture formation. 71 As Dillamore and Katoh observed [20], a successful recrystalli-72 sation theory must explain both the origin and orientation de-128 73 pendence of the recrystallised grain with the deformed matrix. 74

In the case of pure Fe and low carbon steels, the recrystalli-75 sation texture is typically characterized by a strong  $\{111\}\langle 112\rangle$ 76 (component of the  $\gamma$ -fibre) texture with the peak intensity be-77 ing dependent on the cold rolling reduction [21]. Meanwhile,<sup>132</sup> 78 the  $\{411\}\langle 148\rangle$  component is sometimes present in the recrys-79 tallisation texture, as it has been reported for interstitial free<sup>134</sup> 80 and electrical steels [22, 23]. The origin of this texture com-81 ponent shares some attention in literature, yet the mechanism 82 of its formation is still under debate, as direct observations are 83 generally lacking. The formation of  $\{111\}\langle uvw \rangle$  texture can be 84 influenced by the thermomechanical process parameters and the 139 85 chemical composition of the alloy [22]. As far as the chemical<sup>140</sup> 86 composition is concerned, the recrystallisation texture can be<sup>141</sup> 87 affected by the presence of elements such as P, Se, Sb, Sn etc. 88 142 even in low concentrations [24]. In several occasions the in-89 tentional addition of solutes is accompanied by a weakening of143 90 the  $\gamma$ -fibre [25, 26, 27]. In particular for Sn addition, previous<sup>144</sup> 91 research showed that Sn reduced the  $\{111\}\langle uvw \rangle$  texture com-<sup>145</sup> 92 ponent intensity, while the intensity of  $\{110\}\langle 001\rangle$  increased in<sup>146</sup> 93 the recrystallisation texture [28, 29]. The mechanism via which<sup>147</sup> 94 Sn is thought to affect texture formation is often attributed to

the enhanced mobility of some CSL interfaces related to segregation phenomena. However, these texture observations with solute addition were performed in a post-mortem fashion, as samples were characterized at the end of the processing and with no association to Sn segregation state.

Latest technological developments in characterisation methods now allow for the first time to correlate orientation preferences with local chemical composition information. Correlative site-specific characterisation is undoubtedly opening more avenues for deeper understanding in materials science, which means that we have an unprecedented capability of analyzing complex physical mechanisms in recrystallisation texture development. Here, we study the effect of Sn addition on the static recrystallisation of Fe-Si model alloys. Si is a common alloying element in steel and is usually added for the fabrication of core materials in magnetic flux carrying devices [30]. In a previous study, the Sn solute effect on the recovery prior and concurrent to recrystallisation was investigated illustrating the retardation in the softening of Fe-Si-Sn alloys [31]. Nevertheless, the underlying mechanism, with which Sn modifies the crystallographic texture formation during recrystallisation, is not yet fully understood. Materials were studied in the as cold deformed state and as recrystallisation initiated and advanced. A sequential measurement procedure using the EBSD technique was used to monitor the evolution of the microstructure and to assess the role of parent grain orientation in recrystallisation texture development. In parallel, site-specific chemical analysis with atom probe tomography (APT) was performed to measure solute segregation at grain boundaries. Chemical information was used to support the crystallographic orientation observations and to explain the effect of Sn solute addition.

#### 2. Experimental

#### 2.1. Materials

A binary Fe- 3 wt.% Si (Fe- 5.8 at.% Si) and a ternary Fe- 3 wt.% Si- 0.1wt.% Sn (Fe- 5.8 at.% Si- 0.045 at.% Sn) were lab produced with maximum impurity level of 150 ppm. Si is added to stabilize  $\alpha$ -Fe and produce a single-phase ferritic alloy in order to study recrystallisation only under the solute effect, both Si and Sn concentrations are within the solid solubility in  $\alpha$ -Fe. Therefore, the key factor on recrystallisation development will be solute segregation. The Fe-Si binary alloy provided a reference material to isolate the silicon contribution, assessing merely the Sn effect in the microstructure evolution of iron. Both materials were similarly processed, hot rolled, annealed and then cold rolled at a Von Mises true strain of about  $\epsilon = 2$  as described elsewhere [31].

#### 2.2. EBSD measurements

Subsequent static recrystallisation was monitored in a sequential manner with EBSD starting from the as cold deformed state and after fast annealing treatments, following the microstructure evolution in a region of interest (ROI) with a sequence of EBSD scans after each annealing step.

The sequential EBSD experiments were performed in the 148 Centre of Materials Forming (CEMEF), Mines ParisTech, in 149 Sophia Antipolis, using a heating stage that has been developed 150 in CEMEF and set up in a FEI XL30 scanning electron mi-151 croscope (SEM) equipped with a TSL EBSD acquisition sys-152 tem. Both binary and ternary alloys in the as deformed state, 153 were subjected to an interrupted annealing performed at 700° C 154 within the SEM chamber and characterized by EBSD to mon-155 itor the recrystallisation evolution. Fig. 1 displays a typical 156 heat treatment during the sequential EBSD experiment as mea-157 sured by the thermocouple welded on the sample surface. The 158 heating device has a thin Ta foil on which the sample is point-159 welded, shown schematically in Fig. 1a. An electrical current 160 goes through the Ta foil so that both the Ta foil and the sam-161 ple (with dimensions of about  $3 \times 4 \times 0.2 \text{ mm}^3$ ) are heated by 162 Joule effect. This particular experimental setup enables very 163 fast heating and cooling rates up to about  $100^{\circ}$  C s<sup>-1</sup>, while 164 temperature is regulated by two thermocouples welded close 165 to the region of analysis. More details on the experimental 166 setup can be found elsewhere [32]. EBSD mappings were per-167 formed at room temperature after each annealing cycle, so that 168 the microstructure does not evolve during the acquisition time, 169 which would be a serious drawback for fast-kinetic phenom-170 ena. EBSD maps were obtained in a selected region of interest 171 in the initial condition and after each heat treatment cycle. For 172 efficiency purposes, given the repeatability and duration of the 2013 173 experiment, the EBSD mappings were performed with a step<sub>204</sub> 174 size of  $2\mu m$  to be completed within less than 60 minutes cov-175 ering an area of  $400 \,\mu\text{m} \times 600 \,\mu\text{m}$ . Additional EBSD measure-176 ments were performed on the initial state of the materials using 177 a Zeiss Gemini 500 field-emission gun scanning electron mi-178 croscope (FEG-SEM) equipped with an EDAX Hikari Super<sup>207</sup> 179 acquisition system. These EBSD scans were performed using  $a^{^{208}}$ 180 finer step size down to  $0.05 \,\mu m$ . Crystal orientations throughout 181 this manuscript are expressed using the conventional orienta-210 182 tion representation shown as Orientation Distribution Functions<sup>211</sup> 183 (ODFs) within the Euler space ( $\phi_1$ ,  $\Phi$ ,  $\phi_2 = 0.90^\circ$ , due to the or-<sup>212</sup> 184 thorhombic sample-crystal symmetry of the bcc structure) and<sup>213</sup> 185 214 their intensity is expressed in multiple random density [33]. 186 215

#### 187 2.3. Heat-treatment in SEM chamber

Starting from the as deformed state, samples were heat 188 218 treated inside the SEM on the heating stage. The heat treatment 189 applied to both binary and ternary was similar, but longer accu-220 190 mulative holding time was used in the case of the ternary due<sub>221</sub> 191 to slower recrystallisation kinetics [34]. During the 1st heating<sub>222</sub> 192 step, a heating rate of 35° C/s was followed, while the follow-223 193 ing heating steps were performed with the maximum  $\operatorname{possible}_{\scriptscriptstyle 224}$ 194 heating rate ~ 100° C/s. Short isothermal holdings at 700°  $C_{_{225}}$ 195 were applied and holding time was gradually increased once the<sub>226</sub> 196 kinetics became slower, i.e. during the second half of the trans-227 197 formation, and until almost full recrystallisation was achieved.228 198 To analyse the kinetics of recrystallisation, the non-isothermal<sub>229</sub> 199 heat-treatments followed in this study (including heating/cooling<sub>30</sub> 200 sections) Fig. 1b, needed to be converted into isothermal. For<sub>231</sub> 201 this purpose, the approach of Whittaker and Sellars was used<sub>232</sub> 202



Figure 1: a) Sample configuration tested with EBSD and b) heat-treatment scheme followed during the sequential EBSD experiment in the SEM.

to convert the heating time into an equivalent isothermal time [35].

#### 2.4. Data treatment

All EBSD data post-processing was performed with the OIM Analysis platform. The low-to-high angle boundary transition was set at 15° [36], hence low-angle boundaries (LAGBs) were defined for  $2^{\circ} \le \theta \le 15^{\circ}$  and high-angle boundaries (HAGBs) for  $\theta \ge 15^{\circ}$ . Recrystallised grains were defined as grains bounded by HAGBs and by imposing the following additional misorientation criteria; a recrystallised grain must have a low internal average misorientation (GOS  $\leq 1.8^{\circ}$ ). The areas that did not fulfill the aforementioned criteria, were considered as nonrecrystallised (non-RX) or recovering microstructure and mainly consisted of a subgrain structure (minimum angle for subgrain definition,  $\theta_c \ge 2^\circ$ ). To include all newly formed grains, after a thermal treatment, a grain definition at the subgrain level was necessary 5° and a more relaxed orientation spread threshold of  $\leq 2.5^{\circ}$ ; such data segmentation will be referred to as all new grains. In other words, these grains are products of both recovery and recrystallisation excluding the not recrystallised regions of the microstructure.

Besides the recrystallisation state, further analysis of the EBSD data was conducted by segmenting the microstructure also as a function of parent grain orientation, i.e. as deformed prior grains. For this purpose, the image quality (IQ), orientation information from the IPF maps and prior grain boundary locations were combined. In the as deformed state (Fig. 2 and Fig. 4 initial), bright and dark regions represent high IQ corresponding to low stored energy (LSE) regions and low IQ corresponding to high stored energy (HSE) regions, respectively.

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For simplicity, in the present study we consider the as deformed 233 state to be binary consisting of HSE and LSE regions only. HSE 234 regions include the main components of the  $\gamma$ -fibre {111}(110), 235  $\{111\}\langle 112\rangle$  and the higher dislocation density components at  $\Phi$ 236 > 35° of the  $\alpha$ -fibre. LSE regions include the orientations of the 237  $\alpha$ -fibre centered at the {001}(110) component and its vicinity 238 (at  $\Phi = 0^{\circ}$ ). LSE also include the {112}(110) of the  $\alpha$ -fibre [37] 239 as this orientation is also known to recrystallise at a later stage 240 of static recrystallisation. The reader is referred to the Supple-241 mentary Material for more details on the data segmentation. 242

#### 243 2.5. Atom probe characterisation

From the EBSD scanned areas, regions of interest were se-244 lected and prepared into tips for APT. Site-specific sample prepa-245 ration was performed in a FEI Helios NanoLab 600 Dual Beam 246 focused ion beam (FIB - SEM), following the lift-out technique 247 as described elsewhere [38]. Atom probe experiments were 248 conducted in a LEAP 3000XHR instrument with a detector ef-249 ficiency of 0.37. All measurements were performed in voltage 250 mode, with the sample base temperature and the pulse fraction 251 set at 70 K and 20%, respectively. The APT data were analysed 252 with the IVAS software after calibration of the reconstruction 253 parameters [39]. Finally, segregation at the interfaces was quan-254 tified using the well-established method of the Gibbsian excess 255 number,  $\Gamma_i^{GB}$  [40]. Such analysis yields a value of concentration 256 insensitive to the measurement resolution and inclination angle 257 of the interface with respect to the axis of analysis [41]. 258

#### 259 3. Results

#### 260 3.1. The as deformed state

In our previous study, the deformed state ( $\epsilon = 2$ ), at the RD-<sub>291</sub> 261 ND plane, was analysed in-depth using EBSD measurements<sub>292</sub> 262 and performing a dislocation density calculation based on mis-293 263 orientation data [31]. Overall, the analysis of the deformed<sub>294</sub> 264 state, on the basis of deformation features, fraction of main tex-295 265 ture components and stored energy analysis from EBSD [31], 266 suggested that the addition of Sn neither induced noticeable<sub>296</sub> 267 solid solution strengthening nor alter the known stored energy<sub>297</sub> 268 sequence (in agreement with the work of Dillamore et al.  $[16])_{208}$ 269 and that the binary and ternary alloys have very similar as de-299 270 formed microstructures at this stage. Although in the annealed 271 state prior to cold rolling, the ternary alloy had a finer mi-272 crostructure as a result of Sn addition, the grain sizes after inter-273 mediate annealing were 145 and 104  $\mu$ m for binary and ternary<sub>303</sub> 274 alloys, respectively. 275

The regions of interest investigated during the experiment 276 were chosen to be as representative as possible of the overall as  $_{306}$ 277 deformed state. In the initial scans, as deformed  $\{111\}\langle uvw\rangle$  as  $_{307}$ 278 well as  $\{001\}\langle 110 \rangle$  oriented grains were included in the area of  $_{_{308}}$ 279 analysis. Their respective volume fraction in the as deformed<sub>309</sub> 280 state, may not represent accurately the global one but the focus 281 is to study their behavior and interaction during recrystallisa-311 282 tion. In both materials large non-indexed areas are also included 283 in the EBSD maps (black zones in Fig. 4). Fig. 2 shows a high<sub>313</sub> 284 resolution EBSD overview of the as deformed microstructure 285



Figure 2: An overview of the as deformed (initial) microstructure in the ternary alloy measured by EBSD. a-b) in the ND-RD section: the ND-IPF map a)through the entire sample thickness and b) at a ROI between different prior grain orientations. c-f) in RD-TD section, at a similar position in the ND axis as in the sequential experiment: c) the ND-IPF mapping, d) the IQ image, e) the 1st neighbor KAM mapping and f) the average KAM measured in the high stored energy (HSE) and the low stored energy (LSE) regions.

in RD-TD section, acquired at a similar position in the ND axis as the one in the sequential EBSD experiment. It can be seen that non-indexation can be correlated to higher amount of local dislocation density and is orientation dependant. For example, the kernel average misorientation (KAM) of the as deformed  $\{111\}\langle 112 \rangle$  orientation is seen to be more than double that of the  $\{001\}\langle 110 \rangle$  orientation (Fig. 2e-f). Hence, it is suggested that these highly deformed, non-indexed regions within the area of analysis in Fig. 4 have a near  $\{111\}\langle uvw \rangle$  orientation, something that is also confirmed by their neighboring indexed orientations.

#### 3.2. Global recrystallisation behaviour

The global recrystallisation behaviour in the two alloys, Fe-Si (binary) and Fe-Si-Sn (ternary) has been reported in detail in a previous work [42] (see Supplementary Material). That study focused on the effect of Sn addition on recrystallisation texture analysing several thousands of grains by EBSD. Furthermore, the recrystallisation kinetics in the two alloys was quantified by interrupted dilatometer heat treatments and optical image analysis. Fig. 3 shows the overview of the recrystallisation texture for similar recrystallisation fractions ( $X_V = 0.2, 0.5, 0.8$ ) in the two alloys. It was observed that Sn resulted in a softening of the recrystallisation texture by reducing the intensity of  $\{111\}\langle 112\rangle$ - from x12.9 mrd to x4.0 mrd at  $X_V = 0.2$  (Fig. 3a-b). Whereas the peak intensity in the ternary microstructure was the Goss - $\{110\}\langle 001\rangle$  component, which suggested that shear band nucleation of recrystallisation is less (if at all) affected by Sn. Both the EBSD data and recrystallisation kinetics suggested that the most significant effect of Sn was during nucleation and early

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Figure 3: Micro-texture ODFs at the  $\phi_2 = 45^{\circ}$  section of the Euler space show-<sup>357</sup> ing the evolution of recrystallisation texture at 700° C from large-scale ex situ<sub>358</sub> EBSD measurements for (a,c,e) binary and (b,d,f) ternary alloy at 0.2 - 0.5 - <sup>359</sup> 0.8 fractions recrystallised, respectively. The max. intensities and numbers of RX grains included are given for each ODF plot (more details are provided<sup>360</sup> elsewhere [42]).

stages of recrystallisation [42]. At later stages of recrystallisation, these observations remained and in the case of the ternary alloy other orientations emerged in the recrystallisation texture (Fig. 3d and f). This behaviour will be discussed in detail in the following sections, in view of local, sequential, EBSD measurements and chemical analysis with APT.

#### 3.3. Sequential observation of recrystallisation

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EBSD analysis after sequential annealing at 700° was performed to evaluate the evolution of the microstructure during recrystallisation in the two alloys and the role of the parent grain orientation. Fig. 4 shows the ND- inverse pole figure (IPF) maps as recrystallisation progresses in the two alloys. The IPF maps reveal a clearly different recrystallisation behaviour for the two alloys. The binary alloy was observed to recrystallise rapidly, with small recrystallised grains appearing already after the first heating step (without soaking at  $700^{\circ}$  C, t = 0s). Moreover, recrystallisation was nearly completed after only 10 seconds of accumulative soaking time. On the other hand, the ternary alloy exhibits much slower recrystallisation kinetics, suggesting that Sn retards the onset of recrystallisation. Indeed, first recrystallised grains were observed after more than 10s accumulative time and more than 2000s accumulative time was necessary to complete recrystallisation within the analysed region. Nevertheless, in both alloys recrystallisation initiates in the  $\gamma$ -fibre parent grains (blue / non-indexed areas in Fig. 4), as expected from stored energy considerations.

Fig. 5 shows the evolution in normalised surface area of the main annealing texture components of all the new grains in the two alloys - as recrystallisation initiates and advances to almost completion (i.e. relative recrystallisation fraction,  $X_V^{rel}$ = 0.84 - 0.93). In the recrystallisation texture of the binary alloy, the  $\{111\}\langle 112\rangle$  component appears early in the process and becomes dominant at the later stages of recrystallisation. The  $\{111\}\langle 112\rangle$  component grows to almost twice the area fraction of the other orientations, and a substantial growth occurs at about  $X_{V}^{rel} = 0.7$ . The texture evolution for the ternary alloy (Fig. 5b), shows in contrast to the binary microstructure, that the  $\{111\}\langle 112\rangle$  component stays at rather low values of area fraction during recrystallisation. Moreover, an increase of the  $\{100\}\langle 120\rangle$ ,  $\{411\}\langle 148\rangle$  and  $\{111\}\langle 110\rangle$  orientations is observed. These observations relate to the parent grain orientations and stage of recrystallisation, and this will be further discussed in the following sections. The experiment shows that the parent grain orientation plays a crucial role in the recrystallisation sequence, while their kinetics is quite diverse. Hence, a segmentation of the microstructure as a function of parent grain orientation is relevant. The length scale and orientation data of the sequential EBSD experiment permit to analyse the recrystallisation kinetics at the parent grain scale and this subject will be considered in the following sections.

#### 364 3.3.1. Recrystallisation as a function of parent grain

The segmentation of the as deformed microstructure as a function of stored energy enables the evaluation of the individual kinetics within either the HSE or the LSE regions of the



Figure 4: Sequential EBSD ND-IPF + GBs mappings of recrystallisation during annealing in a) binary and b) ternary alloy. The relative recrystallisation fractions are given at each step, lines represent GB misorientations of  $8 - 65^{\circ}$ .

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microstructure using the Johnson-Mehl-Avrami-Kolmogorov383 368 (JMAK) analysis [43]. Fig. 6 shows the individual recrystallisa-384 369 tion kinetics in the HSE and LSE regions for the binary (Fig. 6a)385 370 and for the ternary alloy (Fig. 6b), respectively. The parameters<sub>386</sub> 371 of the JMAK model fitted to the EBSD data are listed in Table387 372 1. The overall JMAK exponent in the kinetic law for the binary<sub>388</sub> 373 alloy is close to unity. For the ternary alloy, the JMAK exponent389 374 is different depending on the prior grain stored energy. HSE re-390 375 gions were found to recrystallise with an exponent n of about<sub>391</sub> 376 0.6, while the LSE regions recrystallise later but much faster,  $n_{392}$ 377 = 1.2.378

Table 1: The JMAK kinetic model parameters from EBSD. The parameters of <sup>394</sup> the recrystallisation kinetics are only given for fractions  $0.1 < X_V < 0.9$  [44].<sup>395</sup> The  $t_{50}$  [1] is given in seconds.

$\begin{array}{c ccccccccccccccccccccccccccccccccccc$			Overall	HSE	LSE
$\frac{1}{10000000000000000000000000000000000$	Binary	п	1.0	-	
n 0.7 0.6 1.2		$t_{50}$	2.4		
	Ternary	п	0.7	0.6	1.2
$t_{50}$ 565 81 1080		$t_{50}$	565	81	1080

Fig. 7a shows the growth rates of recrystallised grains within<sub>404</sub> each region. Black arrows in Fig. 7a designate the moment<sub>405</sub> when recrystallisation proceeds in the LSE regions. The binary<sub>406</sub> alloy recrystallises rapidly and the recrystallised grains grow<sub>407</sub> fast. Overall the kinetics cannot be separated since HSE regions recrystallise and migrate into the LSE regions. Hence, the kinetics of the binary alloy represents that of recrystallised grains from the HSE regions. On the contrary, the ternary alloy recrystallises in a more individual manner; HSE and LSE regions recrystallise separately and their kinetics are independent. The grains recrystallising within LSE regions nucleate at a later stage and their growth is faster. The overall kinetics show a mean behaviour resulting from the contribution from both HSE and LSE regions.

#### 3.3.2. Texture formation as a function of parent grain

Regarding the texture formation, Fig. 7b-c shows the texture ODFs of the recrystallised grains within the HSE (Fig. 7 b.1-b.2) and LSE regions (Fig. 7 c.1-c.2) at the end of recrystallisation for the two alloys – for a limited number of grains, N = 280 - 630. Circles at Fig. 7a designate where the texture plots correspond in the growth process. In both alloys, recrystallisation initiates in the HSE regions. The high local disorientation within these regions suggest high dislocation density that provides the necessary driving force to initiate recrystallisation [2]. Indeed, in the binary alloy recrystallisation was observed to initiate and grow dominantly from the HSE regions (Fig. 4a). Moreover, as it was seen earlier, the recrystallisation texture evolution showed that the  $\{111\}\langle 112 \rangle$  component increased significantly towards the latter stages of recrystallisation, at about



Figure 5: The evolution in normalised surface area of the main annealing texture components during sequential heat-treatment at  $700^{\circ}$  C for a) binary (open points) and b) ternary alloy (solid points) taking into account all the new grains. Arrows on the plots designate the stage when recrystallisation proceeds in the low-stored energy regions.



Figure 6: Microstructural experimental data for recrystallisation from sequen-<sub>427</sub> tial EBSD the recrystallisation kinetics for the two alloys after separating the deformed state into HSE (blue squares) and LSE (red circles) regions for a) binary (open points) and b) ternary (solid points) alloys.



Figure 7: Evolution of microstructure in the high (HSE) and low stored energy (LSE) regions respectively, a) the growth of new grains with annealing time for binary (open points) and ternary (solid points), arrows indicate the stage when recrystallisation proceeds in the LSE regions. The micro-texture ODF plots at the  $\phi_2 = 45^\circ$  section of the Euler space of the new grains at the end of recrystallisation as a function of parent grain: for binary b.1) HSE (N=631), c.1) LSE (N=386) and for ternary b.2) HSE (N=328), c.2) LSE (N=282), respectively.

 $X_V^{rel} = 0.7$ . This corresponds to a stage at which recrystallisation is completed in the HSE regions and is expanding into the LSE regions, arrows on Fig. 5 indicate the point where recrystallisation proceeds in the LSE regions. Recrystallised grains from the HSE grow into the LSE consuming these regions. This can be observed in Region 'A' of Fig. 4a where some grains from a recrystallised HSE region are migrating into an adjacent LSE region as recrystallisation progresses in the binary alloy. This behaviour shows characteristics of strain induced boundary migration (SIBM) mechanism as will be discussed more extensively in a following section. Thus, in the binary alloy a similar texture is observed after recrystallisation in both HSE and LSE regions as shown in Fig. 7b.1 and c.1. This leads to a peak at the {111}(112) component of the  $\gamma$  -fibre with an area fraction almost twice that of the other orientations.

Fig. 7c shows the texture evolution as a function of parent orientation for the ternary alloy. Despite that the HSE regions also recrystallise first, the onset of recrystallisation occurred after a prolonged incubation period and kinetics is particularly sluggish in these regions. In general, the  $\{111\}\langle 112 \rangle$ orientation in the recrystallisation texture of the ternary alloy stays at low values. Recrystallised grains from HSE regions

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show a weak  $\gamma$ -fibre with the peak intensity shared between the {111} $\langle 110 \rangle$ , {411} $\langle 148 \rangle$  and slightly deviated {001} $\langle 120 \rangle$  components (Fig. 7b.2).

At a later stage, LSE regions were observed to recrystallise 433 independently. The point when recrystallisation started in LSE 434 regions is indicated by the arrow on Fig. 5b. The recrystalli-435 sation of LSE in the ternary alloy was accompanied by an in-436 crease in the  $\{001\}\langle 120\rangle$ ,  $\{411\}\langle 148\rangle$  and  $\{111\}\langle 110\rangle$  orienta-437 tions, as shown in Fig. 5b. Towards the end of recrystallisation, 438 recrystallised grains from the LSE regions (Fig. 7c.2) show a 439 strong maximum on the  $\theta$ -fibre and near the  $\{h11\}\langle 1/h \mid 2\rangle$  fi-440 bre first reported by Homma et al. [45], with a peak intensity 441 of  $\times$  8.5 random near {611}(16.12). The texture illustrates that 442 the recrystallisation of HSE and LSE regions in the ternary al-443 loy occurs independently as the kinetics have shown (Fig. 6b). 444 The ternary recrystallisation texture shows thus, a clear texture 445 inheritance from the parent grain orientation. 446

In conclusion, the segmentation of the EBSD dataset into
HSE and LSE regions revealed that due to Sn addition the two
alloys behave very differently in the progress of recrystallisation with respect to the strain heterogeneity of the as deformed
state.

#### 452 3.4. Segregation

#### 453 3.4.1. Prior grain boundaries

As a starting point, solute segregation was investigated at 454 the prior GBs (PGBs). The material after intermediate anneal-455 ing, i.e. before cold rolling, was preferred for characterization 456 since the cold deformed state was found to be quite brittle for 495457 APT analysis. Fig. 8a shows the EBSD ROI from which  $a_{496}^{7}$ 458 random GB,  $\theta = 28^{\circ}[123]$  was selected for chemical analy-459 sis, views of the APT volume containing the PGB of interest 460 are shown in Fig. 8b. In the APT volume, all Sn atoms (green 489 461 spheres) are shown and for simplicity the Fe matrix atoms are  $\frac{1}{490}$ 462 only partially shown. The concentration profile for Sn across 463 the GB is given in Fig. 8c. APT analysis revealed a segregation $_{492}$ 464 amount of Sn of 2.3 at.nm<sup>-2</sup>. Additionally, interstitial C and 465 B atoms were observed to segregate at this boundary, although  $A_{404}$ 466 both were at residual levels in the nominal composition of these  $_{495}$ 467 alloys. Segregation of C and B is considered to occur also in the 468 binary grain boundaries and is thought to exert negligible solute 469 drag effects. Nonetheless, site competition and potential inter-470 action of these elements with Sn atoms cannot be dismissed,  $\frac{1}{499}$ 471 especially in the case of C and Sn [24]. 472 500

#### 473 3.4.2. Onset of recrystallisation

After the thorough examination of the recrystallisation ori-474 entation preferences and kinetics with the addition of Sn, it is 504 475 evident that the HSE regions are of great importance. There-476 fore, the substructure of a HSE region of the ternary alloy was<sub>506</sub> 477 further investigated. The EBSD characterization of the ND-RD<sub>507</sub> 478 section (Fig. 9), revealed that the HSE region had a finer intra-479 granular cell-structure of subgrains, while LSE regions were 509 480 considerably coarser. If the subgrain model of Cahn [3] is as-481 sumed to describe the mechanism of nucleation of recrystallised 482 grains, a nucleus derives from a successful subgrain that grows<sub>512</sub> 483



Figure 8: The ternary alloy prior to cold rolling a) EBSD ND-IPF mapping of the microstructure (black lines representing GBs with  $15-65^{\circ}$  misorientation), b) different views of the APT volume with Sn segregation (green spheres) at the prior GB, c) the concentration profile for Sn perpendicular to the interface.

during the incubation period. Markedly, these regions of the microstructure are very important to understand the nucleation mechanisms. Fig. 9a shows the EBSD GAM map at the onset of recrystallisation in the ternary material ( $X_V \sim 0.07$ ). The fine subgrain structure inside the deformed grain can be observed, along with the presence of a recrystallised grain having very low intra-granular misorientation variation. The recrystallised grain represents the successful nucleation process from subgrain growth selectivity. Subgrain boundaries (SGBs) at the onset of recrystallisation were analysed with APT after isothermal holding equivalent to 1.6 seconds at 700° C. Fig. 9a-b shows the EBSD ND-IPF maps of the material at different magnifications, and the mint green circle shows the region of subgrain structure from which a tip for APT was prepared by FIB-SEM. The orientation of the parent grain area is near  $\{111\}\langle 110\rangle$  along the  $\gamma$ -fibre. The APT analysis revealed the presence of a triple junction between three SGBs, in Fig. 9c, Sn atoms are represented as green spheres. Remarkably high Sn segregation was observed to decorate the SGBs already at this very early stage of recrystallisation. Besides Sn atoms, no other element was found to meaningfully segregate at the SGBs. Si concentration shows some variation at the boundary but the enrichment factor (concentration at boundary over the concentration at the bulk) is near unity and is thus considered negligible.

The segregation profiles of Sn across the different SGBs are shown in Fig. 10a. The segregation was quantified with the Gibbsian excess number (Fig. 10b) and was found to be different for the three SGBs: 2 interfaces are found to have similar amount of Sn, SGB1 0.91 and SGB3 0.94 at.nm<sup>-2</sup> while the

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Figure 9: The ternary alloy at the early stages of recrystallisation ( $X_V = 0.07$ ) analysed in EBSD before APT analysis. EBSD maps shown at various magnifications with the region of interest where the APT tips were taken from (mint-green circle) a) GAM+IQ map, b) IQ map and orientation colored map showing the grains with {111} and {100} parallel to ND, c) different views of the APT volume with Sn segregation (green spheres) at the subgrain boundaries.



Figure 10: (a) the concentration profiles perpendicular to the interface for the  $^{533}$ subgrain boundaries and (b) integral profiles that yield the Gibbsian excess534 number at each SGB. 535

other one, SGB2, has a lower amount of Sn of 0.59 at.nm<sup>-2</sup>. 513 The Sn segregation is probably varying depending on subgrain 514 angle, as Read and Shockley observed for low-angle bound-515 aries [46]. Additionally, a segregation misorientation depen-516 dence has been previously reported for Sn for a binary Fe-Sn 517 alloy [47]. Unfortunately, the exact misorientations associated 518 to the SGBs were not accessible as the ROI was very small and 519 it was challenging to control the exact location during FIB sam-520 ple preparation. Nevertheless, by segmenting the EBSD map to 521 include only the not recrystallised structure of that particular 522 prior grain, it was found that nearly 75% of the SGBs had a 523 misorientation  $2.5^{\circ} \le \theta < 9.5^{\circ}$ , while the mean value was,  $\theta$ 524 =  $8.2^{\circ}$ . Hence, statistically it is reasonable to assume that the 525 analysed SGBs may have a misorientation near these values. 526

Maruyama et al. [48] have also observed Nb solutes at subgrain boundaries of a Fe-Nb alloy at the stage of recrystallisation incubation and the excess number is comparable at about 529 1.19 at.nm<sup>-2</sup>.

#### 4. Discussion

The as deformed microstructure constitutes the initial state and will define the path for the thermally activated phenomena that will take place during annealing. Thus, it is important to have a good understanding of the microstructure at this

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stage. The alloys were cold rolled at a true strain of about  $\epsilon = 593$ 536 2. At these large levels of strain, the deformed microstructure is594 537 known to be comprised of a well-defined network of subgrains595 538 [2]. The subgrain structure is formed by dislocation accumula-596 539 tion arranged in a cell like structure in the grain interior [49].597 540 The dislocation arrangement within the deformed grains differs598 541 with grain orientation as the energy of deformation is known to599 542 be stored heterogeneously across different orientations [16]. It<sub>600</sub> 543 is suggested that subsequent nucleation of recrystallisation pro-601 544 ceeds in an orientation-dependent manner [17]: for cold rolled602 545 metals, recrystallisation takes place primarily in orientations603 546 with high stored energy [18]. More specifically, according to<sub>604</sub> 547 the Taylor factor criterion, the  $\{110\}\langle 110\rangle$  orientation (at  $\Phi =$ 548 90° of the  $\alpha$ -fibre) is expected to demonstrate the highest ca-605 549 pacity to store the energy of deformation [16]. However, such 550 orientation is scarcely observed in the cold rolled microstruc-607 551 ture of conventionally processed single-phase  $\alpha$ -Fe [50]. In<sub>608</sub> 552 cold rolled  $\alpha$ -Fe the most abundant, high Taylor factor orien-609 553 tations, are  $\{111\}\langle uvw\rangle$  ( $\gamma$ -fibre) orientations. During rolling, 610 554 the  $\{111\}\langle uvw \rangle$  grains exhibit higher slip activity than the other<sub>611</sub> 555 orientations and as a result store higher amount of energy dur-612 556 ing deformation. In particular, the  $\{111\}\langle 112\rangle$  oriented grains<sub>613</sub> 557 are prone to form shear bands during deformation [51]. On the614 558 contrary, the  $\{001\}\langle 110\rangle$  (at  $\Phi = 0^{\circ}$  of the  $\alpha$ -fibre) component615 559 are the lowest dislocation density grains [16]. Therefore, during616 560 recrystallisation of the cold deformed materials, the  $\{111\}\langle uvw\rangle_{617}$ 561 grains usually recrystallise readily, while the {001}(110) orien-618 562 tations are more reluctant to recrystallise [22]. 619 563

The sequential EBSD experiments showed that the solute620 564 addition altered markedly the recrystallisation preferences. In621 565 brief, it was observed that during annealing, HSE regions re-622 566 crystallise promptly without Sn addition however, when Sn is623 567 added, the onset of recrystallisation is greatly retarded in these 624 568 regions of the as deformed microstructure. A distinct difference625 569 observed in the ternary alloy is that LSE regions recrystallise626 570 independently at a later stage of the process. As a result, the re-627 571 crystallized texture is weaker compared to the binary alloy and628 572 the presence of some  $\{001\}\langle uvw \rangle$  texture components are also<sub>629</sub> 573 observed. 574 630

Overall, the texture observations made from the sequen-631 575 tial EBSD experiment are in accordance with ex situ observa-632 576 tions [34], where the analysis of several thousands recrystal-633 577 lized grains showed that Sn is affecting significantly the recrys-634 578 tallisation texture from the onset. Additionally, the overall re-635 579 crystallisation kinetics of the two alloys, shown in the global636 580 study [34], displayed the same retardation of recrystallisation637 581 nucleation with the addition of Sn shown in the present work. In638 582 section 3.2.1, we saw that the recrystallisation kinetics of HSE639 583 and LSE regions in the ternary alloy showed a distinct change640 584 in the JMAK exponent. 641 585

In an earlier study [52], we have reported the segregation<sup>642</sup> of Sn at half-recrystallisation in migrating GBs. Here, the fo<sup>-643</sup> cus is shifted to the very beginning of recrystallisation and the<sup>644</sup> initial segregation state. The retardation of recrystallisation in<sup>645</sup> the HSE regions, in the presence of Sn, was identified to be a key point to understand the solute effect on texture evolution. Hence, such regions were selected to perform a site-specific chemical analysis. Sn segregation was measured with APT at SGBs of the non-RX structure within a substructured HSE region (within a  $\{111\}\langle 110\rangle$  oriented region). The APT analysis revealed a remarkably high Sn segregation decorating the SGBs at the onset of recrystallisation, shown in Fig. 9. This experimental finding of Sn segregation at this early stage of recrystallisation, is of great importance for understanding how the nucleation of recrystallisation may be affected by segregation. All segregation data measured at different stages of recrystallisation, reported here and in previous works, are summarised in Table 2 together with crystallographic information. First, we should consider the diffusion processes involved.

#### 4.1. Diffusion of Sn to subgrain boundaries

The segregation results showed that a swift annealing of 1.6 seconds at 700° C was sufficient for Sn solutes to segregate at SGBs of the non-RX structure with levels of 0.6-1 at.nm<sup>-2</sup>. Hence, this raises the question of the diffusion mechanism that transported the Sn atoms so efficiently to the SGBs. SGBs consist of dislocation accumulation, initially they are created due to the large strain applied during cold deformation; at this stage, Sn atoms should be mainly in solid solution in the grain interior prior to the recrystallisation annealing and the SGBs should not be solute enriched. Sn atoms are considered to be in solid solution in the grain interior at t = 0. Nevertheless, during annealing below the recrystallisation temperature, the dislocations at the SGBs tend to re-arrange via dislocation glide and climb. The authors have previously discussed that Sn solutes could already affect dislocation motion and retard the subgrain formation during recovery processes [31].

Making these assumptions (no segregation of Sn at SGBs at t = 0), kinetics can be calculated tacking into account diffusion from the grains to the SGBs and assuming local equilibrium between grain and SGBs. Indeed, it is usually assumed that during segregation, a local equilibrium between the GB and its vicinity is attained since the time required to equilibrate the GB should be faster compared to the time needed for diffusion from the grain interior that controls the concentration at the GB vicinity. Moreover, the temperature dependence of grain boundary segregation can be described by the Langmuir-McLean isotherm. To account for these conditions and following the analysis of Fournier Dit Chabert et al. [53], the segregation kinetics of Sn was calculated by finite difference simulation in one dimension using: (i) the second Fick law for the bulk diffusion in the grain, and (ii) the Langmuir- McLean isotherm to calculate the local equilibrium between the GB and its vicinity. A segregation energy at random GBs at 700° C,  $\Delta G_{Sn}^0 = -51.3$  kJmol<sup>-1</sup> and a site saturation  $x_{Sn}^0 = 1/3$  were used as reported for Sn in  $\alpha$ -Fe in [54]. The segregation kinetics were calculated for a semi-finite volume with a half interface and half grain [53]. A GB width,  $\delta$ =0.5 nm was selected as in the study of Inoue et al. [55]. For the purpose of the present simulation the subgrain width is assumed to be comparable to grain boundary width [42]. The grain with a half grain size<sup>1</sup>, x = 300 nm was divided in ele-

<sup>&</sup>lt;sup>1</sup>the half grain size, x = 300 nm was selected as this value corresponds to the half of the mean subgrain size,  $d_0/2$  experimentally observed in the ROI.

Table 2: Data on segregation of Sn during different stages of recrystallisation. Grain boundary crystallographic information as obtained from the EBSD orientation mappings and the values of interfacial segregation for Sn,  $\Gamma_{Sn}^{GB}$  as analysed with APT.

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	Adjacent grains	Euler Angles $(\phi_1, \Phi, \phi_2)$	Boundary	Grain boundary, $\theta$ [h,k,l]	Interfacial excess of Sn (at. nm <sup>-2</sup> )	Ref.
Prior to	Grain 1	31°, 52°, 43°	Grain 1/ Grain 2	random GB, 28°[123]	$2.3 \pm 0.1$	This work
cold rolling	Grain 2	28°, 26°, 63°				
Recrystallisation incubation	Subgrain 1		SGB 1		$0.91 \pm 0.09$	
	Subgrain 2	matrix: 0°, 55°, 45°	SGB 2	-	$0.59 \pm 0.04$	This work
	Subgrain 3		SGB 3		$0.94 \pm 0.08$	
Recrystallisation migration	RX Grain 1	13°, 32°, 40°	Grain 1/ Non-RX	random GB, 48.2°[4 -1 2]	$0.60 \pm 0.08$	
	RX Grain 2	49°, 27°, 21°	Grain 2/ Non-RX	random GB, 27.5°[-4 3 2]	$0.41 \pm 0.06$	[52]
	Non-RX matrix	30°, 55°, 44°				



Figure 11: Kinetics of Sn segregation at the grain boundaries compared to the segret experimental APT values of Sn at subgrain boundaries and changing the dif- $_{684}$  fusion coefficient from  $D_{bulk}$  [56] to an efficient diffusion coefficient  $D_{eff} = D_{3}_{685}$  [57].

ments with a size equal to half the GB size, dx = 0.25 nm. Diffusion in the grain was solved by explicit finite difference using 647 the second Fick law in one dimension. At each time step of the 648 diffusion with the finite difference simulation, the Sn concentra-649 tions in the SGB and in the grain element in contact with the  $\mathrm{GB}^{^{689}}$ 650 were equilibrated using the Langmuir- McLean equation and 690 651 the conservation of mater. Note that under these assumptions, 652 the kinetics of segregation is mainly controlled by the diffusion 653 in the grain but depends also on the equilibrium segregation that  $^{\rm 693}$ 654 694 fix the boundary condition at the grain/SGB interface. 655

<sup>656</sup> Fig. 11 shows the segregation kinetics of Sn segregation to <sup>695</sup> the GBs (or SGBs) from the APT segregation data and the cal-<sup>696</sup> culated segregation kinetics. In the case of pure bulk diffusion <sup>697</sup> of Sn atoms from the bulk to the SGBs (Fig. 11 solid line), the <sup>698</sup> diffusivity of Sn in  $\alpha$ -Fe below the Curie transition temperature <sup>699</sup> was reported by Torres et al. [56] as:

$$D_{bulk} = 3.5 \cdot 10^{-2} \exp(\frac{-275000}{RT})$$
 m<sup>2</sup>s<sup>-1</sup> (1)<sup>702</sup><sub>703</sub>

using the  $D = D_{bulk}$ , the predicted segregation amount cor-<sup>704</sup> relates well with the APT measured values of Sn segregation.<sup>705</sup> In other words, the diffusion length,  $\sqrt{D_{bulk} \cdot t}$  for pure bulk diffusion could explain the Sn atom transport to the SGBs at the onset of recrystallisation.

Nevertheless, acknowledging the large amount of cold strain applied, the short annealing time and the fact that the APT analysis was performed in the interior of a high dislocation density region, it is reasonable to assume that dislocations may have a contribution to the overall diffusivity of Sn atoms. Diffusion through dislocation pipes is considered to have a diffusion coefficient of up to three orders of magnitude, in the case of pure pipe diffusion, higher compared to bulk diffusion [58, 59, 16]. In the current circumstances, it is more appropriate to think that the transport of atoms is controlled by an effective diffusion coefficient,  $D_{eff}$  which incorporates both the contributions from pipe and bulk diffusion [60]. Klinger et al. [57] and more recently Divinski et al. [61], have given a theoretical basis for the  $D_{eff}$  variance with the individual contributions. For instance, dislocation enhanced diffusion has been discussed for its importance in the diffusion of Nb in  $\alpha$ -Fe [61].

In these works, the bulk diffusion coefficient in the classical model from Fisher of GB diffusion, is substituted by  $g_d D_d$ , where  $g_d$  is the volume fraction of sites belonging to dislocation pipes. Then  $D_{eff}$  is expressed as:

$$D_{eff} = D_3 = g_d D_d + (1 - g_d) D_{bulk}$$
(2)

 $g_d$  in Eq. 2 is calculated using the dislocation density of the as deformed state for the {111}(110) orientation,  $\rho = 10^{-15} \text{ m}^{-2}$ as reported in a previous study from EBSD data [31]. It is noted that the  $D_3$  diffusion coefficient is an approximation as the dislocation density used here represents the total number of geometrical necessary dislocations (GND) estimated from EBSD data. In principle, the relevant dislocations in the case of efficient diffusion would be the ones within the bulk, subtracting from the total density the dislocations that form the subgrain walls. A detailed assessment of such diffusion mechanism is complex. More segregation data at different annealing times are needed. Additionally, dislocation density data regarding mobile dislocations from the grain/subgrain interior and as a function of crystallographic orientation would be essential.

In comparison to the APT experimental data at the onset of recrystallisation, the dashed line in Fig. 11 shows that segregation kinetics with an efficient diffusion in the  $D_3$ -regime, can be considered to be relevant as well in the present conditions. Nonetheless, further work will be needed in this direction.

#### 706 4.2. Texture evolution with Sn

### 707 4.2.1. Recrystallisation affected by segregation

With respect to the recrystallisation texture development,<sup>764</sup> 708 the observed segregation is also central. The presence of Sn<sup>765</sup> 709 at SGBs can explain the retardation of recrystallisation by de-766 710 creasing the rate of subgrain coarsening due to solute drag pin-<sup>767</sup> 711 ning. The HSE regions should be more affected by Sn segrega-768 712 tion at the SGBs, since hard Taylor factor orientations tend to<sup>769</sup> 713 form a finer substructure during deformation and to contain a<sup>770</sup> 714 higher density of intra-granular defects. 715

We have observed that the binary alloy recrystallises mainly<sup>772</sup> 716 from the grain interior of the  $\{111\}\langle uvw \rangle$  prior grains produc-<sup>773</sup> 717 ing a strong  $\{111\}\langle uvw \rangle$  texture. Essentially, the recrystallisa-<sup>774</sup> 718 tion mechanism in these regions is the one introduced by Cahn<sup>775</sup> 719 [3]; a successful recrystallisation nucleus is formed during sub-776 720 grain coarsening processes. Several possible mechanisms may777 721 come into play during recrystallisation to assist the subgrain778 722 formation and coarsening [62]: (i) recovery phenomena, re-779 723 arrangement and accumulation of dislocations at the SGBs; (ii)780 724 subgrain coalescence phenomena via migration of low angle781 725 boundaries increasing gradually their misorientation with the<sup>782</sup> 726 matrix. Static recrystallisation of HSE regions, usually pro-783 727 motes a strong  $\{111\}\langle uvw \rangle$  recrystallisation texture well known<sup>784</sup> 728 in low carbon steels [22, 21]. Similarly, in our binary alloy, re-785 729 crystallisation proceeds heterogeneously with the HSE regions786 730 reaching full recrystallisation whilst nucleation is still not initi-787 731 ated in the LSE. Eventually, there is a critical time for which the<sup>788</sup> 732 dislocation density in the LSE regions becomes higher than in789 733 the recrystallised HSE regions. This dislocation density differ-790 734 ence provides the driving force for the recrystallised grains of<sup>791</sup> 735 HSE regions to migrate into the LSE regions. Such recrystalli-792 736 sation process is known as strain induced boundary migration<sup>793</sup> 737 (SIBM) or GB bulging. It occurs as a crystal within a lower<sup>794</sup> 738 dislocation density region is favoured to grow into an adjacent795 739 region with a higher dislocation density [5, 63], once a criti-796 740 cal size and dislocation density difference is acquired [64]. The<sup>797</sup> 741 role of SIBM during static recrystallisation has been previously798 742 reported in the literature [17, 65, 66]. An example of SIBM<sup>799</sup> 743 can be observed in the sequential EBSD mappings of the bi-800 744 nary alloy Fig. 12. The region 'A' shows the recrystallisation<sup>801</sup> 745 between a HSE and a LSE region separated by a prior GB. It<sup>802</sup> 746 is observed that after HSE regions are fully recrystallised, re-803 747 crystallised grains from the HSE regions bulge into the slug-804 748 gish LSE regions - yellow arrows in Fig. 12 indicate grains<sup>805</sup> 749 which bulge into LSE. The majority of grains migrating into ad-806 750 jacent LSE regions had the  $\{111\}\langle uvw\rangle$ , consisting about 75% at<sup>807</sup> 751 t = 2s, 72% at t = 4s, 65.5% at t = 6s of the migrating grains. 752 From these, were often  $\{111\}\langle 112 \rangle$  grains, about 45% at t = 2s,<sup>808</sup> 753

<sup>754</sup> 44% at t = 4s, 38% at t = 6s, respectively. As a consequence, <sup>809</sup> <sup>755</sup> the recrystallisation texture in the binary alloy is dominated by<sup>810</sup> <sup>756</sup> {111}(uvw) texture components, which nucleated faster in the<sup>811</sup> <sup>757</sup> HSE regions and grew to consume adjacent LSE regions (see<sup>812</sup> <sup>758</sup> Supplementary Material). <sup>813</sup>

On the other hand, the recrystallisation with Sn addition is<sup>814</sup>
 seen to be completely different. Segregation at the onset of<sup>815</sup>
 static recrystallisation, at the substructure of the non-RX HSE<sup>816</sup>

regions, is of great importance for understanding how the nucleation is affected by segregation. In the most accepted subgrain boundary model, by Winning et al. [67], the movement of such boundaries occurs through dislocation climb facilitated by vacancies. The physical concept relates to the vacancy motion, which must first escape from the Sn atoms (overcoming the Sn-vacancy binding energy) so that subgrain boundary dislocations can climb [68, 67]. It is suggested that segregated Sn at this stage, will pin subgrain coarsening. Such an effect of Sn in Cu-Sn alloys was assumed also by Cram et al. [68], who modelled and tested the deformation behaviour of the alloys with various Sn contents during dynamic recrystallisation. In consequence, recrystallisation nucleation selectivity, as described by Cahn nucleation model [3] is retarded due to segregation.

Subsequently, as recrystallisation advances, Sn grain boundary segregation is exerting a drag force at the migrating boundaries of recrystallisation retarding the growth of recrystallised grains and the consumption of the non-RX matrix [52]. Hence, nucleation of new grains in the ternary alloy is delayed as evidenced in the number of recrystallised grains at similar  $X_V$  and areas of analysis, Fig. 3. The segregation data in Table 2 show that the amount of segregated Sn at the SGBs is higher than the one measured for later stages of recrystallisation on migrating HAGBs. This observation is somewhat unexpected and is considered to be due either to desegregation of Sn occurs for longer annealing at 700° C or to impurity C atoms and site competition for segregation [24].

Furthermore, SIBM events of  $\{111\}\langle uvw \rangle$  recrystallised grains into LSE regions is less often observed in the ternary alloy. The reason for this observation could be understood, (*i*) as the size criterion for the subgrains and dislocation density difference does not fulfil the Bailey - Hirsch threshold (Suppl. Mat.) and (*ii*) as Sn enrichment at the PGBs would induce a solute drag effect and could potentially limit GB bulging. For the latter to be valid, some Sn segregation should remain at the PGBs during subsequent cold deformation and recrystallisation annealing. This still remains to be experimentally verified. Additionally, the effect of initial grain size (which was finer for the ternary alloy) would also have favoured SIBM effect if solute drag was not acting on the PGBs.

The overall effect of Sn solute segregation at the prior grain boundaries, subgrain boundaries and recrystallisation interfaces experimentally observed (Table 2), contribute in a significant texture softening compared to the binary alloy. The recrystallised grain intensity of  $\{111\}\langle uvw \rangle$  is reduced (nucleation effect), and  $\{h11\}\langle 1/h \ 1 \ 2 \rangle$  intensity increases.

### 4.2.2. Role of parent grain orientation and formation of $\{h11\}\langle 1/h \ 1 \ 2\rangle$ recrystallised orientation

To understand the formation of such texture we must first consider its origin. Deformed grains with  $\{411\}\langle 148 \rangle$  orientation may form within  $\{112\}\langle 110 \rangle$  and  $\{001\}\langle 110 \rangle$  crystals (main components of the LSE regions), where weak lattice curvatures are created at the vicinities of grain boundaries [69].  $\{411\}\langle 148 \rangle$  regions in the deformation texture are often regarded to form after severe rolling reductions (i.e. higher than 90%). At large

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Figure 12: Magnified region of interest 'A' delimited by a white rectangle within Fig. 4a of the binary alloy, showing the interaction of high-stored energy (HSE) and a low-stored energy (LSE) region in the as deformed microstructure as recrystallisation advances at 700° C in the binary Fe-Si alloy. a) ND-IPF mappings, orientation colored maps with IQ: b) for texture components  $\{111\}\langle 112\rangle$  and  $\{111\}\langle 110\rangle$ , c) for texture fibres  $\{111\}\langle uvw\rangle$  and  $\{100\}\langle uvw\rangle$ . High-angle grain boundaries are shown in black and a prior grain boundary is shown with a white dashed line (PGB). At t = 2s yellow arrows indicate recrystallised grains originated in the HSE regions adjacent to the PGB, which migrate into the LSE region during static recrystallisation via stress induced boundary migration (SIBM).



Figure 13: The growth of the most dominant texture components,  $\{111\}\langle 112\rangle_{869}$ in HSE and  $\{411\}\langle 148\rangle$  in LSE for binary (open points) and ternary alloy (solid points). 871

strains  $\{112\}\langle 110 \rangle$  and  $\{001\}\langle 110 \rangle$  regions, near crystal bound-873 817 aries, can locally fragment and form deformation bands near the874 818  $\{411\}\langle 148\rangle$  component [45, 69] or the  $\{311\}\langle 136\rangle$  component as<sup>875</sup> 819 reviewed by Gobernado et al. [70, 71]. These intra-granular re-876 820 gions, can store more energy during deformation than the sur-877 821 rounding  $\{001\}\langle 110\rangle$  parent grain [72] and thus serve as poten-878 822 tial recrystallisation sites within these regions. For the sake of 879 823 clarity, the  $\{411\}\langle 148 \rangle$  orientation ( $\phi_1 = 20^\circ, \Phi = 20^\circ, \phi_2 = 45^\circ$ )880 824 and its vicinity orientations such as the  $\{311\}\langle 136\rangle$ , will be re-881 825 ferred to as the representative component, R component, after882 826 Gobernado et al. except if the precise orientation needs to be883 827 specified [70]. 828 001

Studies on the recrystallisation of IF steels, confirm that885 829 mainly the *R* component is observed after recrystallisation as-886 830 sociated with the LSE regions and particularly the deformed887 831  $\{001\}\langle 110 \rangle$  orientation [22]. Gobernado et al. suggested that 8888 832 the formation of the *R* component, during recrystallisation in a<sup>889</sup> 833 cross-rolled IF steel, is related to oriented nucleation [70], as890 834 it was initially suggested by Homma et al. [45]. Whereas, ori-891 835 ented growth theory has also been applied to explain the appear-892 836 ance of this texture component [73], as it has a  $26.5^{\circ}(110)$  mis-893 837 orientation in respect to as deformed  $\{112\}\langle 110\rangle$  regions present<sup>894</sup> 838 in the cold rolled state. The increased mobility of  $26.5^{\circ}(110)_{895}$ 839 boundaries in bcc metals (Ibe and Lücke model) could assist the896 840 selective growth of orientations that these interfaces bound. In a897 841 recent study using in situ EBSD experiments, Takajo et al. stud-898 842 ied the formation of Cube oriented grains ( $\{001\}\langle 100\rangle$ ), which<sup>899</sup> 843 nucleated in the  $\{411\}\langle 148\rangle$  as deformed regions and grew pref-900 844 erentially [74]. Thus, the sequential aspect of the recrystallisa-901 845 tion evolution presented here, adds a valuable insight on texture902 846 development in LSE regions and its origin. 903 847

The evolution of the individual texture components in the<sup>904</sup> binary and the ternary alloy (Fig. 5), in combination with the<sup>905</sup> mean growth of the recrystallised grains in HSE and LSE re<sup>-906</sup> gions (Fig. 7), indicated a different mechanism during recrys<sup>-907</sup> tallisation. It was observed that the parent grain orientation is<sup>908</sup> decisive, in particular for Sn addition. LSE regions with Sn<sup>909</sup>

were observed to recrystallise independently during the progress 854 of recrystallisation due to the Sn solute drag at the HSE regions. 855 It is interesting to consider the grain growth kinetics of the dom-856 inant orientations in the primarily recrystallised structure as a 857 function of parent grain orientation. Fig. 13 shows the growth 858 rates of the main texture components,  $\{111\}\langle 112 \rangle$  for the HSE 859 and  $\{411\}\langle 148 \rangle$  for the LSE regions of the binary and ternary 860 allovs. 861

In the binary microstructure, both texture components are observed to have similar growth rates of the mean recrystallised grain diameter (Fig. 13). However, as observed before, the overall intensity of the  $\{111\}\langle 112 \rangle$  is the most dominant in the recrystallised microstructure. In the ternary alloy, HSE regions are also the first to recrystallise but overall Sn segregation is considered to retard greatly the nucleation in these regions. Then, at a later stage, LSE regions start to recrystallise to form mainly  $\{411\}\langle 148\rangle$  (and  $\{100\}\langle 120\rangle$ ) grains. The recrystallisation of the LSE regions, is considered to benefit from recovery processes. Indeed, since LSE regions have a relatively lower dislocation density, their coarsening rate should be faster than in the HSE regions as a result of a coarser structure at the as deformed state and a lower recrystallisation nucleation density. In other words, it can be considered that these regions can potentially coarsen more readily during annealing. Duggan and Roberts [75] studied texture evolution after extensive recovery and subsequent recrystallisation in a low carbon steel and they observed a strong  $\{411\}\langle 148\rangle$  texture component in the annealing texture. Essentially, the  $\{411\}\langle 148\rangle$  component grew faster as it nucleated in LSE zones where the nucleation density is lower, thus grain growth competition and impingement are reduced. In contrast, in the HSE zones the high nucleation density results in sooner impingement, which reduces the recrystallised growth in these regions. Fig. 13 demonstrates these microstructural considerations during the sequential recrystallisation.

Fig. 14 focuses on the recrystallisation of the LSE regions in the microstructure of the ternary alloy. The main texture component observed after recrystallisation of these regions was the *R* component; the peak is observed at  $\{611\}\langle 16 12 \rangle$  ( $\phi_1 = 20^\circ$ ,  $\Phi = 13^\circ, \phi_2 = 45^\circ$  this orientation is near the  $\{411\}\langle 148 \rangle (\phi_1 \otimes \phi_2) \rangle$ = 20°,  $\Phi$  = 20°,  $\phi_2$  = 45°) component. It is observed that the R component was one of the first orientations to recrystallise. Its formation occurred mainly at the vicinity of low-angle GBs, designated by red arrows in Fig. 14. Towards the end of recrystallisation, grains of the  $\theta$ -fibre, near the  $\{001\}\langle 120 \rangle$  appear as well (Fig. 14b). Recrystallisation in the LSE regions, may also be assisted by subgrain coalescence phenomena. Such an example can be seen within a region of interest 'B' in Fig. 14a, with recrystallised grains appearing in microbands oriented near the R component. Recrystallised grains within region 'B', are products of low-angle rotations and nucleate at the vicinity of a LAGB with an orientation relationship of  $9^{\circ}(110)$  with the as deformed matrix. The recrystallisation retardation due to the solute segregation at SGBs and recrystallisation boundaries is believed to favour the LSE regions ( $\{001\}\langle 110\rangle$  deformed grains) to recrystallise at a later stage, with an oriented nucleation mechanism. Subgrain coalescence might explain the formation of these recrystallised grains. However, for subsequent growth,

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Figure 14: Recrystallisation within a parent near {001}(110) grain of LSE regions in the ternary alloy, a) {411}(148) and {611}(16 12) recrystallised grains highlighted. b) the micro-texture ODFs at the  $\phi_2 = 45^\circ$  section of the Euler space, of only the recrystallised grains. The texture maxima and number of grains analysed are for t = 0s: x24.7 mrd at  $\phi_1 = 10^\circ$ ,  $\Phi = 10^\circ$ ,  $\phi_2 = 45^\circ$  (only parent grain) – t = 290s: x12 mrd at  $\phi_1 = 29^\circ$ ,  $\Phi = 29^\circ$ ,  $\phi_2 = 45^\circ$ (N = 40) – t = 590s: x8.5 mrd at  $\phi_1 = 14^\circ$ ,  $\Phi = 0^\circ$ ,  $\phi_2 = 45^\circ$ (N = 71) – t = 1970s: x9.4 mrd at  $\phi_1 = 19^\circ$ ,  $\Phi = 14^\circ$ ,  $\phi_2 = 45^\circ$ (N = 158), respectively.

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<sup>911</sup> the presence of mobile boundaries is needed.

#### 912 4.3. Grain Boundary Mobility

Grain boundary mobility is a necessary criterion for a suc-931 913 cessful recrystallisation nucleus. Furthermore, it is an asset932 914 for already recrystallised grains to acquire a growth advantage933 915 [77]. Mobility of all types of boundaries is dramatically af-<sup>934</sup> 916 fected by solute addition due to solute drag, but the mobility of935 917 CSL boundaries may be less affected compared to general GBs.936 918 In a previous study, we have discussed the solute drag effect<sup>937</sup> 919 of Sn on the migration of general GBs during recrystallisation938 920 [52]. In the presence of solute atoms, the high mobility of low<sup>939</sup> 921 CSL boundaries is believed to be pivotal to the microstructural<sup>940</sup> 922 development, as these interfaces may suffer less drag due to so-941 923 lute grain boundary segregation [11]. Despite that segregation<sup>942</sup> 924 of comparable magnitude is also expected at CSL boundaries943 925 by thermodynamics as reported for Sn in  $\alpha$ -Fe by Lejček et al.<sup>944</sup> 926 945 [54]. 927

From the sequential EBSD experiment, the selective growth possibility of the *R* component was evaluated. CSL distributions in both binary and ternary alloys, showed the increased frequency of some  $\Sigma \langle 110 \rangle$  type boundaries [78]. Especially during the growth stage of recrystallisation, the fraction of  $\Sigma 9$ ,  $\Sigma 19a$ ,  $\Sigma 33a$ ,  $\Sigma 33b$  and  $\Sigma 11$  increased within the polycrystalline structures. These boundaries can potentially promote the growth of  $\gamma$ -fibre orientations by  $\langle 110 \rangle$  type orientation transformation of  $\alpha$ -fibre grains as described by Gangli et. al [18] and computer simulations of orientation selectivity by Kestens et al. [79]. In a re-evaluation of selective growth in bcc steels, Verbeken et al. [73] claimed to have revealed the formation of the *R* component as a selective growth mechanism from the as deformed  $\{112\}\langle 110 \rangle$  orientations due to the increased mobility of  $26.5^{\circ}\langle 110 \rangle$ , near the CSL  $\Sigma 19a$  boundary.

The misorientation distributions in the recrystallised segmented data were evaluated for the LSE regions in the ternary alloy. In these parent grains, the formation of strong R component was observed, which showed a strong correlation with



Figure 15: a) The CSL boundary frequency normalised by the random grain<sup>978</sup> assembly [76] and b) misorientation/axes bounding  $\{411\}\langle148\rangle$  recrystallised<sub>979</sub> grains at  $X_V^{rel} = 0.4$  in the ternary alloy.

high mobility misorientation/axes known in bcc metals. In par-982 947 ticular, the  $\{411\}\langle 148\rangle$  component is highly bounded by  $\langle 110\rangle^{983}$ 948 boundaries. Fig. 15 shows the boundaries of  $\{411\}\langle 148\rangle$  re-<sup>984</sup> 949 crystallised grains in the ternary alloy as a function of area985 950 fraction recrystallised. Fig. 15a shows the CSL frequency of 951 this component normalised by the random grain assembly  $\dot{\mathrm{CSL}}^{_{987}}$ 952 frequency based on boundary length [76, 80]. From the CSL<sup>988</sup> 953 and misorientation distributions, the highest frequency was ob-989 954 served at  $29 - 30^{\circ}\langle 110 \rangle$  near the  $\Sigma 19a$  (26.5° $\langle 110 \rangle$ ) as in Ibe-<sup>990</sup> 955 Lücke's model of selective growth in Fe- 3%Si [81, 19]. Mean-991 956 while, it is interesting to observe that more  $\langle 110 \rangle$  CSL bound-<sup>992</sup> 957 aries were associated with this component, namely  $\Sigma 27a$ ,  $\Sigma 33a^{993}$ 958 and  $\Sigma 33b$ . 959

On the other hand, the  $\{611\}\langle 16\ 12\rangle$  component (peak inten-960 sity within the LSE region) revealed a high frequency of GBs<sup>996</sup> 961 about 30°(111) instead, which is near the  $\Sigma 13b$  (27.8°(111)).<sup>997</sup> 962 Sanjari et. al [82] observed a similar behaviour of selective<sup>998</sup> 963 growth of the *R* component having a high frequency of  $30^{\circ} \langle 111 \rangle^{999}$ 964 GBs. All aforementioned HAGBs could acquire the necessary 965 mobility to assist the growth of the R component grains and 966 survive the grain growth process [12, 23]. Perhaps the selec<sub>7001</sub> 967 tive growth of  $\{411\}\langle 148\rangle$  due to the near 26.5° $\langle 110\rangle$  orienta-968 tion relationship, might be a key factor in why these grains have<sup>002</sup> 969 shown to dominate the primary recrystallisation texture of the003 970  $\{001\}\langle 110\rangle$  parent orientations, as they have been regularly re<sup>1004</sup> 971 ported to be present in the secondary recrystallised microstruc1005 972 1006 ture as well [83]. 973

Recently, Suehiro et al. reported on the effect of Sn during<sup>007</sup> the grain growth following primary recrystallisation of Fe3%<sup>008</sup>



Figure 16: Schematic of recrystallisation sequence in the two alloys constructed from the texture densities and recrystallisation kinetics observations: a) binary (no Sn) similar to [22] and b) ternary, with Sn addition.

Si and with Monte Carlo simulations [84]. They observed that the growth of the  $\{411\}\langle148\rangle$  grains increased when Sn was added, while in contrary the growth of  $\{111\}\langle112\rangle$  grains reduced. These authors attribute the growth behavior with Sn to a difference in solute drag effect on low-angle and highangle GBs. Such non-isotropic solute drag can potentially be of importance, although the size advantage of the  $\{411\}\langle148\rangle$ grains already acquired from primary recrystallisation should not be overlooked. Grain growth after recrystallisation, essentially is a process during which the larger grains consume the smaller ones. Finally, it is interesting to note that when only the  $\{411\}\langle148\rangle$  grains are concerned Suehiro et al. also observed a peak near the 26.5° misorientation as we report here from the sequential EBSD observations.

Overall, from the recrystallisation observations reported here and previous *ex situ* study, the recrystallisation sequence in the two studied alloys: with and without Sn can be summarised schematically in Fig. 16. Fig. 16a shows the case for the binary alloy (no Sn), which as it was shown here, has a similar recrystallisation behaviour to low carbon steels as proposed by Hutchinson [22]. Fig. 16b shows the sequence in the presence of solute Sn. The schematic is semi-quantitative, including information on the recrystallisation retardation and orientation magnitude proportional to the texture densities measured experimentally taking into account several thousands of grains [34].

#### 5. Conclusions

The static recrystallisation in cold rolled Fe-Si model alloys was investigated with sequential EBSD assessing for the effect of Sn solute addition during recrystallisation annealing. Sn segregation was analysed site-specifically with APT, revealing segregation at different stages of recrystallisation that is crucial in understanding the texture formation. The orientation dependence of stored energy is proven essential for the understand-

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ing of texture modification with Sn. As in the case of no Sn, the063 1009  $\{111\}\langle uvw \rangle$  parent grains were the highest stored energy orien<sup>1064</sup> 1010 tations, while the low  $\Phi$  angles along the  $\alpha$ -fibre and  $\{001\}\langle 110 \rangle_{106}^{1065}$ 1011 were the lowest stored energy orientations. Without Sn, the,067 1012 fastest regions to recrystallise ( $\{111\}\langle uvw\rangle$ ) were observed to<sup>068</sup> 1013 dominate the microstructure during recrystallisation and essen1069 1014 1070 tially defined the texture. 1015 1071

Recrystallisation texture with the addition of Sn can be sum  $_{1072}$ 1016 marized in the following. Solute affected significantly the onseto73 1017 of recrystallisation in {111}(uvw) oriented regions by segrega<sup>1074</sup> 1018 tion at the subgrain boundaries of the as deformed/recovering  $d_{076}$ 1019 microstructure, prolonging the incubation period. At this stage<sub>1077</sub> 1020 Sn segregation was suggested to have a pinning effect during<sup>078</sup> 1021 subgrain coarsening and hence, retard the onset of recrystallised<sup>079</sup> 1022 grains. Recrystallisation proceeds with parent grains recrys<sub>1081</sub> 1023 tallising independently in the Sn-containing alloy; segregation<sub>082</sub> 1024 at prior grain boundaries could be responsible for the reduction083 1025 of SIBM from faster recrystallising regions. The growth of new<sup>1084</sup> 1026 grains was dictated by the available stored energy, nucleation  $\eta_{086}$ 1027 density within each parent grain and solute drag of Sn on mi+1087 1028 grating interfaces. Low stored energy regions recrystallised in1088 1029 dependently at later stages of the process. Oriented nucleation<sup>1089</sup> 1030 of  $\{411\}\langle 148 \rangle$  grains was observed in these regions. Segregation 1031 phenomena were shown to promote {411}(148) recrystallisation<sup>092</sup> 1032 as a consequence of solute drag in as deformed  $\{111\}\langle uvw \rangle$  re<sup>1093</sup> 1033 gions. The growth of such grains was one of the fastest  $ob_{1095}^{1094}$ 1034 served within the primarily recrystallised microstructure. Ibe<sub>096</sub> 1035 and Lücke known theory of selective growth of 26.5°(110) bound= 1036 aries might explain why these grains grew so rapidly. 1037

These results showed that the combination of local chemical  $\frac{1}{100}$ 1038 element distributions and thermodynamic study with crystallo<sub>1101</sub> 1039 graphic analysis can be very insightful and can pave the way102 towards understanding the active physical mechanisms not only<sup>1103</sup> 1041 in the Fe-Si system, but may be expanded for other single-phase 1042 metals were solute segregation takes place. 1043 1106

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