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RESEARCH ARTICLE

Improvement of low temperature impact toughness through flux modification for submerged arc welded low carbon steel E350 plates

Lucky[1]*, **Harish Arya[2]**

1 Department of Mechanical Engineering, SLIET Longowal, 148106, Punjab,
Tel.: +919417424879
2 Department of Mechanical Engineering, SLIET Longowal, 148106, Punjab

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***Corresponding author**.
Lucky
Department of Mechanical Engineering, SLIET Longowal, 148106, Punjab,
Tel.: +919417424879
luckysaklani94@gmail.com

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Abstract

Objectives: To analyse the effects of rutile (TiO_2) and Alumina (Al_2O_3) on impact toughness properties of Submerged arc welded joints for low temperature applications. **Methods**: Al_2O_3 and TiO_2 have been added to F7AZ/PZ-EL8 Submerged arc welding (SAW) flux in varying quantities and the base material used for joining is low carbon structural steel having E350 grade. To verify the effect of these elements by flux modification, the Charpy V-Notch impact tests, scanning electron microscope (SEM) fractography and visible light microscopy (VLM) have been conducted. **Findings**: Charpy V-Notch impact testing and analysis have shown that there is considerable improvement in the impact properties of welds with the addition of 20% TiO_2 and 20% Al_2O_3 in the 80% of flux as separate constituents i.e., when these elements have been added in the flux in separate form or two new modified fluxes were prepared, where one is doped with alumina and other is with rutile. Fractography of welds with addition of 20% alumina in 80% F7AZ/PZ-EL8 SAW fluxes are having best impact properties at lower temperature and addition of 20% rutile also as almost effective as alumina in flux. consequently, mode of failure is ductile fracture with dimples as a result of good amount of acicular ferrite microstructure. **Novelty**: Analysis of previous studies implicated that there is lack of significant studies in the low temperature impact properties of low carbon steel submerged arc welds. Present study shows that the significant improvement in the toughness properties of welds at lower temperature of -40°C can be achieved through flux modification, it would ensure the application of Low carbon steel structures at lower working temperature. The applications may be pressure vessels, ships or containers.

Keywords: Submerged arc welding (SAW); low temperature impact toughness; flux modification; low carbon steels

1 Introduction

Submerged arc welding (SAW) is the essential welding operation needed to fabricate the ship hulls, containers, boilers and pressure vessels [1, 2]. Their operating conditions of these components are severe and challenging depending upon external environment and temperature conditions. One of greatest obstruction which limit the use of SAW welded low carbon steel components, is low impact toughness at low temperature. Low carbon steel having BCC structure tends to lose the ductility at temperature just below the freezing point of water. Numbers of fatalities have been happened when the importance of impact toughness is ignored. There have been significant practices to improve the toughness of the low carbon steels. The factor which is responsible for the improvement is the amount of acicular ferrite present in the weldment, which has been proved by various studies. Researchers have tried to improve the toughness of welds by addition of titanium, manganese, nickel etc., however their results seems to be contradictory at some point. Impact toughness properties have not been studied in detail at low temperature. While studying the microstructure and properties of pipeline steels having acicular ferrite (AF), mechanical as well as microscopic examinations were also conduced to compare the above said steel with the steel merely having ferrite and pearlite. Analyses have proved the steels with AF have high strength, improved toughness and lowering of ductility to brittle transition temperature (DBTT) as compared to steels with simple pearlite and ferrite. This is result of uniform microstructure and clean steel [3] . Acicular ferrite nucleates heterogeneously, and its nucleation is inter-granular. It is also called as intergranular nucleated bainite. The formation temperature of the acicular ferrite and bainite is similar however these differ in nucleation mechanism. The grains of bainite nucleate and grow parallel to each other and the nucleation of the acicular ferrite is chaotic and it tends to spread in all directions [4] . Researchers have studied the capabilities of the different inclusions to form intergranular ferrite. It has been found that the TiO_2 and Al_2O_3 inclusions form ferrite layers between the steel and oxide interfaces. TiO_2 can promote the nucleation of intergranular ferrite and it has ability to change the chemical composition of micro metal zones adjacent to manganese depleted zones [5] therehas been attempt on improvement of alloy design of X70 pipeline steels to study its effect on the impact toughness and its analysis concluded that the increase in Ti/N ratio has resulted in the higher percentage of average finer grains at coarse gain heat affected zone (CGHAZ). Increase in Ti/N ratio has not significantly altered the precipitate size. The Ti/N ratio of 3.2 has resulted in maximum density of precipitates and have maximum Charpy V-notch Impact (CVN) as well crack tip opening displacement (CTOD) strength values [6].

Generally, two types of inclusions i.e. spherical and faceted inclusions appear in the weld metal. The spherical inclusions promote the nucleation of acicular ferrite (AF). This ends up in chaotic grain structure and reduction of other ferrites, resulting in increase in impact toughness. This makes the fracture mostly ductile in nature. It has also been found that the formation of AF depends on size of pre-austenite grains, smaller pre-austenite grains help in the formation of AF. Also, the oxide inclusions enhance the promotion of formation of AF [7]. Some studies have found that the increase of Ti from 0.014% to 0.048% has resulted in significant growth of acicular ferrite from 30% to 80%. Yield strength (YS), ultimate tensile strength (UTS) and vickers hardness number (VHN) slightly reduced, however there is small improvement in the CVN energy [8]. While comparing annual metal arc (MMA) welds and SAW, and Ti composition in the weld metal, studies have concluded that the variation of Ti in weld metal from 690 to760 ppm in SAW has resulted in the increased hardenabilty, while there has no significant change in microstructure of MMA welds. SAW welds have higher volume fraction of inclusions and Mn, Ti, Si were inclusions in both types of welds, yet MMA welds have the Al as inclusions [9]. Three process i.e., SAW, SMAW and four wire tandem SAW with the addition of Ti on the process have been compared with each other. Base metal having fine grains have been assumed to restrict the grain growth during heat treatment due to pinning effect. SEM and electron dispersive spectroscopy (EDS) analysis showed that inclusion of 2 μm and major constituting elements are Fe, Ti, O, Si and Mn. In Titanium doped welds, there is the highest amount of acicular ferrite followed by polygonal ferrite and FS. Higher TiO_2 has ended up in higher recovery of Mn due to significant slag-metal reactions. Also, the doping with 'Ti' has resulted in slight improvement of mechanical properties. CVN impact energy has been drastically reduced in case of SAW welds at -10°C, SMAW welds show very little improvement while in T-SAW impact strength has significantly improved and all these were obtained due to significant variation in acicular ferrite (AF) and polygonal ferrite (PF) [10]. Ti and Mn have been added to the SAW flux as the alloying elements. ANOVA plots have shown that the tensile strength of the welds increases up to 13% addition of TiO_2 and it reduces after further addition of rutile. Microscopic examinations revealed that the weld metals have the Equiaxed ferrite and acicular ferrite as majority and small portion of pearlite colonies. Mn has resulted in recovering the tensile strength of the weld metal, increased TIO_2 has resulted in increased ductility and impact strength at the cost of tensile strength [11].

In this paper, the SAW flux F7AZ/PZ-EL8 having neutral nature is doped with rutile and alumina in different proportions and their effects on impact properties of low carbon steel welds at low temperature as well as microstructure has been analysed. The details of work are given in upcoming sections.

2 Materials and work methodology

2.1 Consumables Used

To conduct the experiments, the base material selected for the purpose of study was E350 grade low carbon steel whose chemical composition is given in the Table 1. This steel is extensively used for boiler and pressure vessel manufacturing. The welding was done on the faying surfaces of two plates having square groove geometry. Dimensions of plates were 75×250×10 (W×L×T) mm. The filler wire used for the deposition of the welds was copper coated 3.15 mm diameter EH 14 wire (AWS code F7A2 -EH14). The chemical composition of filler wire is given in Table 2. The flux used for the welding and modifications was F7AZ/PZ-EL8 flux manufactured by Ador India Limited. It is aluminate rutile type having basicity index of 0.9. The nature of this flux is neutral, and it is preferred for multi-pass welding due to good slag detachability.

Table 1. Chemical composition of the base metal (E350 grade)

Element	C	S	P	Mn	Fe
Percentage by weight	0.18	0.008	0.015	0.95	rest

Table 2. Chemical composition of the filler wire used

Element	C	Mn	Si	S	P	Cu	Fe
Percentage by weight	0.077	1.64	0.33	0.015	0.022	0.12	rest

2.2 Work Methodology

The experimentation methodology for this research work is given in Figure 1 and after the selection of consumables for welding, the fluxes had been modified as given on the Table 3. The unmodified flux was coded as 'O' and others had been coded as 'A', 'B', 'C', 'D', and 'E'. The base flux 'O' was

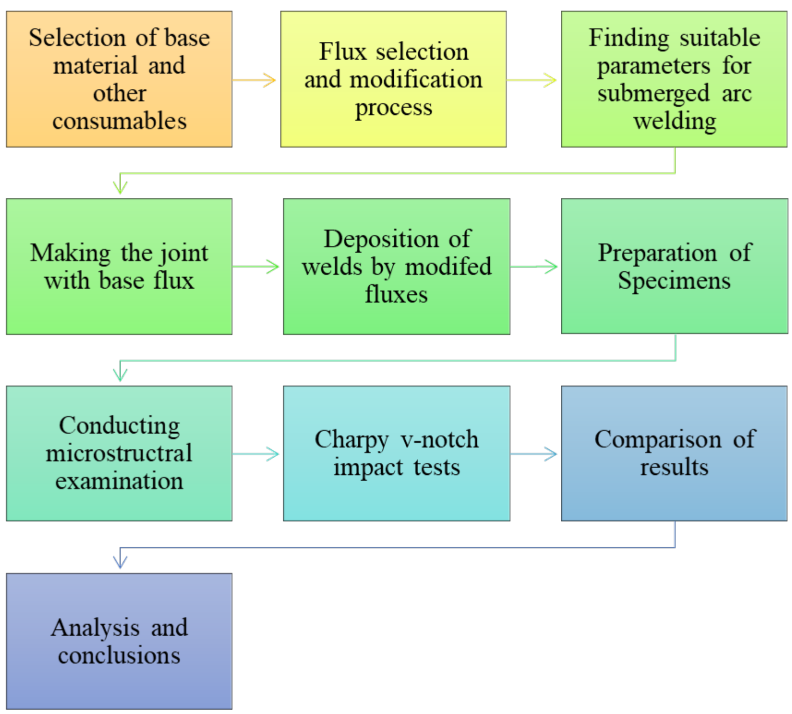

Fig 1. Experimentation methodology

subjected for different additions of TiO_2 and Al_2O_3 as given in Table 3 and the sodium silicate was used as the binder for mixing all the elements when it had the powdered form. Flux coded as 'O' was crushed and sieved to powder from before mixing other components. Then the crushed flux and other additives were dry mixed after weighing to proportion. After dry mixing sodium silicate and water were added to make the paste form. Then the wet mixture had been passed through a sieve which is having 2 mm size as per ASTM-E11 standard and left for drying in still air for 24 hours at room temperature. After drying, modified fluxes were baked at 700°C in muffle furnace for 2 hours. When baking was completed, followed by cooling in air, uniform grain size of 2 mm is obtained by further crushing of modified and baked fluxes.

Table 3. Composition of modified fluxes

Code	Percentage of TiO_2	Percentage of Al_2O_3	Percentage of base flux
A	10	10	80
B	6	14	80
C	14	6	80
D	20	NIL	80
E	NIL	20	80
O	NIL	NIL	100

The Figure 2 shows the photographs of unmodified as well as the modified fluxes. When the fluxes had been modified, these were stored and heated in oven before the welding is done. The parameters selected for the welding process are shown in Table 4. The plates have been joined with the help of Tornado SAW M-800 submerged arc welding machine. The two plates were joined as per parameters given in the Table 4, the plates were having square groove geometry at faying surface and the gap between joining surfaces was 1.5-2 mm. When all the plates had been joined, these have also been coded as same as the fluxes were. For e.g. plates joined with the help of flux 'A' have been coded as 'A' and so on. Specimens for CVN impact testing, chemical analysis and chemical composition were taken from the weld zone.

Fig 2. Modified Fluxes after addition of $TiO_2$and Al_2O_3

Table 4. Welding parameters

Parameters	Value
Current	550 A
Voltage	28.5V
Nozzle to plate distance(NPD)	35 mm
Speed	20 m/h

The chemical analysis was done to check the chemical composition of weld metal and spectroscopic technique was employed to detect the presence of different alloying elements. Charpy V-Notch impact tests were conducted for all types of specimens from temperatures -60°C to +40°C with difference of 20°C. After the impact tests, Ductile to brittle transition temperature (DBTT) curves were drawn for each plate welded from modified fluxes and unmodified flux. The specimens which showed good results were further subjected to SEM-fractography and microstructural analysis. The SEM-Fractographs of impact tested specimens 'D', 'E' and 'O' were taken at -60°C and +40°C in SAI Labs, TIET, Patiala, Punjab INDIA. The magnifications of fractography were 500x, 1000x and x2000x. These specimens had also been subjected to microstructural analysis by visible light microscopy (VLM) at 200x. The specimens were polished and etched with Nital solution having 96-98% methanol and 2-4% of nitric acid.

3 Results and discussion

3.1 Chemical analysis

Spectroscopic techniques have been employed to detect the presence of various alloying elements in weld metal. These tests have been done at Sunbeam Auto Pvt. Ltd. Ludhiana, Punjab INDIA**.** The given Table 5 shows the chemical composition of the weld metals deposited with the help of different coded fluxes.

Table 5. Chemical composition of different weld specimens codded according to flux types

Code (TiO_2 / Al_2O_3)	% C	% Si	%Mn	% P	% S	%Cr	%Mo	Ni	**%Al**	%B	%Co	%Cu	**% Ti**	%V	%Fe
A(10/10)	0.130	0.227	0.868	0.026	0.022	0.084	0.036	0.119	**0.028**	0.0008	0.023	0.103	**0.015**	0.006	rest
B(06/14)	0.132	0.221	0.889	0.026	0.022	0.084	0.036	0.119	**0.032**	0.0008	0.019	0.105	**0.012**	0.006	rest
C(14/06)	0.132	0.198	0.896	0.025	0.022	0.083	0.036	0.117	**0.023**	0.0008	0.019	0.103	**0.018**	0.006	rest
D(20/nil)	0.135	0.199	0.856	0.026	0.022	0.083	0.034	0.115	**0.020**	0.0008	0.018	0.103	**0.023**	0.005	rest
E(nil/20)	0.150	0.268	0.991	0.026	0.023	0.085	0.037	0.114	**0.045**	0.0007	0.018	0.104	**0.004**	0.007	rest
O(nil/nil)	0.147	0.269	0.985	0.025	0.023	0.084	0.035	0.115	**0.021**	0.0007	0.017	0.0105	**0.004**	0.007	rest

The results given in Table 5 show that addition of TiO_2 and Al_2O_3 alone has resulted in the significant difference of above elements' chemical composition, however overall alloy composition has increased marginally, which has improved the impact properties. There has been significant improvement in the alloying elements pickup of the weld metal in 'D' and 'E' as compared to others, which has been deposited with the help of modified flux having 20/0 and 0/20 percentage addition of TiO_2/Al_2O_3. Its alloying pickup properties were good as compared with welds deposited with non-modified flux. The Mn and Si in other specimens 'B' and 'C' help in improving the little bit of toughness properties of the weld metal, which makes it to bear the shocks and impact loads. By seeing the composition of specimens 'D' and 'E', here we have observed a notable increase in the weight percentage of titanium and aluminium in the weld metal. Which in result has led to considerable improvement of impact properties at lower temperatures, which is discussed in upcoming sections.

3.2 Charpy V-Notch impact Test

The Charpy V-Notch (CVN) impact tests have been conducted as per ASTM E23 standard at 6 different temperatures ranging from -60°C to +40°C. For every temperature point, five Charpy impact tests were performed. The maximum and minimum values were removed, and average of rest three readings are presented in Table 6. Only two curves from specimens 'D' and 'E' have been found to produce the good results with variation of temperature as shown in the Figure 3. It means that the addition of TiO_2 and Al_2O_3 has been become beneficial which these were added alone to the flux. When added in combined forms 'A' 'B' and 'C', there has not positive effect on the impact strength of welds. This may due to insufficient alloying elements' addition to the weld metal or poor solubility of oxides to the weld metal. This on the other hand, resulted in the poor properties of weld metal due to poor pickup of carbon, manganese and silicon as shown in the Table 5.

The Figure 4 shows the comparison of the curves obtained from specimens 'D' and 'E' to specimens 'O'. These two curves are comparable to curve obtained from reference flux 'O'. Seeing the graphs, the impact strength of the specimen 'D' is higher as compare to the specimen 'O'. It is somewhat greater than reference curve obtained from non-modified flux 'O'. This means that the addition of titanium has proved to improve the mechanical properties of the welds. The addition of 20% of Titanium Dioxide along with 80% of un-modified flux has not affected the impact properties to an extent, however it resulted in a good improvement of properties at lower point of temperature ranges. As we know, the non-metallic inclusions act as the sites for nucleation acicular ferrite. The titanium inclusion might have been helped to promote the formation of acicular ferrite. Spherical inclusions enhance the probability of formation of acicular ferrite, however it may not be always true [7] . While the specimen 'E' has resulted in significant improvement in Charpy impact values starting from -40°C and it is up to +40°C. It has been significant improvement at -20°C as we observe the Figure 4 by comparing 'E' and 'O'. There has been the difference of absorbed

Table 6. Charpy impact energy values of weld specimens deposited with modified fluxes

Code	TiO_2	Al_2O_3	Impact values in Joules at Various Temperatures (in Joules)					
			-60°C	-40°C	-20°C	0°C	+20°C	+40°C
A	10	10	24	35	62	105	120	126
B	6	14	28	36	63	102	130	136
C	14	6	32	42	68	96	120	128
D	20	nil	38	50	90	120	146	154
E	Nil	20	42	63	99	130	145	160
O	NIL	NIL	28	40	76	101	120	129

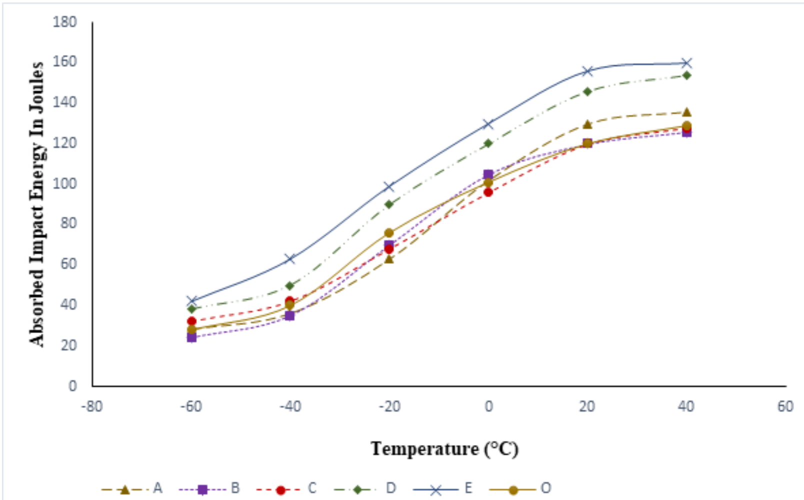

Fig 3. Comparison of absorbed impact energies of specimens tested from -60 °C to +40°C

energies of 23J at -40°C between welds made from flux 'E' and 'O'. The addition of 20% Al_2O_3 has been beneficial for the impact bearing capability of the welds. In introduction section of this paper, it is already discussed that the Al_2O_3 has good capability to formation of acicular ferrite which has chaotic structure. This chaotic structure of acicular ferrite prevents the brittle fracture of the specimen.

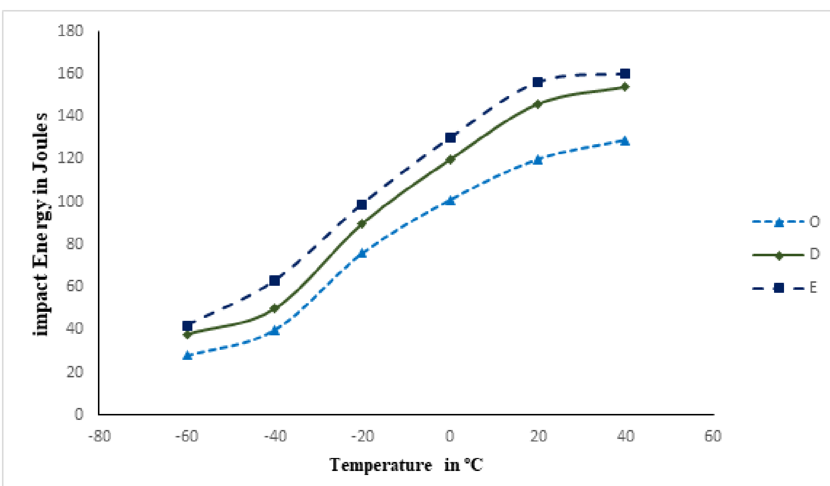

Fig 4. Comparison of impact toughness of specimens 'D', 'E' and 'O'

3.3 Fractographs

Figure 5 shows the fractographs of CVN test's broken specimens 'D', 'E' and 'O' at -60°C and x500 magnification obtained in SEM. By observing the above figures, we can say that the majority type of fracture is brittle or cleavage fracture. The brittle fractured surfaces consist of tearing ridges and cleavage facets. The cleavage facets have smoother appearance under higher magnification. These facets offer less resistance for the propagation cracks generated due to impact loadings. Tearing ridges have river like appearance. There is also partial ductile fracture at some locations in all above three specimens. The BCC crystal structure is more severe to brittle fractures at lower temperature while FCC structures are not generally subjected to brittle fracture at lower temperatures [12].

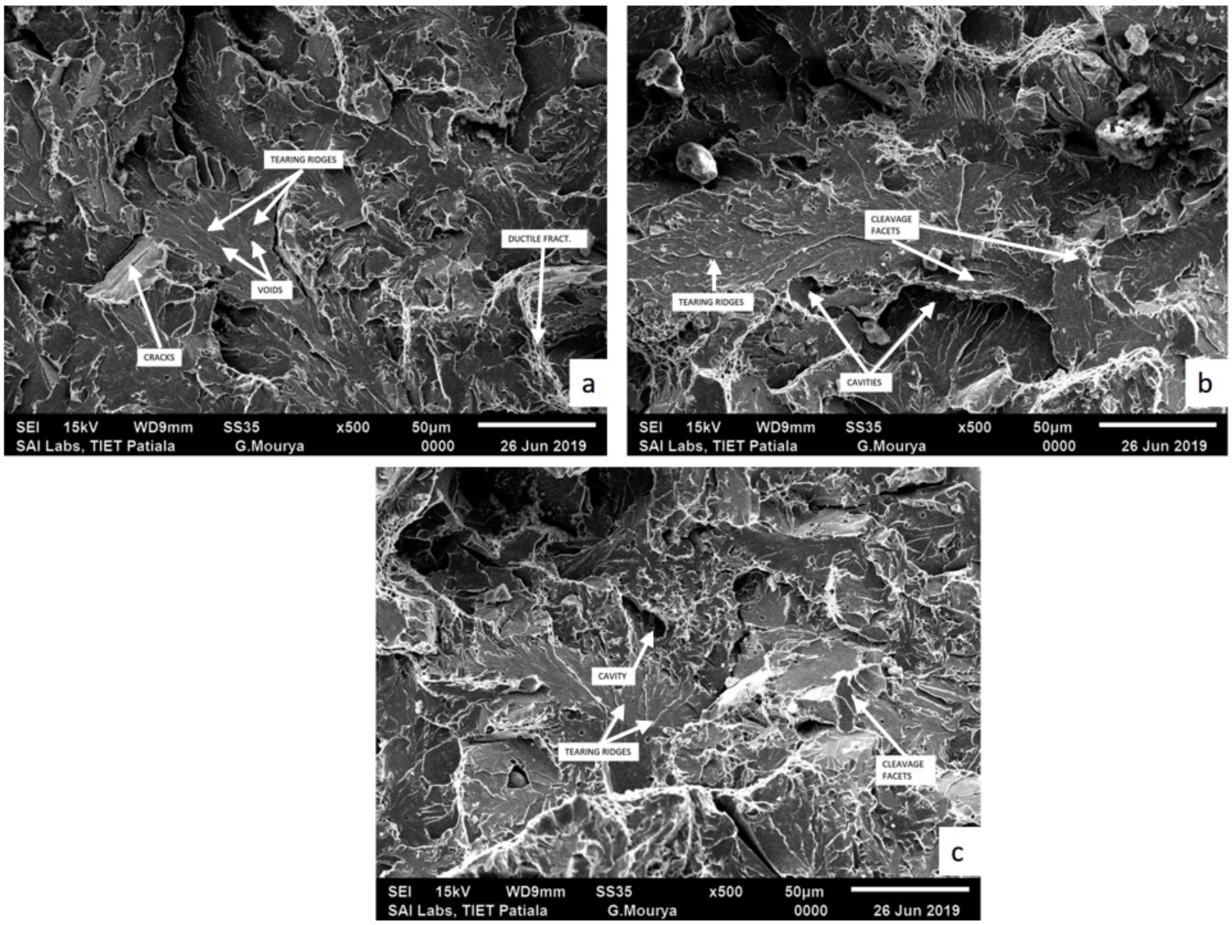

Fig 5. Fracto graphs of welded specimens: a). 'D' b) 'E' and c) 'O' at x500 and -60°C

Figure 6 shows the fractographs of impact tested specimens 'D', 'E' and 'O'. As discussed in previous section at x1000, there is also revelation of cleavage facets, tearing ridges. Along with them, we can see some fractured surface with dimples i.e., ductile surface, which means the fracture is not purely brittle at all. Some cavities and voids can also be seen here, which are generated during the fracture. These are due to vacancies created during the propagation of cracks. Inclusions during the welding may also be the cause of the voids and cavities because of the poor mixing with metal during solidifications. However micro-inclusions are not always proved to be harmful for the weld metal's mechanical properties. Their size and shape affect the grain type and geometry, type of ferrite etc. In previous researches, it has been found that the spherical inclusions of size 1-3 μm have proved to be beneficial for the impact properties and ductility by promoting the formation of acicular ferrite [4, 7, 13].

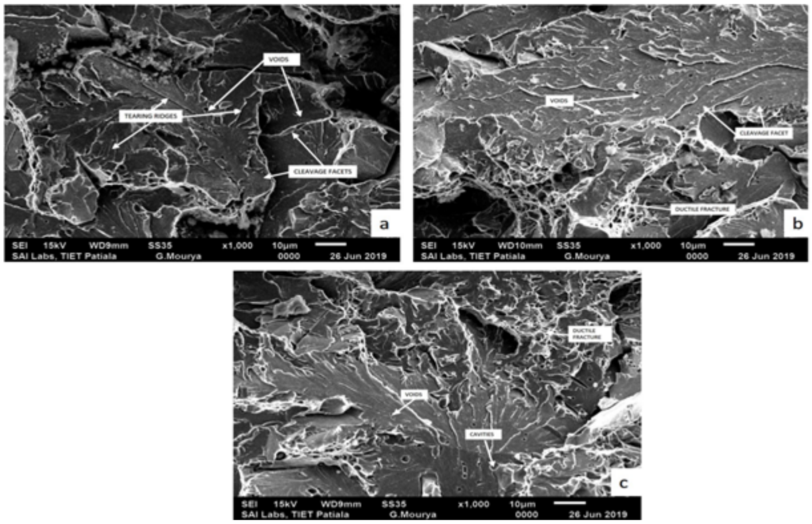

Fig 6. Fractographs of welded specimens: a). 'D' b) 'E'and c) 'O' at x1000 and -60°C

Fractographs of specimens, 'D', 'E' and 'O' at x500 and CVN impact test at +40°C are represented in the Figure 7. Observing the SEM images, we can see that the nature of fractured surface is ductile. It consists of honeycomb like structure which is indicator of ductile fracture. In these fractographs, we can also see the cavities as well as voids. Cavities are formed when a large surface area is fractured. Voids are formed at stress concentration points which are generated during the loading. These voids can also be formed due to imperfect crystal structure of the material and entrapment of micro inclusions during welding. Generally, the ductile fracture occurs in intergranular manner. It progresses in grain boundaries which have lower strength at higher temperature.

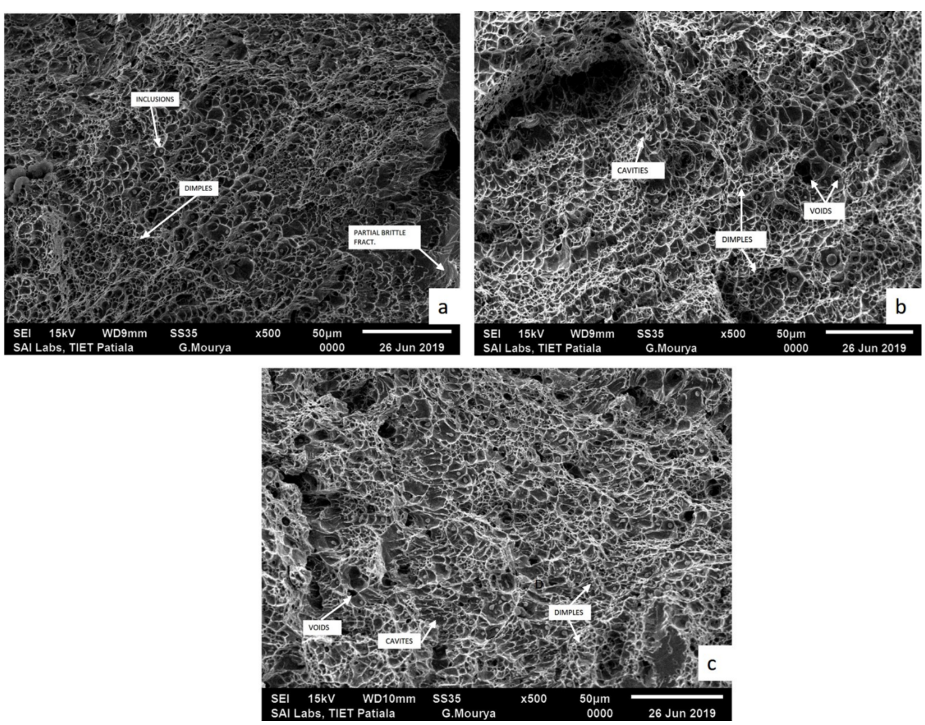

Fig 7. Fractographs of welded specimens: a) 'D' b) 'E' and c) 'O' at x500 and +40°C

Images given on Figure 8 show the fractographs of 'D', 'E' and 'O' at x1000 and impact test temperature was +40°C. The properties of fractured surface are analysed at higher magnification. At x1000 magnification, all three specimens' surfaces show the significant number of inclusions at the pints where the fracture has been occurred. Most of the inclusions are spherical and oval shapes. As analysed, the spherical inclusions help to prevent the cleavage fracture. These tend to increase the toughness properties of the weld metal. There may be a large amount of the acicular ferrite which increase the impact energy by restricting the crack propagation. The chaotic and non-ordered structure of acicular ferrite is major factor which helps to prevent the brittle fracture. These act as the pinning elements across the grains thus resisting the propagation of cracks. Interestingly the number of round and oval inclusions have been significant in the welds made with modified fluxes 'D' and 'E', here 'E' has highest fraction of these metal oxide inclusions which are really helpful for the formation of acicular ferrite [7] .

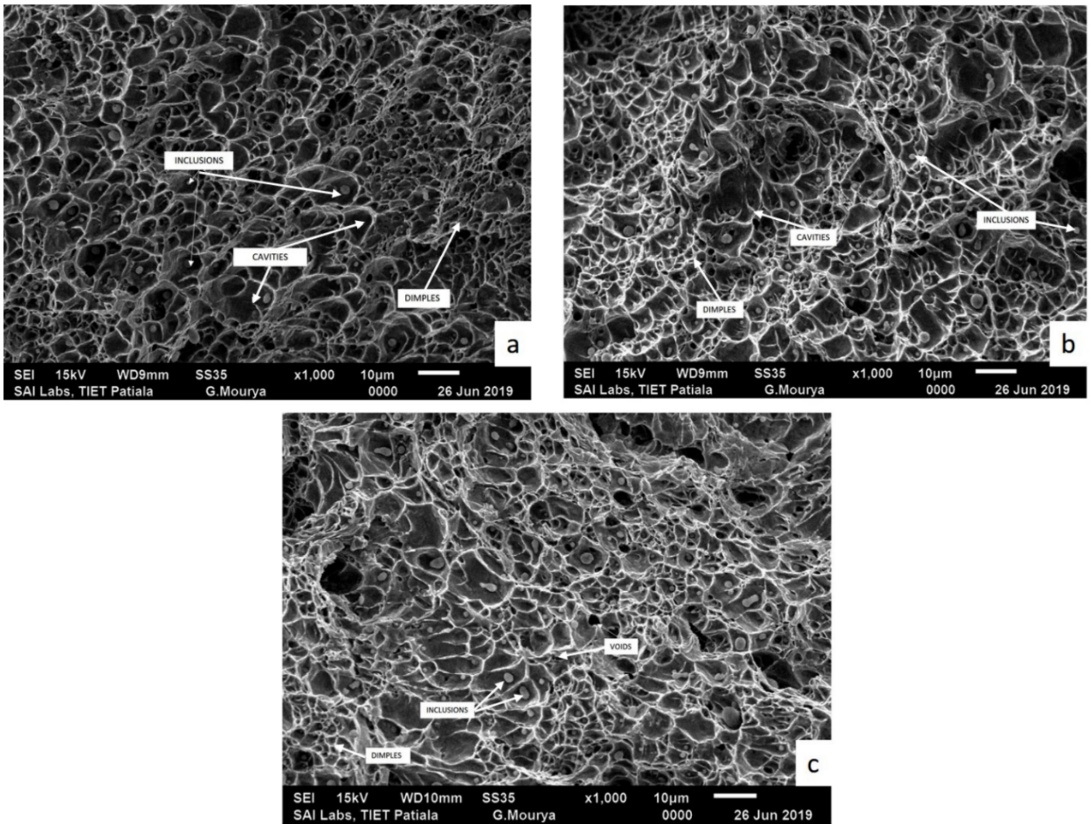

Fig 8. Fractographs of welded specimens: a) 'D' b) 'E' and c) 'O' at x1000 and +40°C

3.4 Microstructure of welds

The Figure 9 shows visible light micrographs (VLM) of welded specimens 'D', 'E' and 'O'. As we can see, there is no specific single kind of ferrite present in the weld metal. There is acicular ferrite, grain boundary ferrite, Equiaxed ferrite and pearlite segregated across the weld metal. Acicular ferrite is the major concentration on which our study is focused. We know that acicular ferrite's chaotic structure resists the crack propagation rate and slows it down under high strain rates i.e., impacts [4, 7, 13] .Looking at micrograph of 'D', the distribution of various kinds of ferrites is not uniform, it is scattered. The acicular ferrite is dominating the weld metal structure, which should be present. There is the presence of large size ferrite laths and GBF at some locations. GBF is characterized by elongated ferrite grains having some thickness. While ferrite laths are characterized by large grains spread in all directions.

This microstructure of 'E' is same as that of the 'D' and it also have much of the AF spread across the weld metal. The GBF and Equiaxed ferrite grains have been acted as the separators for AF at some locations. The acicular ferrite and large ferrite laths dominate the microstructure. These two structures might have been helped for the improved impact energy values at the lower temperatures. In 'O' there is significant presence of the large sized ferrite grains, scattered across the weld metal. Acicular ferrite, however, is the major component present in the weld metal, but there is observation that the GBF is having very small area fraction in weld metal as we can see from micrographs at X200. It is also not ignored that GBF may be present in the other locations. It is generally non-uniformly distributed in the weld metal zone. It can be concluded from these figures that the amount of AF is higher in specimen 'E' as compared to 'D' and 'O'. Effect of other forms of ferrite may have appeared to be positive on the impact strength of welds. This higher composition of AF and smaller grain sizes have led towards the improvement in impact toughness.

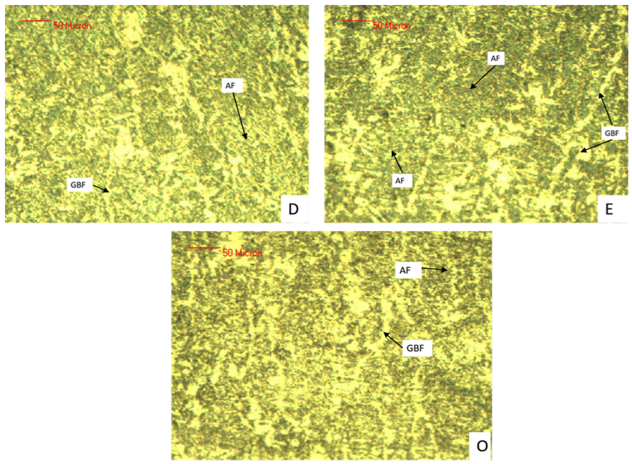

Fig 9. Visible light micrographs (VLM) of welded specimens, 'D', 'E' and 'O' at x200

4 Conclusions

The available flux has been modified by using various percentages of rutile (TiO_2) and alumina (Al_2O_3) powder. Weldment have been prepared with modified flux and impact strengths of welded specimens have been compared at various temperature conditions. The thorough analysis of results concludes the following outcomes:

1. Low temperature impact toughness graphs (DBTT curves) for submerged arc welded low carbon steel E350 joints have been drawn. These graphs may help to predict the loss of impact toughness for low temperature applications.
2. Specimen 'E' (welded by flux with 20% alumina) has maximum percentage of AF structure and hence provide maximum improvement in impact toughness at lower temperature ranging from 0°C to -40°C.
3. Welds with rutile and alumina mixed together have not shown good properties at lower temperature. The possible reason may be poor alloying, grain geometry as well as inclusions properties (lack of spherical and optimum size of inclusions).
4. Visible light micrographs of 'D', 'E' and 'O' clearly indicate that alloying elements like alumina and titanium dioxide work as inclusion which helps in formation of acicular ferrite structure and hence the percentage of acicular ferrite increased with modified fluxes along with smaller grain size.
5. SEM images of fractured surfaces showed that the number of inclusions having round & oval shape increased with specimen welded with 20% alumina and hence ductility and impact toughness improved at low temperature.
6. At -60°C, there is mostly brittle fracture of the weldments even with modified flux, this may be due to transition of the properties of welds at lower temperature, lack of new bonds formation. This results in faster crack propagation after initiation. However, the specimens 'D' and 'E' has absorbed significant good amount of energy higher than specimens 'O'.

57% improvement in impact strength of specimen 'E' as compare to 'O' has been observed at -40°C. The micro inclusion size observed with SEM images of specimen 'E' found in the range of 0.5-2 μm which is ideal for formation of acicular ferrite structure.

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