Modeling of Tests of Primary Water Stress Corrosion Cracking of Alloy 182 of Pressurized Water Reactor According to EPRI and USNRC Recommendations

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Abstract: One of the main degradation mechanisms which cause risks to safety and reliability of pressurized water nuclear reactors is the primary water stress corrosion cracking (PWSCC) in nickel alloys, such as Alloy 600 (75Ni-15Cr-9Fe), and its weld metal Alloy 182 (67 Ni-15Cr-8Fe). It can appear at several reactor nozzles dissimilarly welded with Alloys 182/82 between steel ASTM A-508 G3 and stainless steel AISI316L, among others. The hydrogen which is dissolved to primary water to prevent radiolysis, can also have influence on the stress corrosion cracking behavior. In this article one departs from a study of Lima based in experimental data obtained from CDTN-Brazilian Nuclear Technology Development Center, in slow strain rate test (SSRT). It was prepared and used for tests a weld in laboratory, similar to dissimilar weld in pressurizer relief nozzles, operating at Brazilian NPP Angra 1. It was simulated for tests, primary water at 325°C and 12.5 MPa containing levels of dissolved hydrogen: 2, 10, 25, and 50 cm^3 STP H_2/kgH_2O. The objective of this article is to propose an adequate modeling based on these experimental results, for PWSCC crack growth rate according to the levels of dissolved hydrogen, based on EPRI-MRP-263 NP. Furthermore, it has been estimated the stress intensity factor applied for these tests: according with these, some another models described on EPRI-MRP-115, and an USNRC Technical Report, have been tested. According to this study, CDTN tests are adequate for modeling comparisons within EPRI and USNRC models.

Keywords: Hydrogen Effect, Modeling, Pressurized Water Stress Corrosion Cracking, Slow Strain Rate Tests, Weld Nickel Alloys 82/182.

1. INTRODUCTION

The PWSCC is a very complex degradation mode in Nuclear Pressurized Water Reactor (PWR) ´s thick-walled components of nickel alloys – such as Alloy 600 and its weld metals (Alloy 182 and Alloy 82). It has been identified as an important mode affecting the safety and reliability of this type of plant operation. A constant effort has been done to develop and identify technologies to mitigate this damage mechanism. Until now, the main ones are hydrogen optimization and zinc injection, during PWR plant operations. The zinc injection is used to delay the PWSCC initiation due to its incorporation in spinel oxide films, thus enhancing their stability [1].

The hydrogen injection on primary water is normally applied to prevent radiolysis. The hydrogen optimization which consists in different hydrogen injection levels in primary water of the operating unity has been demonstrated to strongly mitigate the PWSCC growth rate, mainly in Alloy 182 and Alloy 82 weld metals. With respect to the PWSCC initiation time, most of available data did not show a conclusive effect on this, mainly due to the high degree of scatter. Also, the hydrogen increase above current operational levels did not have an enhancement effect on PWSCC initiation time [2]. Also, according to these authors, the best for this case is zinc addition, which has been demonstrated strong mitigation of the initiation of PWSCC in Alloy 600: further, the zinc addition is an additional way of decreasing occupational radiation exposure of plant staff, according to Nordmann(1).

It has been departed from a study of Lima [3] which investigates the influence of dissolved hydrogen contents on the susceptibility to PWSCC of Alloy 182, used as weld metal in a dissimilar weld between the steel ASTM A-508 G3 and stainless steel AISI 316L, similar to the weld which exists in a pressurizer nozzle of Angra # 1 nuclear power plant: it was evaluated. In this study was used a simulated PWR primary coolant water chemistry at 325ºC and pressure of 12.5 MPa with different levels of dissolved hydrogen: 2, 10, 25, 50 cm^3H2/kg H_2O at standard temperature and pressure (STP). Slow strain rate test (SSRT) was used to evaluate the Alloy 182 PWSCC susceptibility. Open

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circuit potential was measured in different hydrogen concentrations to evaluate their effect in the material electrochemical corrosion: the main study results indicated that Alloy 182 is less susceptible to PWSCC at 50 cm^3H_2 (STP) /kg H_2O at 325ºC, and showed the positive effect in to keep hydrogen concentration in a high level in the PWR primary coolant water.

The objective of this article is to study and to discuss an adequate modeling based on the experimental results from Lima´s study [3] for PWSCC growth rate according to the levels of dissolved hydrogen. It has been used from Section 7 of EPRI-MRP 263 NP, a numerical model describing the effect of hydrogen on PWSCC growth rate which takes the form of a Gaussian distribution centered at the Ni/NiO transition. This model is function of the difference in electrochemical potential (ΔECP) between the Ni/NiO transition and the test condition. Typical fitted parameters are the peak width and the peak to baseline ratio [2]. The SSRT did not include the stress intensity variation (ΔK), then it would be possible according to the CDTN data, to apply only the referred model from MRP-263 NP, now appealed "hydrogen ΔECP submodel". But it has been estimated the stress intensity factor applied for these tests: according to this, one has been possible to do some comparison with another technical recommendations from EPRI [4] and USNRC [5].

1.1. The Hydrogen ΔECP Submodel

The Sections 6 and 7 of EPRI-MRP 263 NP [2] treat about respectively mitigation of PWSCC initiation through elevated hydrogen content, and mitigation of PWSCC propagation through elevated hydrogen content. The conclusion of section 6 is based only in the Alloy 600 case, and is that according to the available data did not exist a hydrogen effect on PWSCC initiation in this nickel alloy: the case of Alloy 182 was not considered. But in Section 7, it was studied the hydrogen effect on Alloy 600, and Alloy 182 (here the interest case). The considered model described the effect of hydrogen on PWSCC propagation in the form of a Gaussian distribution centered at the Ni/NiO transition. This model is function of the difference in ECP between the Ni/NiO transition and the test condition (Equation (1)). Typical fitted parameters are the peak width (c) and the peak to baseline ratio (*P*). The concentration of hydrogen corresponding to the potential at the Ni/NiO transition is temperature dependent (Equations (2) and (3)).

$$CGR = CGR_{\max}\left[\left[\frac{1}{P}+\frac{(P-1)}{P}\exp\left(\left(-0.5\left(\frac{\Delta ECP_{Ni/NiO}}{c}\right)^2\right)\right)\right] \quad (1)$$

$$\Delta ECP_{Ni/NiO} = 29.58\left(\frac{T_{ref}+273.15}{298.15}\right)\log\left(\frac{[H_2]}{[H_2]_{Ni/NiO}}\right) \quad (2)$$

$$[H_2]_{Ni/NiO} = 10^{(0.0111Tref-2.59)} \quad (3)$$

where: *CGR* is the crack growth rate*, CGR_{max}* is the maximum CGR at the Ni/NiO transition, *P* is the ratio of the maximum to minimum expected CGR, *c* is the peak width, $\Delta ECP_{Ni/NiO}$ is the ECP difference from the Ni/NiO transition, T_{ref} is the reference temperature (in Celsius degrees) in test condition, $[H_2]$ is the hydrogen concentration on environment, $[H_2]_{Ni/NiO}$ is the $[H_2]$ on the Ni/NiO transition.

On the Table 7-2 of EPRI-MRP 263 NP [2] are presented model parameters for Alloy 182 data sets from various authors, as well as its average and standard deviation values. On the Figures 7-7 to 7-10 of EPRI-MRP 263 NP are presented graphics CGR *vs.* $\Delta ECP_{Ni/NiO}$ departing from raw data for Alloy 182 from researchers Andresen and Toloczko [2].

1.2. The MRP-115 Model

The MRP-115 is a technical recommendation which has been done with the objective for a qualified equation for CGR to evaluate defects found by in-service inspection. It is applied to wrought Alloy 600 base material, and also to its weld metals, the Alloys 182, 82, and 132. The nuclear components included the primary circuit welds with high residual stresses and in some J-groove welds attaching control rod drive mechanism (CRDM) and bottom mounted instrumentation (BMI) nozzles to the reactor upper head. The weld metals are by definition "as-cast" structures and, consequently are much more inhomogeneous than wrought materials. This characteristic generates data scattering due to the variations on the microscopic scale of weld metals. So, the simple multiplication factor approach is not suitable for extensive use, and necessitated the development of a more sophisticated methodology. The main model which would be applicable to the CDTN data is given by equation (4) [4].

$$CGR = \exp\left[-\frac{Q}{R}\left(\frac{1}{T}-\frac{1}{T_{ref}}\right)\right]\alpha\, f_{alloy}\, f_{orient}\, K^{\beta} \quad (4)$$

where: *CGR* is the crack growth rate at temperature *T, Q* is the thermal activation energy for crack growth, *R* is the universal gas constant, *T* is the absolute operating temperature at location of crack in the test condition, T_{ref} is the absolute reference temperature at location of crack in test condition, α is a power law constant, f_{alloy} is a constant value depending on material, f_{orient} is a constant value depending on the dendrites solidification direction in the weld, *K* is the crack tip stress intensity factor, *β* is a proper exponent.

1.3. The USNRC Model

The USNRC model is extracted from a report which has been done with the objective to study the CGR results and related metallography for field and laboratory specimens of Alloy 600 and its weld alloys tested in PWR environments. This report also has been presented CGR results for a shielded-metal-arc weld of Alloy 182 in a simulated PWR environment as a function of temperature between 290°C and 350°C. These data were used to determine the activation energy for crack growth in Alloy 182 welds. The tests were done by measuring the changes in the CGR as the temperatures were varied during the tests. The difference in electrochemical potential between the specimen and the Ni/NiO line was kept constant at each temperature by adjusting the hydrogen overpressure. The model has been extracted from the normalization for the nickel weld data to study the use of different activation energies, and is showed in equation (5) [5].

$$CGR_{Ni-weld} = 1.1\times10^{-12}\exp\left[-\frac{Q}{R}\left(\frac{1}{T}-\frac{1}{T_{ref}}\right)\right]K^{1.6} \quad (5)$$

where: $CGR_{Ni\text{-}weld}$ is the nickel-weld crack growth rate at temperature *T*, and the other symbols are the same as in equation (4). Note also that equation (5) is basically the same equation (4).

2. MATERIAL AND METHODS

The original study of Lima [3] comprised: a) Dissimilar material weldment from Angra reactor #1 reproduction in CDTN mechanical workshop, according to the ASME Boiler and Pressure Vessel Code – Section IX, Welding and Brazing Qualification, and the AWS specifications to the welding electrodes: this weldment has been done between two plates of AISI 316L and ASTM A-508 filled with Alloy 182 in a chamfer previously buttered with Alloy 82; b) Chemical-mechanical-structural characterization of weld and related materials, according to ASTM E4, and ASTM E8; c) Evaluation of the corrosion potential of the Alloy 182 at high-temperature; d) Obtaining and characterization of oxide passive film formed in Alloy 182 on primary water environment at high-temperature; e) SSRT with Alloy 182 specimens at different levels of dissolved hydrogen in the test environment, according to ASTM G 49, and ASTM G 129.

The materials are basically done at Table **1**, divided in base (two upper rows) and weld filler metals [3].

The basic mechanical properties of the main studied material, the Alloy 182 are given at Table **2** [3].

The quantification of the PWSCC brittle fracture surface and its depth was obtained departing from the scanning electron microscope (SEM) micro fractographies of the tested specimens: the images

Table 1: Chemical Composition of the Base and Filler Metals (wt%) [3]

	C	Mn	Si	P	S	Cr	Ni	Nb	Ti	Mo	Fe
AISI 316L	0.023	1.46	0.475	0.021	0.003	16.7	9.8	0.02	0.03	2.10	Bal.
ASTM A508	0.21	1.34	0.227	0.005	0.003	0.09	0.68	0.002	0.001	0.51	Bal.
Alloy 182	0.05	6.16	0.341	0.01	0.009	14.3	70.3	2.07	0.049	0.24	Bal.
Alloy 82	0.04	3.4	0.141	0.012	0.005	18.9	73	2.47	0.25	0.16	Bal.

Table 2: Basic Mechanical Properties of the Filler Metal Alloy 182 [3]

	Temperature (°C)	Yield Strength $YS_{0.2\%}$ (MPa)	Ultimate Tensile Strength (MPa)	Percentual Elongation (%)
Alloy 182	25	400±40	620±12	44±3
Alloy 182	325	328±28	500±30	47±3

were processed by a Quantikov equipment, and the evaluation of the CGR according to a method developed by Totsuka *et al*., quoted in the references of Lima [3]: see also pp.63-64 from this study for more information.

The main results of Lima study, concerning the value to modeling data, according to EPRI-MRP 263 NP [2], were the Tables **3** and **4** below reproduced from this: these Tables correspond graphically to the originals Figure 4.38 (page 100), and Figure 4.37 (page 99) from the study of Lima [3]. In Figure **1**, it is reproduced the original Figure 4.38.

In the Table **3**, E_{cor} represents the corrosion potential, and $\Delta ECP_{Ni/NiO}$ is the electrochemical potential (ECP) difference from that of the Ni/NiO transition: this is a very important parameter, because changes in the hydrogen concentration in primary water enforce that the corrosion potential reaches the Ni/NiO transition line in the Potential versus pH equilibrium diagram between Ni and water, and have a strong influence in the stress corrosion cracking behavior. In the Table **4**, A_{IGSCC} is the brittle fracture surface area ratio to the total fracture surface area, remembering that IGSCC is the intergranular stress corrosion cracking, the predominant type of PWSCC in this case.

Table 3: Resulting Values of E_{cor} and $\Delta ECP_{Ni/NiO}$ to Alloy 182 in PWR Primary Water at 325°C [3]

Test Environment	E_{cor} (mV_{SHE})	$\Delta ECP_{Ni/NiO}$ (mV_{SHE})
2 cm^3 H_2 (STP)/kg H_2O	-717	-18
10 cm^3 H_2 (STP)/kg H_2O	-735	0
25 cm^3 H_2 (STP)/kg H_2O	-756	21
50 cm^3 H_2 (STP)/kg H_2O	-776	41

Table 4: Crack Growth Rate (CGR) to Alloy 182 in PWR Primary Water at 325°C [3]

Dissolved Hydrogen	Crack Depth (mm)	A_{IGSCC} (%)
2 cm^3 H_2 (STP)/kg H_2O	0.836	14
10 cm^3 H_2 (STP)/kg H_2O	1.3	33
15 cm^3 H_2 (STP)/kg H_2O	1.18	25
25 cm^3 H_2 (STP)/kg H_2O	1.04	20
50 cm^3 H_2 (STP)/kg H_2O	0.573	6
Dissolved Hydrogen	Time to failure t_f (h)	CGR (mm/s)
2 cm^3 H_2 (STP)/kg H_2O	324	1.3×10^{-7}
10 cm^3 H_2 (STP)/kg H_2O	216	5.0×10^{-7}
15 cm^3 H_2 (STP)/kg H_2O	273	2.9×10^{-7}
25 cm^3 H_2 (STP)/kg H_2O	278	2.1×10^{-7}
50 cm^3 H_2 (STP)/kg H_2O	384	2.9×10^{-8}

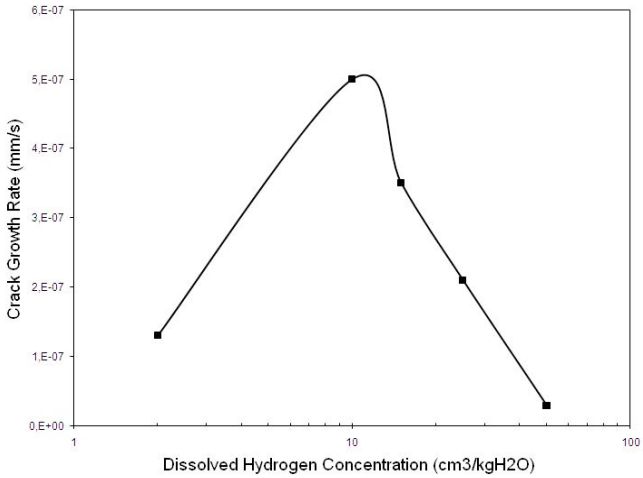

Figure 1: Experimental result for Alloy 182 SSR tested at 325°C in primary water with strain rate 3.10^{-7} s^{-1}, from Lima [3].

3. RESULTS

3.1. The Hydrogen ΔECP Submodel

The obtained result applying experimental CDTN data results on equations (1) to (3) is done by simple algebraic substituition, and the main model equation is a normal/gaussian distribution according to equation (6).

$$CGR = 1.7\left[\left[\frac{1}{17.2} + \frac{(17.2-1)}{17.2}\exp\left(\left(-0.5\left(\frac{\Delta ECP_{Ni/NiO}}{15.2}\right)^2\right)\right)\right]\right] \quad (6)$$

$(mils/day)$

3.2. The MRP-115 Model

The SSRT of CDTN were not performed with controlled stress intensity factor (K) variation, but it´s possible to estimate a K-value where probably the tests were done. To estimate it, one is departed from equation (7) which represents the K in mode I to a fully circumferential crack in a bar [6] (that is a simplification related to the actual case, because the axial crack depth is not completely constant).

$$K_I = (Y\sigma)\sqrt{\pi a} \quad (7)$$

where: σ= stress, a= crack depth, Y= factor which represents the total contribution of the primary and secondary stresses.

The considered values to be used in Equation (7) were: σ= 440 MPa, a= 1.04 x 10^{-3} m – both values corresponding to the case where the hydrogen concentration is equal to 25 cm^3 H_2/kg H_2O (see also Figure 4-24 from Lima, showing the stress versus strain curves obtained on SSRT at CDTN and the stress is the average stress of the SSRT applied to the specimen at 325°C), Y≈ 1, considering contribution of primary stress far greater than the contribution of secondary stress. Then, the found estimated K-value is 25.15 MPa √m.

So, it can be considered the point (CGR, K) obtained in CDTN test at 25 cm^3 H_2/kg H_2O and 25°C (see also Table **4**) equal to (2.1 x 10^{-10} m/s; 25 MPa √m). This test point can be plotted on the disposition curve of the MRP-115 [4] according to Figure **2**.

3.3. The USNRC Model

The resulting modeling is almost immediate when it has been substituted the K-value on equation (5), remembering that T=T_{ref}=325°C:

$CGR_{Ni\text{-}weld}$= 1.1 x 10^{-12} exp (0) $(25.15)^{1.6}$ = 1.9 x 10^{-10} m/s.

From Table **4**, it has found *CGR* for CDTN test at reference temperature equal to 2.1 x 10^{-10} m/s.

4. DISCUSSION

4.1. The Hydrogen ΔECP Submodel

Based on equation (6), one has been plotted in Figure **3**, the corresponding normal curve (in blue), and also plotted the red curve from Andresen raw data [2], as the closer in the MRP-263 NP relating to the CDTN data.

It can be noted that the peak/baseline ratio (P) in the CDTN case is greater than in the Andresen raw data case. Probably this is due to the different test methodologies used: CDTN used an accelerated test, the SSRT; despite in the Andresen raw data case the used tests were not identified (that is because the Andresen´ s references were deleted from the document, as "proprietary information"), normally this researcher uses the constant load test. But the "hydrogen ΔECP submodel" is applicable in both cases, because the Gaussian regression is valid to them. The differences are in the parameters (*P, c*) as showed in equation (1). Maybe could be interesting to include in the MRP-263 NP data sets of SSRT to be compared.

4.2. The MRP-115 Model

Although the CDTN data set was not performed considering K-variation, and the data sets in the MRP-115 [2] did not consider the hydrogen variation, it is possible to estimate a K value to the CDTN tests, and considering only one level of dissolved hydrogen (25

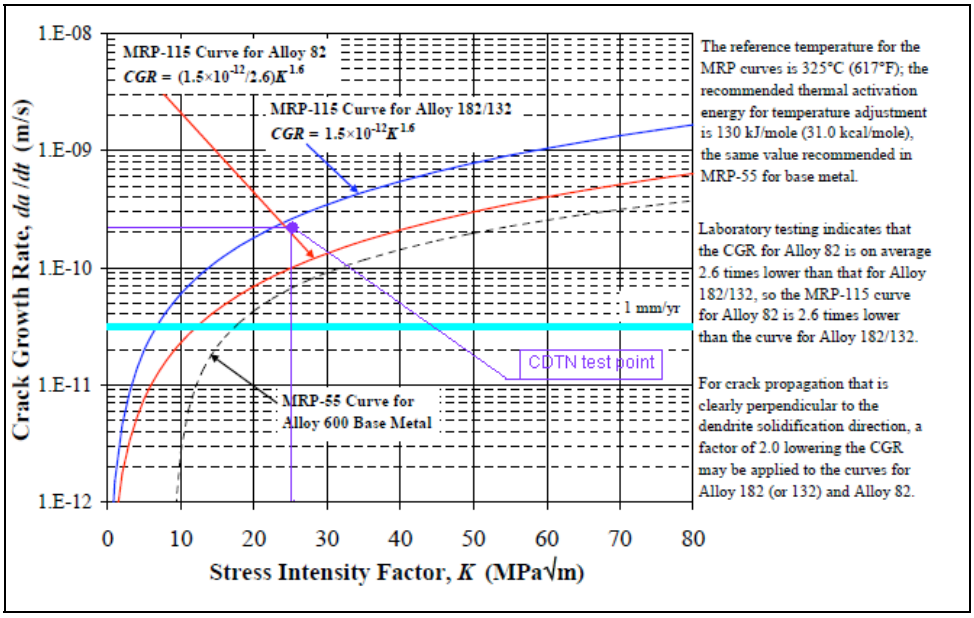

Figure 2: Plotting modeling to the CDTN test point (in purple) over the MRP-115 disposition curves to the Alloys 182/132 and 82: the original graphic is the Figure 4-6 from reference [4].

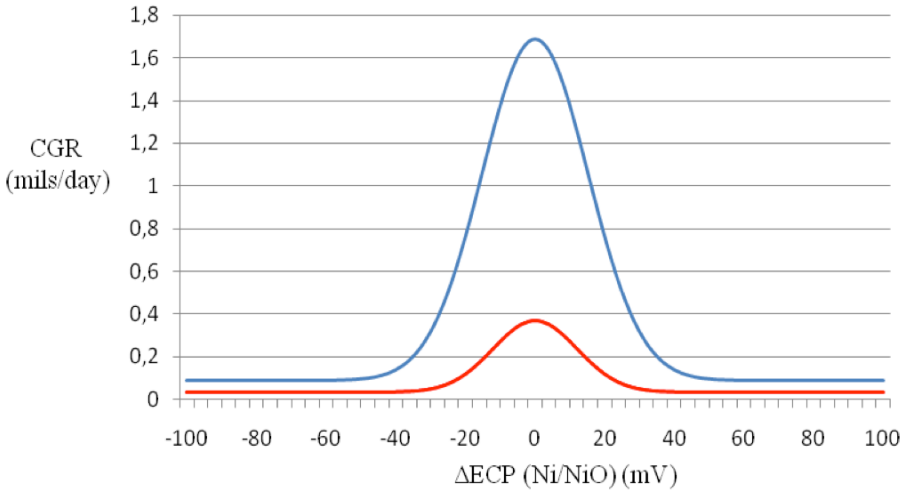

Figure 3: Equation (6) modeling fit to Alloy 182 CDTN data (blue) [3] compared to raw data from Andresen (red) [2]. Plotted through Microsoft Excel 2007.

cm^3 H_2 (STP)/kg H_2O) in the environment, to compare the obtained results. This result plotted in Figure **2** shows that the "CDTN test point" (in purple) at 325°C is close to the MRP-115 disposition curve (in blue) for Alloy 182/132 at 325°C. Naturally more tests with K-variation should be performed at CDTN laboratories to confirm the MRP-115 disposition curve modeling tendency, but only one estimated result so close to this curve that allows to expect, in a complete data set according to CDTN methodology added with the K-variation condition, an almost coincident regression according to MRP-115 and to CDTN.

4.3. The USNRC Model

The result of CDTN experimental CGR value compared to the correspondent modeling value according to the USNRC is about 9.5% higher than that. So, the modeling appears to be less conservative than the CDTN experimental methodology, and it seems to be easily explicable due to different test methodologies using in various data sets used to do the USNRC model.

5. CONCLUSION

This paper presented a modeling and modeling study of tests of primary water stress corrosion of Alloy 182, realized at CDTN installations and comprising the hydrogen ΔECP submodel, which relates the CGR with the dissolved hydrogen variation in the tested environment: the result showed a good qualitative adjustment, but different quantitative adjustment, due probably to different test methodologies used to obtain the data sets. Also it has been done a study considering a modeling comparison with the disposition modeling curve in the EPRI-MRP-115, and also in an USNRC Technical Report. Both results a good initial adjustment, but more tests with K-variation should be done at CDTN to produce complete regression curves according to these models. It should also include in the EPRI-MRP-263 NP more study concerning SSRT data sets.

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