A Comparative Study of the Effects of Methanol and Ethanol Solutions on the Bulk Etch Rate of CR-39

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1. Abstract

CR-39 is a plastic polymer detector used on the magnetic recoil spectrometer (MRS) diagnostic on the OMEGA laser system. Charged particles produced by neutrons emitted from fusion reactions disrupt chemical bonds within the plastic, leaving tracks. The detectors undergo a series of three chemical etching processes to reveal the tracks. The tracks are recorded using an optical microscope in conjunction with image processing software, which uses a coincidence counting technique to distinguish true tracks from background noise. This project investigates a new bulk etching technique using a methanol/NaOH solution with the goal of increasing the etch rate. Varied concentrations of methanol/NaOH solutions were tested. Using a 2.5 molar concentration of methanol/NaOH solution compared to the standard ethanol/NaOH solution increased the bulk etch rate from ~18 microns removed/hour to ~37 microns removed/hour. This promises to decrease the amount of time needed to process MRS and other data.

2. Introduction

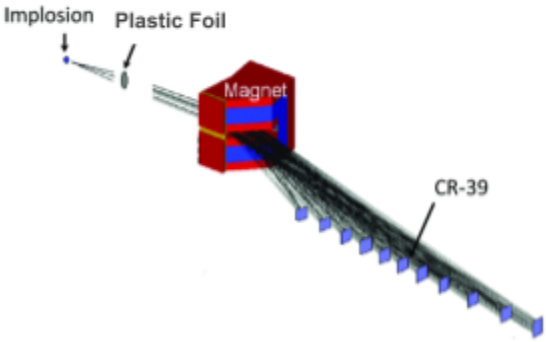

Figure 1: A schematic illustration of the magnetic recoil spectrometer (MRS). Neutrons emitted from the implosion hit the foil, producing charged particles that are selected to be momentum analyzed by an aperture in front of the magnet. The charged particles are then directed onto the CR-39 detectors located on the windows of the MRS.

A nuclear fusion reaction occurs when two light atomic nuclei fuse together to form a heavier nucleus with the release of energy in the form of particles. Although various isotopes of light elements can be paired, the Laboratory for Laser Energetics (LLE) uses the hydrogen isotopes deuterium (D) and tritium (T) to produce the most energetically efficient results. Inertial confinement fusion (ICF) is a method implemented at LLE that uses high-power lasers to uniformly irradiate a cryogenic capsule containing DT fuel.[1] The energy from laser beams ablates the surface of the capsule and causes an implosion, which means the inner fuel quickly collapses inward. As a result, the target is compressed and heated to the extreme temperature and pressure necessary to initiate the DT reaction. When the interior fuel undergoes fusion, helium is formed and a high energy neutron is released.

The magnetic recoil spectrometer (MRS) on the OMEGA laser system shown in Figure 1 consists of three main parts: a plastic foil, a focusing magnet, and an array of CR-39 plastic detectors.[2] Neutrons emitted from an implosion hit the plastic foil and produce recoil charged particles (deuterons or protons). An aperture in front of the magnet selects some of the particles to be momentum analyzed and focused onto the CR-39 detectors.[3] Although the neutrons are generated at a fixed energy, the energies with which they reach the MRS vary. The energies of the recoil particles vary correspondingly. The recorded energies thus indirectly create a neutron spectrum that can quantify the energy lost between neutron production and the detectors.[4] Almost all the energy loss is caused by collisions between ejected neutrons and particles of the unfused fuel between the DT fuel core of the implosion and the detector. This energy loss depends on the fuel areal density (product of density and radius), which is a measure of the radial compression and

determines how much fuel was fused.[5] Measurements of fuel areal density from cryogenic DT implosions are essential to the National Ignition Campaign of achieving ignition. Ignition is attained when the output of energy from a fusion reaction exceeds the input of energy used to create fusion conditions. Thus, CR-39 detectors provide a means to quantify the success of individual reactions.

CR-39 is a special type of plastic (polyallyl diglycol carbonate) designed to be clear with high optical quality. When charged particles pass through the sensitive material, they leave molecular damage that can be observed and recorded under a digital microscope using MIT's charged particle spectroscopy (CPS) scanning program. The Laboratory for Laser Energetics receives CR-39 detectors from Track Analysis Systems Ltd in England. During manufacturing, the plastic is compromised by defects known as background noise or noise pits. Other detector manufacturers also produce noise-pitting CR-39, which means that background noise is not specific to Track Analysis Systems Ltd. Although the CPS scanning program is usually able to recognize valid data points, analyzed noise pits may show similar eccentricity and contrast to ejected particles.[6] To reduce background noise, irradiated CR-39 detectors go through a successful coincidence counting technique (CCT) that takes advantage of the fact that particles penetrate deep within CR-39, while noise pits only show up on one surface or the other.

The CCT uses a standard 6 N NaOH track etch, a bulk etch, and a final 6 N NaOH track etch, along with track microscope scans and analysis. Figure 2 shows the series of etches and their effects on CR-39. In the first standard track etch, the detectors are placed in a hot water bath at 80°C in a beaker containing 6 N NaOH. The chemical etchant infiltrates the broken chemical bonds in the irradiated plastic material, opening the tracks of particles and background noise. After six hours, the detectors are removed from the hot water bath, dried, and placed on the stage of an optical microscope for the pre bulk etch (PRBE) scan. The CPS scanning program records the surface of the CR-39, which can take 1.5 hours to over a day to complete.

In the bulk etch, the detectors are placed in a hot water bath at a temperature of 55°C in a beaker containing 2 M ethanol/NaOH. The aggressive chemical etchant removes the bulk of the material and erases all the opened particle and noise tracks. After 12 hours, the detectors are removed, dried, and measured before reopening the tracks in the final standard track etch.

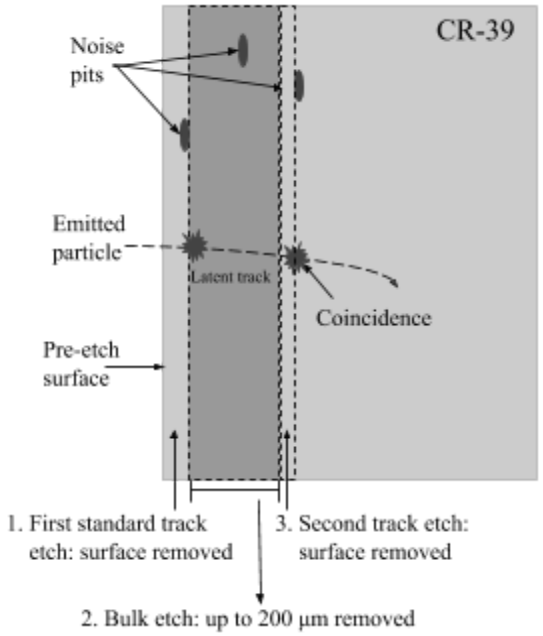

Figure 2: A schematic drawing of the three-series etch process of CR-39 used in the coincidence counting technique (CCT). Emitted particles leave tracks of molecular damage that travel deep within the plastic as opposed to the noise pits that appear sporadically throughout. The standard NaOH track etch reveals the trails. The bulk etch removes much of the bulk of the material and erases the trails. The second track etch reopens the trails. The points that coincide with each other before and after bulk etching are valid data.

The second standard track etch follows the same procedure as the first standard track etch. Figure 3 shows a generalized track profile of molecular damage in the plastic after the two etch processes occur. A conical pit forms in the place of both embedded background noise and true data points on the etched surface.[7]

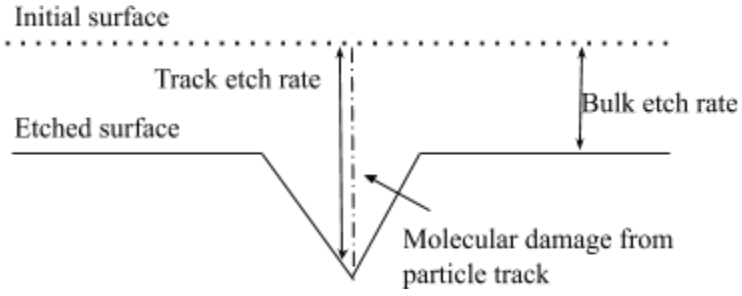

Figure 3: A generalized track profile. During the standard track etches, the solution infiltrates the molecular damage and opens the particle track. During bulk etching, the aggressive etchant removes the bulk of the material. The simultaneous action of the two etches in which the track etch rate is greater than the bulk etch rate results in a conical pit.

The final step in the time-consuming CCT is the CCT scan, which can take 1.5 hours to over a day to complete. The reopened tracks are again recorded under an optical microscope with the CPS scanning program. As shown in Figure 4, the surface of CR-39 from the PRBE scan is aligned and compared with the CCT scan. The points that coincide with each other before and after are true data points because noise has been eliminated.

In this study, a different chemical etchant, methanol/NaOH solution, was chosen to improve the bulk etch technique and decrease the data processing time. While the hot water bath was maintained at 55°C, the concentration of the solutions was manipulated to investigate if the methanol/NaOH solution has a faster bulk etch rate and produces the same quality of CCT data as the ethanol/NaOH solution.

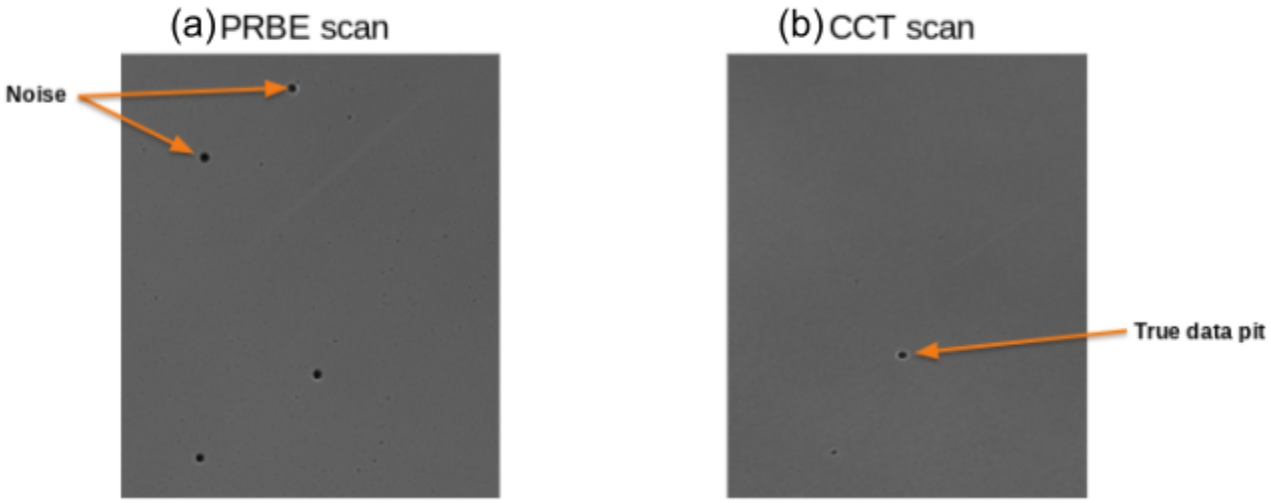

Figure 4: Pre bulk etch (PRBE) and CCT images of a CR-39 surface under an optical microscope after standard track etches. The noise on image (a) appears similar to the true data pits and would be identified as a true data pit by the scanning program. On image (b), the noise has disappeared and only a true data pit remains.

3. Methods and Materials

Prior to the present experiment, the detectors had been subjected to the first standard track etch. They were placed in a 6 N NaOH standard track etch at 80°C. After six hours, they were removed from the hot water bath, dried, and placed under an optical microscope that recorded an image of the CR-39 surface.

After the first standard etch, a micrometer was used to measure the thickness of each CR-39 detector at eight locations. The measurements were averaged and recorded to ensure that ~180 to 200 microns of plastic material would be removed during bulk etching. If less than 180 microns of bulk were removed, the noise pits seen in the first scan would not be eliminated. If more than 200 microns of bulk were removed, then all the particle tracks would be lost.[8]

In the present experiment, the bulk etch rates of four irradiated CR-39 samples were tested in 2 M ethanol/NaOH solution and compared to twenty irradiated CR-39 samples tested in methanol/NaOH solutions ranging from 1 M to 4 M. These chemical etchants were prepared in beakers using 10 N NaOH. The beakers were placed in a hot water bath maintained at a constant temperature of 55°C and covered to prevent evaporation.

Every few hours, the detectors were removed from the hot water bath and the thickness of each sample was measured at eight locations. The average thickness measured was subtracted from the initial average thickness to determine how much bulk was removed. The samples continued to undergo bulk etching until the average bulk removed was between 180 and 200 microns. Then, the bulk etch rate was calculated by dividing the average thickness of the material removed by the number of hours it took to bulk etch. The averages were plotted on a graph (Figure 5) with x-values representing time in hours and y-values representing the bulk etch rate in microns removed/hour. Figure 6 compares the bulk etch rate using various concentrations of methanol/NaOH solutions. Overall, a 2.5 M methanol/NaOH solution reduced the approximately ten-hour procedure using the 2 M ethanol/NaOH solution to under six hours.

Although the efficiency of bulk etching nearly doubled, the final steps of the CCT were executed to ensure that the methanol/NaOH solution produced the same quality of data as the ethanol/NaOH solution.

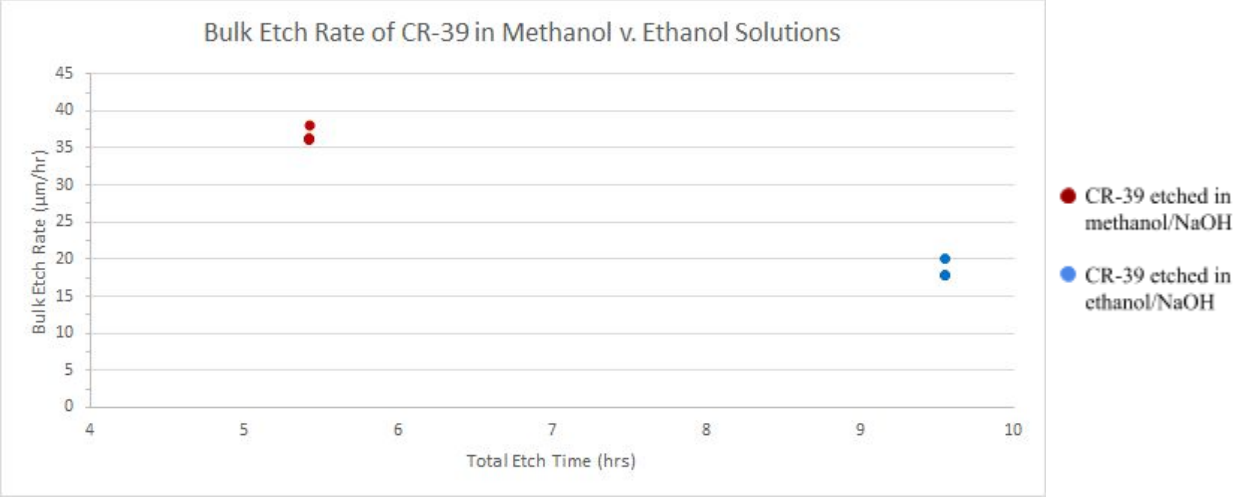

Figure 5: Scatterplot of the bulk etch rate of CR-39 comparing methanol/NaOH solutions and ethanol/NaOH solutions. Time is plotted on the x-axis and the bulk etch rate is plotted on the y-axis. The bulk etch rate of CR-39 in methanol/NaOH is about twice as fast as it is in ethanol/NaOH.

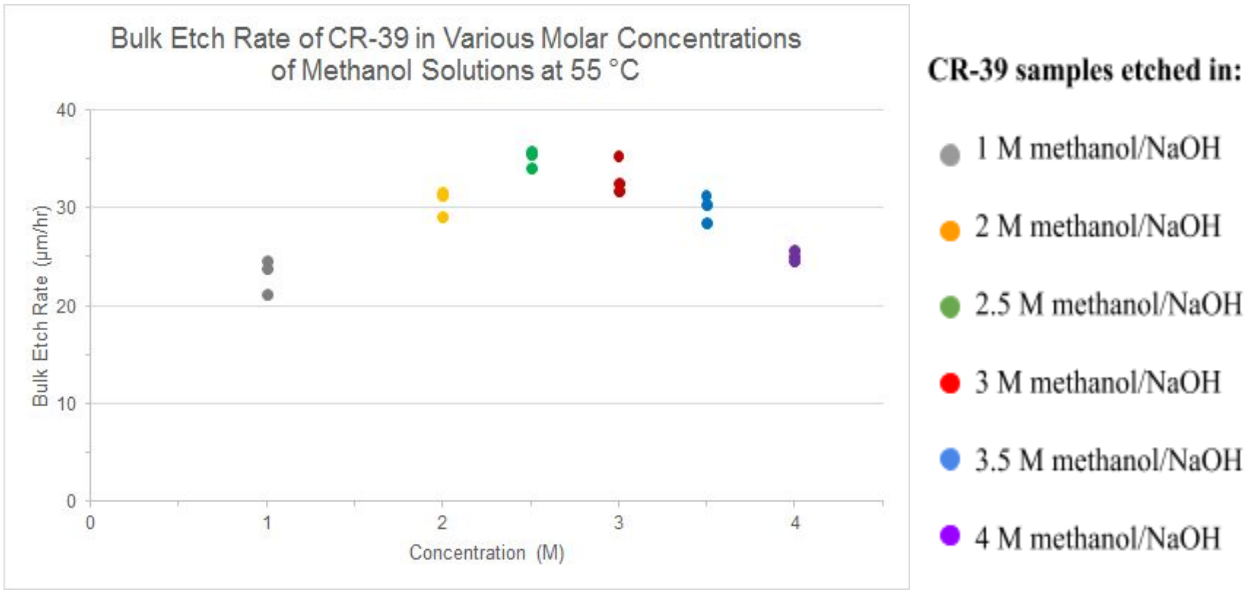

Figure 6: Scatterplot of the bulk etch rate of CR-39 in methanol/NaOH solutions. Molarity is plotted on the x-axis and bulk etch rate is plotted on the y-axis. The bell-shaped curve shows that the fastest bulk etch rate was ~37 microns removed/hour at a molar concentration of 2.5 methanol/NaOH.

4. Results

After completing the CCT, the methanol/NaOH solution produced comparable particle track results to the ethanol/NaOH solution. Figure 7 shows the track counts from PRBE scans and CCT scans of methanol and ethanol etches. The total track counts from ethanol etches compared to methanol etches is greater because the total neutron yield was higher on those implosions. After analyzing the scans, the difference in tracks between PRBE and CCT scans of methanol etches is comparable to the difference in tracks between PRBE and CCT scans of ethanol etches. The ethanol bulk etches have been shown to work with accelerator exposures to a known number of protons.[9] One specific shot (94008) shows a greater track count in the CCT scan than in the PRBE scan, which is most likely due to the background noise being over-subtracted in the PRBE analysis. Separating tracks from background noise is not always straightforward, which is why there is not a perfect agreement between the PRBE and CCT analysis.[10]

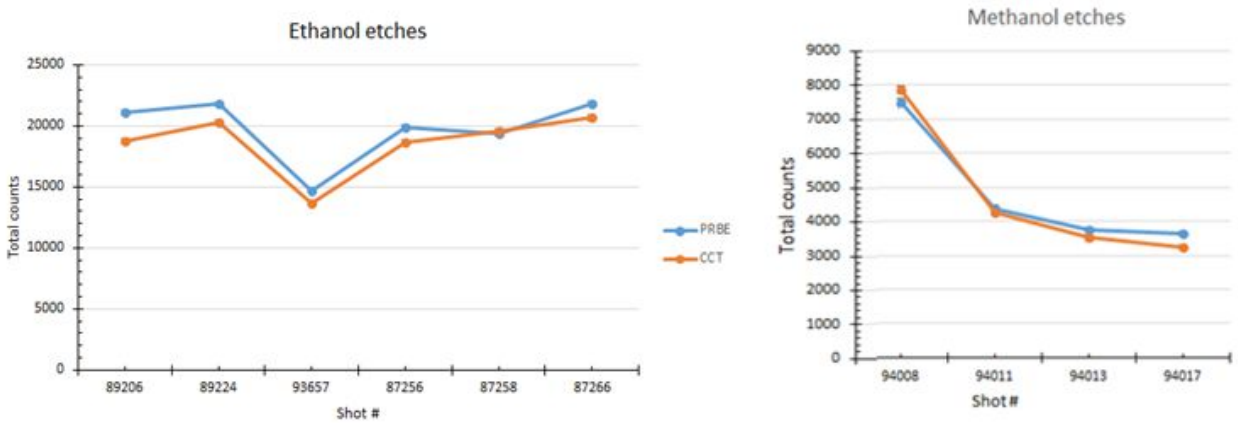

Figure 7: A comparison of the total counts of tracks from PRBE and CCT scans between methanol/NaOH bulk etches and ethanol/NaOH bulk etches. The implosion shot number is plotted on the x-axis and the total count of tracks is plotted on the y-axis. Methanol/NaOH bulk etches reproduce the PRBE and CCT track number estimate trend seen on the ethanol etch graph.

The two contour plots that are shown in Figure 8 graph the number of tracks as a function of diameter and contrast. Based on previous experience with CR-39 response, the tight region highlighted in red is a cluster of tracks with similar diameter and contrast identified as the signal deuterons. Because the signal characteristics are very similar for the two etch scenarios, the methanol etch does not negatively impact CR-39 response. Other contour clusters, such as the cluster stretching from ~10-85 in contrast at small diameters, represent noise tracks in the CR-39. A negative result would show fainter tracks (higher contrast) or smaller tracks (lower diameter), which would have been harder to separate from background tracks in the analysis. This allows us to be confident that the methanol/NaOH does not anomalously fade or shrink tracks.[11]

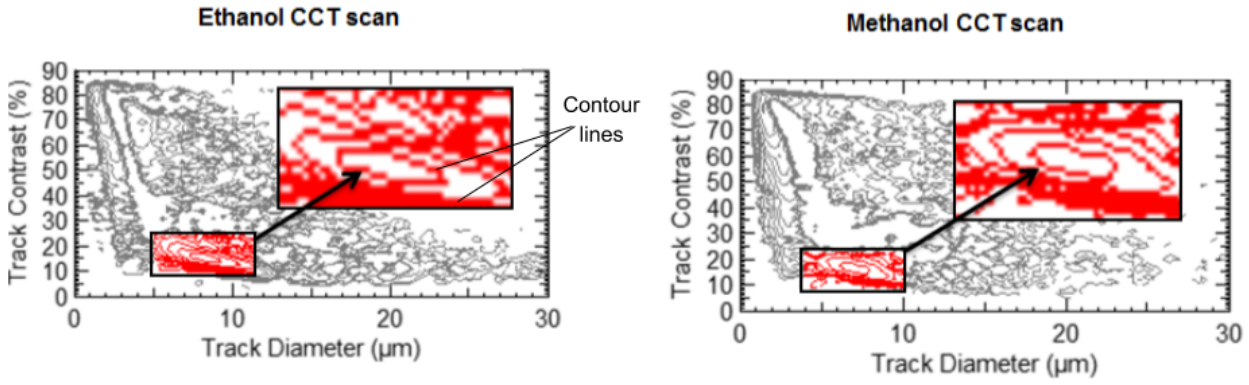

Figure 8: Contour plots of track counts comparing ethanol/NaOH and methanol/NaOH CCT scans. Track diameter is plotted on the x-axis and track contrast is plotted on the y-axis. The areas boxed off in red are the signals picked up from the CPS program. The signal characteristics (track contrast and diameter) are comparable post ethanol and methanol bulk etches, which shows that methanol/NaOH does not negatively impact CR-39 response.

5. Conclusion

In this project, the etching properties of methanol/NaOH solutions have been investigated and compared to the standard 2 M ethanol/NaOH solution. The goals were to verify that methanol/NaOH etchants have a faster bulk etch rate than the standard ethanol/NaOH etchant, find the optimal concentration of the methanol/NaOH etchant that yields the fastest bulk etch rate, and determine if the methanol/NaOH etchant produced the same quality of CCT data as the ethanol/NaOH etchant. Originally, the standard 2 M ethanol/NaOH solution at 55°C had a bulk etch rate of ~18 microns removed/hour with a total etch time of ~10 hours. It was found that a 2.5 molar concentration of methanol/NaOH solution at 55°C more than doubled the efficiency of bulk etching to ~37 microns removed/hour. According to Figures 7 and 8, the data from ethanol etches were comparable to the data from methanol etches. Based on the evidence from this study, the methanol/NaOH bulk etch decreased the amount of time to process data and has replaced the ethanol/NaOH bulk etch as the preferred CR-39 processing method.

6. Acknowledgments

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7. References

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