Original Paper

Investigation of selenium retention in high redox glasses

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Laboratory glass melting experiments were made to study the effect of glass redox on selenium retention. Initially, it was found that as expected selenium retention decreases as glass redox ratio increases. But, unexpectedly, selenium retention then levels off and reaches a minimum upon further increases in glass redox, before once again increasing for very high redox ratios of more than 0.60. Selenium concentrations in the furnace atmosphere at different locations in a commercial float glass furnace were also measured as an indicator of selenium volatility at different stages of glass melting. A study of the effect of glass redox ratio on the spectral absorption of the major selenium containing coloring centers in float glass was also made.

1. Introduction

Float glass products are manufactured today in a wide range of tints for use in both the automotive and architectural industries mainly for reasons of either improved solar absorption or aesthetics. Recently, there has been increased interest in achieving better solar control properties while still maintaining a neutral glass color, which generally requires the development of new glass compositions that have high iron redox ratios (highly reduced) to exploit the better infrared absorbing properties associated with greater Fe^{+2} contents.

For example, various shades of green, blue, brown and gray are now common tints that are manufactured by adding Fe_2O_3 plus different coloring agents, which very often include selenium. Unfortunately, selenium is highly volatile at the glass melting furnace temperatures used in the flat glass manufacturing processes, and typically only about 15 to 20 % is retained in the final glass product. Literature references have reported that neutral or mildly oxidizing conditions are necessary for good selenium retention [1 to 2]. However, this poses a potential conflict when attempting to make better heat absorbing glasses that have high redox ratios. Several float glass producers have investigated methods to further improve selenium retention [3 to 5]. Others have proposed treatment of the furnace exhaust gas to address the environmental issues from selenium emissions [6].

There are many other factors that can affect selenium retention rates during glass melting, including batching considerations such as the type of selenium compound employed [7 to 13] (for example, prereacted materials or high selenium containing glass frits), the use of and amount added of either oxidizing (such as sodium nitrate, sodium sulfate, etc.) or reducing agents (coal, silicon, etc.) to the glass batch [8 and 14], the size of elemental selenium granules, and the cullet content of the batch, whether or not the cullet contains selenium.

Furnace operating conditions can also affect selenium retention rates; these conditions include total melting times [7 and 15], furnace temperatures [7 and 15], and the partial pressure of oxygen in the furnace atmosphere [2, 16 and 17]. Also, selenium can exist in at least four different valence states in glass. Two major coloring species in glass that contain selenium have been previously identified [18 to 22]. They include a pink color from an atomic dispersion of selenium, and a brown color from Se^{-2} due to the formation of iron-polyselenide coloring centers [21]. Several studies have also reported on the redox chemistry between the four selenium valence states in glass [1, 23 and 24].

This paper will investigate the impact of several of the above factors on selenium retention rates, especially the role of glass redox ratio in the presence of appreciable iron oxide in glass. Three key areas were investigated as follows: a) laboratory glass melting experiments using small crucible melts, b) atmosphere sampling tests made in a commercial glass melting furnace, and c) determining any changes in the intensity and spectral shift for the major iron-polyselenide absorption peak, which normally occurs at about 480 nm, due to high glass redox.

2. Laboratory glass melting experiments

Numerous laboratory crucible glass melts of a standard float glass composition were prepared under controlled melting conditions to investigate the effect of glass redox on selenium retention. Typically, 700 g glass melts were made from commercially available batch materials.

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The experimental procedure consisted of melting the mixed batch in silica crucibles for 2.5 h in a gas fired furnace with about 2 % excess oxygen at a peak temperature of 1454 °C, followed by fritting in water and then remelting the dried frit using platinum crucibles for an additional 2 h at 1454 °C in an electric furnace in air. The samples were then cast on a metal table and annealed for about 1 h at 593 °C and then cooled to room temperature at the furnace rate. Test samples, about 38.1 mm in diameter, were ground and polished on both surfaces to about 4.06 mm thickness for spectral solar transmittance measurements and for chemical analysis by X-ray fluorescence (XRF) spectroscopy.

In this paper, glass redox, expressed as the iron redox ratio, is defined as the wt% FeO to total iron oxide wt% as Fe_2O_3. The glass redox ratio was calculated by dividing the wt% FeO content in the glass sample (as measured from the spectral absorption at 1000 nm with a Perkin Elmer Lambda 9 spectrophotometer) by the total wt% Fe_2O_3 content in the glass as determined by XRF spectroscopy (Rigaku 3370 X-ray fluorescence spectrometer). Previously, the spectral absorption at 1000 nm was independently calibrated against a wet chemical technique for the quantification of FeO content in glass. The selenium retention was determined from the actual ppm Se content analyzed in the glass by XRF and the measured amount initially added to the glass batch with an accuracy of ±0.5 ppm.

Tests were made varying the amount of elemental selenium that was added to each crucible melt. Other tests were made by varying the amount of coal added to the glass batch to alter the glass redox ratio of the crucible melts. Usually, selenium was added to the melts as elemental selenium. However, several tests were also made using various selenium containing compounds (selenites, selenides, etc.), different oxidizing agents (nitrates of different metals), and also different glass cullets or frits, which contained different amounts of selenium. Other experiments were made either varying the amount of salt cake (Na_2SO_4) or $NaNO_3$ addition and also the size of the silica crucible (either 100 or 150 mm in diameter) to determine the effect of melting surface area on selenium retention. In these laboratory melts, selenium acts as a very strong oxidizing agent; therefore, unless large amounts of coal were added, glasses with high selenium concentrations inherently had low glass redox ratios.

However, upon further increasing both the amount of selenium and coal added to the glass batch, it was found that the selenium retention first decreased as the glass redox ratio increased. It then leveled off and reached a minimum level before it increased again as indicated in figure 1. This graph shows the percent selenium retained for different glass redox ratios. It indicates that there is a relatively large region for glass redox ratios ranging from about 0.35 to 0.60 where the selenium retention rate is essentially flat at a minimum value of about 3 to 5 %. For either more oxidizing conditions (redox ranging from 0.20 to 0.35) or more reducing conditions (redox > 0.60) the selenium retention rates are significantly higher. The shape of the retention curve in figure 1 relates to the different valence states for selenium that are present in the glass. The redox behavior of selenium has been previously determined from the chemical reaction constants according to the results reported in [9 and 24]. For oxidized glasses, selenium is found as either Se^{+4} ions or Se^{+6} ions, while for reduced glasses, selenium is present as Se^{-2} ions. Selenium ions, whether positively or negatively charged, form relatively strong chemical bonds in glass and are thus chemically more stable than elemental selenium, which is present in glass as an atomic dispersion of selenium in an intermediate redox range [17, 25 and 26]. In addition, reduced selenium exists in our samples even before the second rise in the retention curve, as evidenced by the presence of the selenide absorption peak in the spectral curve for glasses in the redox ratio range corresponding to the nearly flat region of the retention curve as discussed later.

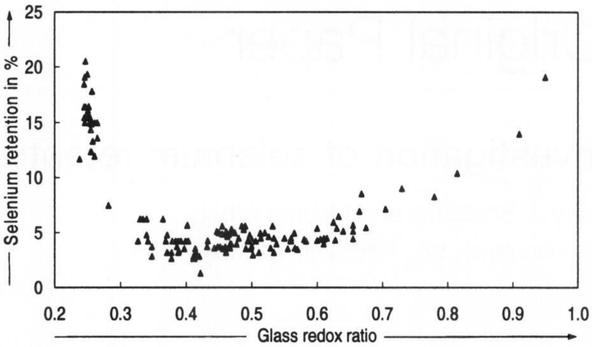


Figure 1. Effect of glass redox ratio on selenium retention in laboratory melts.

Similarly, the redox behavior of sulfur was also investigated. Figure 2 shows the effect of glass redox ratio on sulfur retention for the same set of laboratory melts shown in figure 1. Note a similar shape for both the selenium curve and the sulfur curve, although the minimum occurs at a higher redox ratio for sulfur than for selenium. The effect of glass redox ratio on sulfur retention in silicate glasses was previously reported [27]. Since both selenium and sulfur belong to the same column in the periodic chart, it could be expected that selenium behaves very much like sulfur in glass as a function of redox. As evidenced by the spectral absorption data discussed below, selenide formation occurs at lower redox ratios than does the sulfide. This early formation of reduced selenium likely contributes to the occurrence of the minimum in its retention curve at a correspondingly lower redox ratio.

Typical tinted glasses that are produced on large float glass melting furnaces have iron redox ratios of about 0.25 to 0.30. Therefore, most float glass compositions fall on the left side (oxidized part) of the curve in figure 1. As a conse-

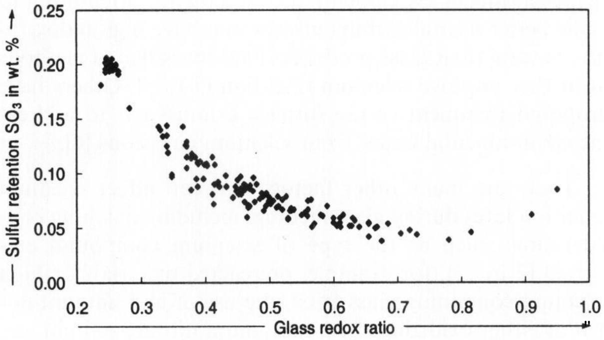


Figure 2. Effect of glass redox ratio on sulfur retention for laboratory melts.

quence, there is the potential for significant process variability in selenium retention during float glass production due to the steeper slope on that part of the curve. This requires close monitoring and very good process control of the float glass melting furnace operations to maintain a constant glass redox ratio, otherwise wide fluctuations in selenium retention can occur that will cause unacceptable changes in glass color. After many years of experience gained while making selenium-containing glasses and by implementing new technical advances that provide improved process control, float glass manufacturers are now able to properly control glass color.

Selenium retention is not entirely determined by the redox ratio variations. For example, the volatilization of elemental selenium from the unreacted batch is one possible source of loss. On the other hand, SeO_2 is the most volatile selenium compound. Thus, if selenium is oxidized before its incorporation in the melt, then its loss is more likely. Therefore, various processes including melting and evaporation, and oxidation of selenium particles followed by SeO_2 volatilization can all be competing to prevent selenium retention. In addition, the sweeping effect of the gaseous species resulting from batch decomposition and other melting reactions will result in additional loss of selenium. The effects of these processes are, in addition to glass chemistry, dependent on the melting processes, which change greatly from laboratory melting to the continuous melting in large furnaces.

3. Furnace atmosphere sampling tests

Because of the low selenium retention rates that normally occur during float glass melting, several experiments were made to measure where the majority of the selenium losses occur in the continuous glass melting process in a production furnace. The tests were performed in a conventional float glass furnace (using overhead air-gas firing) while producing a selenium-containing high redox glass composition. A standard furnace atmosphere sampling technique (EPA Method 4 [28]) was used to determine the concentration of selenium in the atmosphere inside the melting tank at four different locations. The results are given in figure 3, which shows that the selenium concentration in the atmosphere above the glass melt as reported in ppm Se is much greater for the areas where the unmelted batch first enters the furnace (shade cooler, Port 0) than further down tank where the batch has been converted to molten glass (Port 5). This indicates that most of the selenium loss readily occurs during the initial batch melting stage rather than from the molten glass surface, even though glass furnace temperatures are typically higher in Port 5. Oxidation of metallic selenium leading to the formation of the highly volatile SeO_2 compound (which is gaseous down to 300 °C), and the very low boiling point of 685 °C for elemental selenium are possible reasons for the early losses. Under similar furnace conditions, we observed more than a ten times reduction in selenium retention when the glass redox was increased from, e.g., 0.28 to above 0.5. Therefore, higher glass redox also likely elevated the early selenium losses in the float glass furnace. These data suggest that new methods are needed to further improve selenium retention rates including adding high selenium containing cullet, other selenium compounds that have lower vapor pressures (such as $ZnSeO_3$, FeSe, etc.) or better temperature and excess oxygen control in the early ports of the melting furnace. For example, when adding other compounds such as $ZnSeO_3$, its dissociation will then govern the selenium loss from the batch.

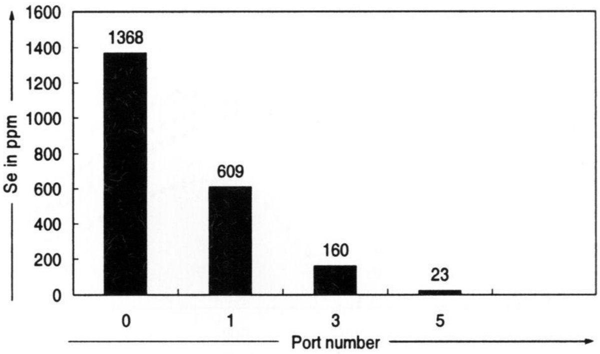
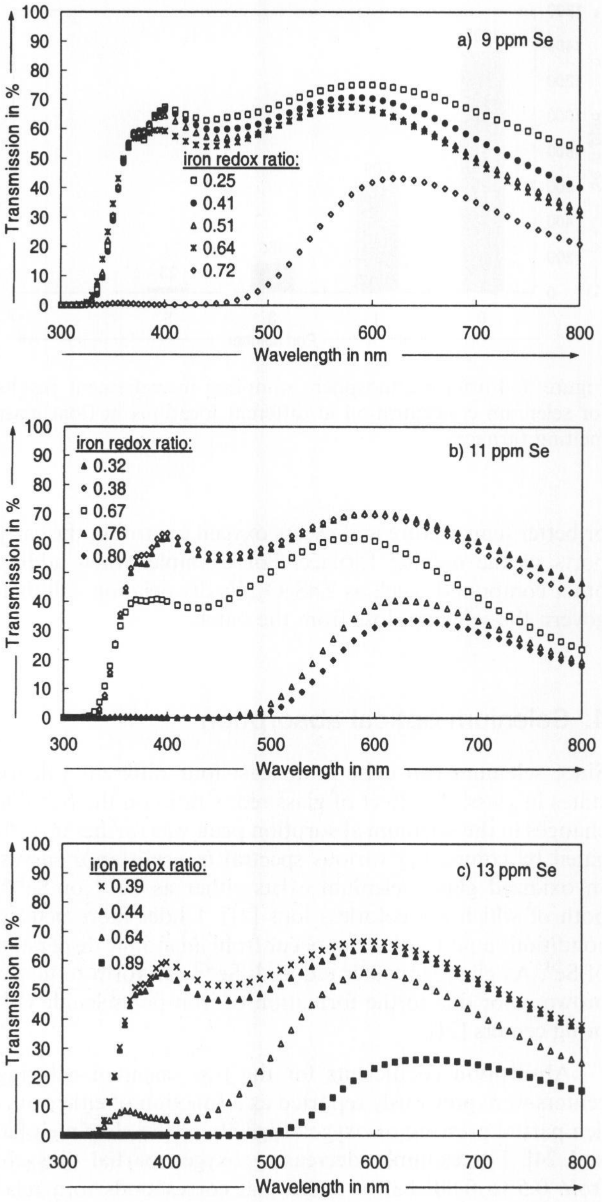


Figure 3. Furnace atmosphere sampling measurement results for selenium concentration at different locations in float glass melting furnace.

4. Selenium optical absorption

Since selenium can exist in at least four different valence states in glass, the effect of glass redox ratio on the possible changes in the selenium absorption peak was further investigated by comparing various spectral transmittance curves. In oxidized glass, selenium exists either as Se^{+4} or Se^{+6}, both of which are colorless ions [21]. Under more neutral conditions a pink color can occur from an atomic dispersion of Se^0. As glass is further reduced, Se^{-2} can form to give a brown color due to the formation of iron-polyselenide coloring centers [21].

Absorption coefficients for the two selenium coloring centers were previously reported as a function of either oxygen partial pressure or oxygen fugacity of the glass melt [20 and 24]. For example, decreasing oxygen partial pressure from 0.5 to 0.001 bar, a range that corresponds to a relatively oxidized glass, significantly decreased the absorption coefficient of Se^0 at about 480 nm from figure 4 in [24] from 0.0070 to 0.0005 cm^{-1}, while the absorption coefficient of iron-polyselenide varied only slightly from 0.0025 to 0.0020 cm^{-1} over this same range of partial pressure of oxygen for glasses containing 200 ppm iron [24]. This behavior is likely due to the disappearance of the Se^0 coloring centers at lower oxygen partial pressure and the subsequent decrease in absorption. In addition, only a slight change was observed in the location of the absorption peak from 440 to 420 nm for the iron-polyselenide coloring center as the oxygen fugacity was lowered from 2.5×10^{-8} to 3.2×10^{-12} bar, corresponding to a highly reduced chemistry in a borosilicate glass [20]. The shift in the peak wavelength was attributed to different distributions of polyselenide ions in the glass. An increase in the intensity of the Se absorption band was also reported at lower oxygen fugacity [20]. In addition, since most glass melts contain tramp amounts of sulfur, upon further increasing the iron redox ratio, amber coloration begins to occur at about 0.6 redox ratios, as calculated in this work, which is equivalent to oxygen partial pressures of about 10^{-7} to 10^{-8} bar, or higher due to the formation of iron-polysulfide coloring centers. At these



Figures 4a to c. Comparison of spectral transmittance curves for various glasses with same total iron and selenium concentrations, but different iron redox ratios; a) 9 ppm Se, b) 11 ppm Se, and c) 13 ppm Se.

higher redox levels, it is proposed that new coloring centers can also occur, which contain iron coordinated with both polysulfides and polyselenides as will be discussed below.

Figures 4a to c give the spectral transmittance curves at 4.06 mm thickness for different glasses that contain about 9, 11 and 13 ppm of selenium and about 0.36 % Fe_2O_3 in a redox ratio range of 0.25 to 0.89. As expected the higher redox glasses show a lower transmittance over the solar spectral range. The broad selenium peak at 480 nm appears to have slightly shifted to shorter wavelengths giving a minimum at about 440 nm. At still higher redox levels, there were also indications of the formation of iron-polysulfide coloring centers that have an absorption peak at about 415 nm. However, since the total sulfur content in the high redox glass melts was typically about 300 to 500 ppm (calculated as SO_3), which is much greater than the selenide content, it can be expected that this region of the transmittance spectrum is dominated by the absorption from the iron-polysulfide coloring centers at higher redox levels. Although an increase in the glass absorption occurred with the increase in glass redox ratio, it is not possible in this case to separate out the higher absorption due to iron-polyselenide coloring centers from that due to iron-polysulfide coloring centers, without performing spectral modeling. Melts would need to be made that do not contain any tramp impurities of sulfur to better determine this relationship, which would require the use of highly pure materials.

At higher redox ratios there is a noticeable shift in the absorption peak to shorter wavelengths giving a minimum at about 440 nm. In general, our results agree with the previously reported observations for the slight shift of the selenium absorption peak due to a lower oxygen fugacity [20]. As suggested above, the shift in absorption is likely due to the formation of new coloring centers that contain iron coordinated with both polysulfide and polyselenide ions. The generation of mixed ligand coloring complexes has been previously reported for water solutions [29], but no other investigations to our knowledge have identified the occurrence of such mixed ligand coloring centers in float glass.

There also appears to be a slight downward shift in the wavelength of the peak transmittance from about 600 nm at low redox levels to about 575 nm at 0.65 redox, then another upward shift to about 650 nm for glass redox levels greater than 0.70. As will be further discussed later, this behavior is caused by the slight changes in the absorption of both selenium and iron-polysulfides as the glass redox ratio increases.

Figure 5 gives the spectral transmittance curves for several laboratory glasses that do not contain any selenium additions. This graph allows the determination of the contribution to the absorption due to the iron-polysulfide coloring centers. The glasses contained about 0.37 to 0.42 % Fe_2O_3 and the glass redox ranged from about 0.26 to 0.71. The shapes of the transmittance curves at 450 to 480 nm are all similar for redox levels between 0.26 and 0.52. However, due to the formation of iron-polysulfide coloring centers the transmittance is significantly decreased for a redox of 0.71. The loss in transmittance at higher wavelengths is due to increasing concentration of FeO at higher redox ratios.

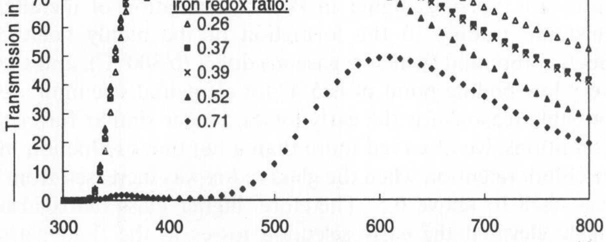


Figure 5. Comparison of spectral transmittance curves for five glasses with same total iron concentration and no selenium addition, but different iron redox ratios.

The optical density (OD) at 450 nm is plotted as a function of the selenium content in figure 6 for two different glass redox ratios of about 0.38 and 0.72. In both cases, the optical density increases as the selenium content increases, which means that the iron-polyselenide coloring centers are significantly contributing to the absorption. As the selenium content increases, there is a slightly greater rate of increase in the optical density at 450 nm for the glasses with a higher redox of 0.72 than for the lower redox of 0.38. As indicated in figure 6, the increase in OD is significantly affected by the concentration of Se. This is to be expected at the 0.38 redox ratio, where there is not a significant contribution from iron-polysulfide color center. On the other hand, the anticipated dominance of the iron-polysulfide center at the 0.72 redox ratio still does not totally mask the iron-polyselenide absorption. In fact, at this high redox level, the contribution due to selenium appears to be increasing at a faster rate as its concentration increases.

To further illustrate this behavior, in figure 7 the transmittance at 450 nm is plotted as a function of glass redox for glasses containing about 0.36 % Fe_2O_3 at various selenium levels ranging between 0 to 13 ppm selenium. For glasses that contain no selenium, the transmittance at 450 nm is essentially flat as glass redox ratio increases from 0.20 to 0.60. However, there appears to be a slight decrease in the transmittance at 450 nm for the selenium containing glasses as glass redox increases over the same range. There is a very rapid decrease in transmittance at 450 nm between glass redox ratios of 0.6 to 0.7. This is mainly due to the onset of absorption from iron-polysulfide coloring centers as indicated by the arrow in figure 7, which dominate the spectral absorption at shorter wavelengths for redox levels greater than about 0.60. A slightly greater decrease in transmittance as the glass redox ratio increases is observed for glasses with higher selenium contents. Therefore, it is concluded that the absorption due to iron-polyselenide coloring centers slightly increases as the glass redox ratio increases in addition to the much larger absorption due to iron-polysulfides that occurs at high redox levels. In summary, the behavior of selenium coloring centers in float glass is in agreement with the results of previous studies in other glass compositions [20 to 24]. In addition, one possible explanation for the observed changes in spectral transmittance is due to the formation of new coloring centers, which contain iron coordinated with both polysulfides and polyselenides.

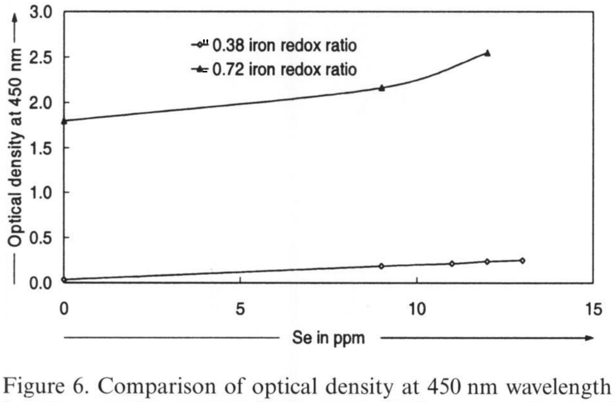


Figure 6. Comparison of optical density at 450 nm wavelength for glasses with same total iron concentration at two levels of iron redox ratio, but different selenium contents.

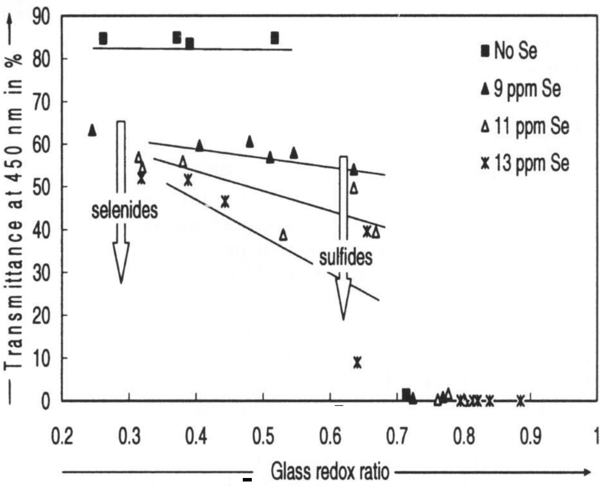


Figure 7. Comparison of transmittance at 450 nm wavelength for glasses with same total iron concentration, but different iron redox ratios and selenium contents.

5. Conclusion

Based on the laboratory glass melting experiments that were performed to investigate the effect of glass redox on selenium retention, it was shown that selenium retention initially decreases as glass redox ratio increases, but then surprisingly levels off and reaches a minimum, before once again increasing for very high redox ratios. In this regard, it is concluded that selenium retention in float glass behaves similar to sulfur retention as a function of glass redox ratio. Measurements of the selenium contents in the atmosphere at different locations of a commercial float glass furnace indicated that the majority of the selenium losses occur very early in the melting process. These results suggest new areas for future research to further improving selenium retention during the melting of glasses with high redox ratios. Finally, by comparing the spectral transmittance curves it was shown that the glass redox ratio has a measurable influence on the absorption of the selenium containing coloring centers in float glass, that is from the absorption of samples with various redox ratios and selenium concentrations we have been able to show evidence for a nonlinear increase in the optical density due to increasing amounts of selenium in glass, particularly for highly reduced glasses.

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