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REVIEW ARTICLE

Concise Reviews and Hypotheses in Food Science

Mechanisms of the initial stage of non-enzymatic oxidation of wine: A mini review

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Abstract: Non-enzymatic oxidation is a primary factor affecting wine quality during bottling or aging. Although red and white wines exhibit distinct responses to oxidation over time, the fundamental mechanisms driving this transformation remain remarkably uniform. Non-enzymatic oxidation of wine commences with the intricate interplay between polyphenols and oxygen, orchestrating a delicate redox dance with iron and copper. Notably, copper emerges as an accelerant in this process. To safeguard wine integrity, sulfur dioxide (SO_2) is routinely introduced to counteract the pernicious effects of oxidation by neutralizing hydrogen peroxide and quinone. In this comprehensive review, the initial stages of non-enzymatic wine oxidation are examined. The pivotal roles played by polyphenols, oxygen, iron, copper, and SO_2 in this complex oxidative process are systematically explored. Additionally, the effect of quinone formation on wine characteristics and the intricate dynamics governing oxygen availability are elucidated. The potential synergistic or additive effects of iron and copper are probed, and the precise balance between SO_2 and oxygen is scrutinized. This review summarizes the mechanisms involved in the initial stages of non-enzymatic oxidation of wine and anticipates the potential for further research.

KEYWORDS
non-enzymatic oxidation, wine oxidation, wine quality

1 | INTRODUCTION

Wine oxidation affects the sensory characteristics of wine, such as color and aroma, which in turn affects consumer choice or acceptance of wine (Lockshin & Corsi, 2012; Rinaldi et al., 2021). Generally, oxidation in red wines stabilizes the color, reduces the astringency, and improves the aroma profile; but for white wines, winemakers try to avoid the negative characteristics of oxidation, such as browning (Andrea-Silva et al., 2014; Bueno et al., 2018; Oliveira et al., 2011; Picariello et al., 2020; Ugliano, 2013). The crucial importance of producing wines that are free of deficiencies and have sensory characteristics that appeal to consumers is largely related to wine oxidation. More and more research is focusing on understanding and applying wine oxidation to produce consumer-appealing wine products.

Wine oxidation can be classified into enzymatic and non-enzymatic oxidation (Li et al., 2008). In general, enzymatic oxidation may occur in the early stages of winemaking and it is facilitated primarily by oxidoreductases (Zhao et al., 2023). As the fermentation process advances and ethanol is produced, the activity of oxidoreductases is gradually suppressed. Subsequently, non-enzymatic oxidation, primarily manifesting during the aging and post-bottling phases, emerges as the predominant factor influencing the

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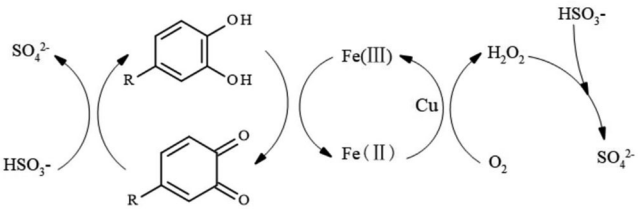

SCHEME 1 Initial reaction mechanism of non-enzymatic oxidation of wine (Danilewicz, 2018).

sensory characteristics of wine (Kilmartin, 2022), which is an essential factor influencing consumers' choice of wine (Vita et al., 2019).

Non-enzymatic oxidation occurs when polyphenol, oxygen, and transition metals are present at the same time (Danilewicz, 2003). Since they are prevalent in wine, this oxidation reaction is universally present in wine. SO_2 has been successfully used as an antioxidant to protect wines from oxidation. The above processes constitute the initial stages of non-enzymatic oxidation of wine as described in Scheme 1 (Danilewicz, 2018; Waterhouse & Laurie, 2006).

The presence of oxygen significantly influences wine by generating H_2O_2 and quinone (formed by oxidizing polyphenols through the presence of iron), which can further oxidize other substrates or form color compounds, thereby impacting the color and aroma profiles of the wine (Zhao et al., 2023). The addition of SO_2 serves as a preventive measure against oxygen-induced wine oxidation by reacting with H_2O_2 and quinone. Quantitatively, 1 mol of SO_2 can react with 1 mol of H_2O_2 and 1 mol of quinone. Theoretically, a 2:1 ratio of SO_2 to O_2 is suggested, a finding substantiated in model wines (Danilewicz et al., 2008). However, in real wines, this ratio occasionally deviates from the anticipated 2:1, likely attributed to the inherent complexity of actual wine compositions (Danilewicz, 2016). SO_2 emerges as a potential predictor of wine shelf life, underscoring its significance in preserving wine quality. The general change in the metal species during the oxidative and reductive aging of wine is not well understood and variation in results was observed in model wine and real wine. For example, Berg and Akiyoshi (1956) reported additive effects of iron and copper in white wine, while a more recent investigation by Danilewicz (2007) identified synergistic effects in model wine. Previous studies have confirmed that both iron and copper can expedite the production of xanthylium derivatives but their catalytic effects in autoxidation differ (Clark & Scollary, 2002; Oszmianski et al., 1996). Clark and Scollary (2002) discovered that the browning rate was not strictly proportional to the concentration of Cu (II) in the model solution. It only exerted an impact when its concentration reached a certain level. In contrast to copper, iron exhibits higher efficacy in catalyzing the formation of xanthylium cations from (+)-catechin and tartaric acid (George et al., 2006).

This review aims to comprehensively examine the impacts of four principal factors, namely polyphenols, oxygen, metals (iron and copper), and SO_2, on the process of non-enzymatic oxidation in wine. Underlying this exploration is a focus on elucidating the intricate mechanisms involved in the interaction of these factors and their collective influence on the oxidation pathways. By delving into the underlying mechanisms, this review seeks to contribute valuable insights that deepen the understanding of the complex interplay among polyphenols, oxygen, metals, and SO_2 in shaping the non-enzymatic oxidative processes within the context of wine.

2 | POLYPHENOLS

Polyphenols are secondary metabolites in plants, consisting of one or more aromatic ring compounds with one or more hydroxyl groups, which are highly relevant to the characteristics and health-improving properties of wine (Gutiérrez-Escobar et al., 2021). When polyphenol concentrations were similar in different wines, comparable antioxidant activity could be achieved (Garaguso & Nardini, 2015). Therefore, polyphenols are also the major substrate for the determination of the antioxidant capacity of wines (Danilewicz, 2015).

Many review articles have categorized polyphenols in wine (Figure 1) (Cataldo et al., 2023; Merkyte et al., 2020). The antioxidant capacity of red wines (stored at 10°C in the dark) was analyzed by Ferric reducing antioxidant power assay (FRAP) and DPPH• assay, and the results showed good correlation between antioxidant capacity and total flavanols content (527 mg L^{-1} catechin) of red wines, with $R^2 = 0.842$ and 0.786, respectively; moreover, sensory testing and the results from potentiometric titration assay were highly correlated ($R^2 = 0.8869$), therefore, it is suggested that wine aging capacity can be predicted by polyphenols (Waterhouse & Miao, 2021). The antioxidant capacity of red wines is significantly higher than white wines because red wines have higher amounts of polyphenols. In general, red wines exhibit an average polyphenol concentration of 2045 mg CE/L (catechin equivalents, with a range of 752 to 3949 mg CE/L), whereas white wines display a lower concentration of 112 mg CE/L (with a range of 53–177 mg CE/L) (Banc et al., 2020; Danilewicz, 2007).

2.1 | Mechanism of polyphenol oxidation

In the wine, polyphenols react with oxygen with the assistance of iron, resulting in the production of quinone and H_2O_2. The presence of SO_2 leads to the consumption of

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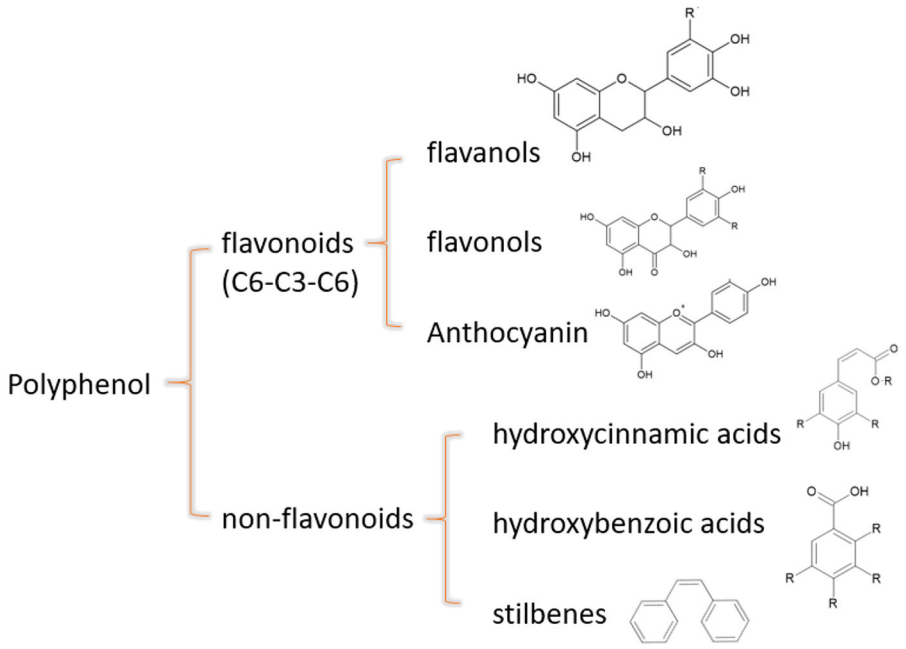

FIGURE 1 Classification and structure of polyphenols in wine.

both quinone and H_2O_2, effectively inhibiting subsequent reaction pathways (Li et al., 2008). The reduction potential of polyphenol/quinone is similar to O_2/H_2O_2, and the net difference in potential (ΔE) for this reaction is close to zero. Therefore, the reaction between these two couples is quite slow and requires the help of a nucleophile (which can react with quinone) (Danilewicz, 2003). As the primary substrates for oxidation, polyphenols in the catechol and pyrogallol moieties are most vulnerable thought wine oxidation, including caffeic acid, (+)-catechin, (-)-epicatechin, quercetin, and gallic acid (Li et al., 2008). Unlike the catechol moieties, the polyphenol containing pyrogallol moieties can be oxidized at significant rates, and both oxidations can be accelerated by SO_2 by reacting with oxidized polyphenol quinone (Danilewicz, 2011a). Although considered as an anti-oxidant, in the presence of SO_2, catechol does not react with the free radicals produced during the oxidation since the probability is very small (~0.5%) (Danilewicz, 2011a). Moreover, not all polyphenols exhibit antioxidant effects. In a study, Marquez et al (2019) observed that caffeic acid, protocatechuic acid, and p-coumaric acid exhibited the capability to stimulate the generation of hydroxyl radicals (HO·), thereby demonstrating a pro-oxidant influence.

Polyphenols could also be categorized into reversible and irreversible polyphenols, which could be identified by cyclic voltammetry (Makhotkina & Kilmartin, 2009). This is the consequence of the reduction of quinone to polyphenol in the presence of certain nucleophiles, such as SO_2. The quinone generated from (+)-catechin and (-)-epicatechin can be reduced back to the polyphenol form by SO_2 with reduction rates of 96% and 79%, respectively (Danilewicz & Wallbridge, 2010). This could be a reason why the antioxidant capacity of wine determined by Folin-Ciocalteu and FRAP was affected by SO_2 (Danilewicz, 2015). As shown in Table 1, SO_2 affected the results of antioxidant determination in wine.

2.2 | Influence of quinone on wine instability

Quinone arises as a product of polyphenol oxidation, displaying instability upon its formation (e.g., reactions with varietal aroma components) (Nikolantonaki et al., 2014). Consequently, it promptly engages with nucleophilic substances, such as thiols and other polyphenols, thereby exerting a pronounced influence on the composition of wine. The presence of antioxidants (e.g., SO_2, glutathione, and ascorbic acid) may sacrificially react with quinone to protect the wine from the change in color and aroma loss while converting the quinone back to polyphenol form (Figure 2) (Geng et al., 2023; Nikolantonaki & Waterhouse, 2012).

The production of quinone leads to the browning of wines, which could be damaging for white wines as white wines are particularly sensitive to oxidation due to their low phenolic compound content (Kanavouras et al., 2020). Flavonoids, a class of phenolic compounds characterized by a three-ring backbone, are acknowledged for their antioxidant properties and their capacity to inhibit browning. However, upon encountering quinones, the

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TABLE 1 Effect of SO_2 on different antioxidant assays (Danilewicz, 2015).

	Sauvignon blanc (with 33.6 mg/L free SO_2)	Sauvignon blanc (without free SO_2)
FRAP (mg/L CafE)	225.2 ± 0.3	85.0 ± 1.0
DPPH• (mg/L CafE)	96.4 ± 1.5	73.2 ± 0.6
F-C (mg/L GAE)	303 ± 9	225 ± 14

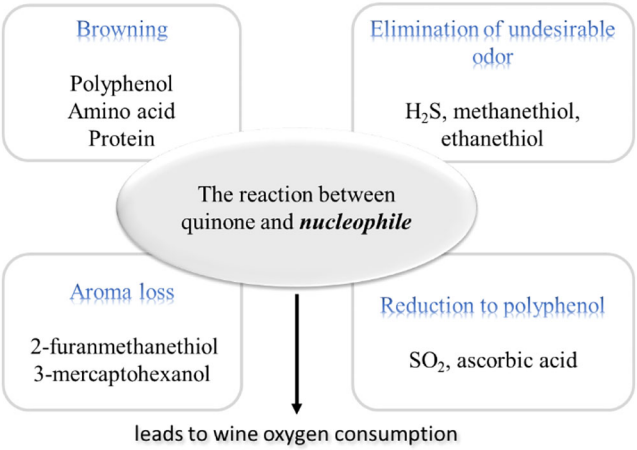

FIGURE 2 Influence of the reaction between quinone with nucleophile on the wine quality.

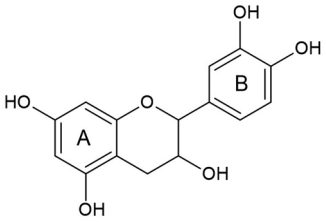

FIGURE 3 A-ring and B-ring of polyphenol in wine.

constituents of flavonoids, specifically the A-ring and B-ring (Figure 3), exhibit distinct reactivity, with the B-ring displaying a heightened reaction rate in comparison to the A-ring, which is attributable to the faster rate of electron transfer reactions than nucleophilic reactions (Ma & Waterhouse, 2018). This discrepant reactivity profile culminates in the generation of precursors associated with the browning process. Furthermore, Ma and Waterhouse (2018) mentioned that the reaction rate of SO_2 is higher than the reaction rate of both A-ring and -B-ring, thus inhibiting the browning. A recent study performed by Su et al. (2022) indicated that chlorogenic acid promotes the oxidation of (+)-catechin to form (+)-catechin quinone, which forms a brown pigment with (+)-catechin in long-term storage of non-wine model. Whether this reaction can occur in the case of wine is not known yet. It is assumed that it depends on the reduction rate of quinone by SO_2.

Volatile thiol is the dominant aroma because of the low odor detection threshold (0.8-60 ng/L) (Coetzee et al., 2012). Loss of thiol occurs through either Cu-mediated oxidation or a reaction with quinone (Kreitman et al., 2016; Ugliano, 2013). In model wines it was shown that 4 mM antioxidants, including ascorbic acid, SO_2, and glutathione react very quickly ($k = -0.3808$ to -0.3343 s^{-1}) with quinone and should protect thiol (i.e., 3-sulfanylhexanol) (Nikolantonaki et al., 2014). Furthermore, ascorbic acid-protected thiol was confirmed in rosé wine (Zhang et al., 2023). However, due to the complexity of the wine matrix and the lack of standards, it is not possible to identify all of the reactants and products of reactions with quinones in wine. Stable isotopic labeling combined with high-resolution mass spectrometry on Sauvignon Blanc and Cabernet Sauvignon showed that sulfur-containing compounds and flavonoids were the main reactants. In white wines, the quinone reaction products contain sulfur, while in red wines, most of them are flavonoid-quinone adducts only few of them contain sulfur (Ji et al., 2022).

Furthermore, Danilewicz et al (2019) mentioned that produced quinone during the oxidation of polyphenols needs to be removed with the help of added antioxidants (SO_2) in order to avoid further oxidation phenomena in wine. Therefore, the author proposed "polyphenol + SO_2" worked as an antioxidant. Moreover, future studies should delve into a more comprehensive exploration of quinone-associated reactions and their respective kinetics. This is particularly critical within the intricate wine matrix, where these reactions can attain heightened complexity. Such endeavors stand to significantly enhance comprehension of wine oxidation mechanisms and subsequently rationalize the use of antioxidants.

3 | OXYGEN (O_2)

Oxygen plays a critical role in wine production. After fermentation, the bottling process results in maximum wine oxygenation (2.99−4.12 mg/L). The maximum wine oxygenation process involves saturating the wine with oxygen to a controlled level, and it is one of the factors that can affect wine color, composition, and antioxidant activity (Kulhankova et al., 2023; Petrozziello et al., 2018). The mechanisms of oxidation are similar in the case of both red and white wines (Danilewicz, 2003; Waterhouse & Laurie, 2006). More specifically, wine oxidation could have a positive or negative impact on the aroma, color, and taste of the wine, depending on the wine varieties (Tarko et al., 2020).

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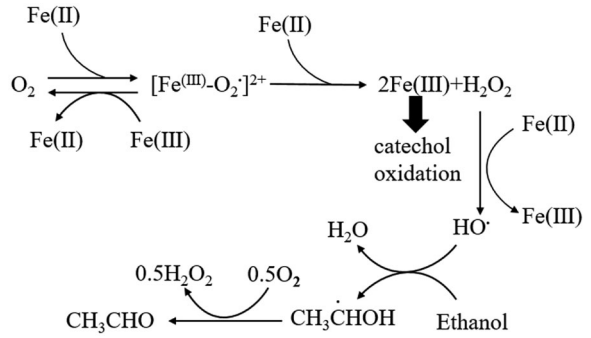

SCHEME 2 Proposed mechanism of oxygen participation in wine oxidation (Danilewicz, 2013).

An aging experiment on Nebbiolo wine showed that moderate oxidation (7–14 mg/L oxygen total intake) contributes to color stabilization by increasing the content of anthocyanins, while excess oxygen (21–28 mg/L oxygen total intake) affects the quality of the wine, increasing methionine content by inducing chemical reaction (Petrozziello et al., 2018). As shown in Scheme 2, oxygen is involved in the oxidation of wine and can form acetaldehyde by oxidizing ethanol. A study of Cabernet Sauvignon wines during post-fermentation stage found that the acetaldehyde content of high-level oxygenation wines (3-3.9 mg O_2/L per time) increased from 4 to 11 mg/L. The acetaldehyde content of wines with low levels of oxidation did not change much (from 4 to 5 mg/L) (Dai, Sun, et al., 2022). The influence of varying acetaldehyde levels (4.86 ± 0.60, 8.67 ± 1.14, and 29.66 ± 1.74 mg/L) on the aging dynamics of Cabernet Sauvignon wines was studied over a 12-month period. Results indicated a direct relationship between acetaldehyde concentration and the rate of change in both polymeric pigment and polymeric tannin content. Specifically, higher acetaldehyde levels corresponded to increased rates of change in polymeric pigment and tannin, with changes of approximately 75%, 125%, and 175% for pigment, and 25%, 37.5%, and 50% for tannin, respectively (Han et al., 2019). Therefore, it is hypothesized that oxygen management may shorten the aging time by promoting the production of acetaldehyde. In comparison to red wines, white wines demonstrate a higher susceptibility to oxidation, leading to a distinct reduction in fruit aromas and the manifestation of noticeable pink and brown colorations (Castellanos et al., 2021; Gabrielli et al., 2021).

3.1 | Reactive oxygen species (ROS) by O_2

Molecular oxygen lacks the inherent capacity for direct interaction with polyphenols or SO_2 due to its electron configuration (Danilewicz, 2003). The exchange of single electrons between O_2 and iron in wine results in the formation of ROS, which participates in the oxidation reaction of wine (Oliveira et al., 2011). O_2 accepts the first electron and forms a superoxide radical anion, which should exist in the acidic environment of wine in the form of protonated hydroperoxyl radical (HOO·), however the presence of this radical was not supported, since no spin adduct with 5-*tert*-butoxycarbonyl 5-methyl-1-pyrroline *N*-oxid (BMPO), BMPO/HOO·, was found by Electron Paramagnetic Resonance (EPR) (Elias et al., 2009; Kreitman et al., 2016). A proposition has been advanced, suggesting that O_2 undergoes direct reduction in a two-electron process, resulting in the formation of H_2O_2 (Kreitman et al., 2016). This H_2O_2 then generates a highly reactive oxidant known as the hydroxyl radical (HO·). This hydroxyl radical exhibits the capability to oxidize the initial organic compound it encounters at diffusion-controlled rates, ultimately yielding H_2O, which serves as the final oxygen reduction product (Danilewicz, 2003). The substance in the wine reacting with HO· would be ethanol, extracted an α-hydrogen atom to produce the hydroxyethyl radical, which turn in forms of acetaldehyde (Elias & Waterhouse, 2010). This is a sign of oxidation and could combine with wine constituents to form components that affect aroma and color (Echave et al., 2021). However, in the case of the Fenton reaction, there is still controversy over the type of radical produced by H_2O_2 to further oxidize substrate (e.g., tartaric acid or ethanol). Coleman et al (2020) and Kremer (2008), who preferred FeO^{2+}, rather than HO, to participate in the next step of the oxidation reaction as a free radical produced by H_2O_2.

3.2 | Oxygen pathways involved in the initial stages of non-enzymatic oxidation of wine

The principal mechanism underlying oxidation involves the interaction of oxygen through its reaction with Fe(II), as shown in Scheme 2 (as described in Section 4 labeled 'Role of transition metals: Fe and Cu') (Danilewicz, 2011a; Danilewicz & Wallbridge, 2010). This is why O_2 absorption was observed in the model wine solution without the addition of SO_2 (Danilewicz et al., 2008). During the oxidation process, O_2 will eventually be reduced to water, leading to the production of H_2O_2 and HO·, two intermediate oxygen species that can oxidize the wine constituents (Elias & Waterhouse, 2010; Waterhouse & Laurie, 2006). In addition to this pathway, it has been found that the addition of H_2O_2 to model wines can cause additional O_2 consumption, suggesting that the Fenton reaction can also uptake oxygen (Danilewicz, 2013). In the Fenton reaction, H_2O_2, the reaction product of oxygen with Fe^{2+} (the first pathway for oxygen to participate in non-enzymatic oxidation),

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oxidizes ethanol to 1-hydroxyethyl, which is proved by EPR, in the presence of Fe^{2+}. Subsequently, oxygen can further oxidize 1-hydroxyethyl to form acetaldehyde, which is also the second site where oxygen is involved in non-enzymatic oxidation, as shown in Scheme 2 (Elias & Waterhouse, 2010).

3.3 | Oxygen content in relation to wine oxidation

Winemaking practices result in different concentrations of oxygen in the wine. Increasing the redox potential by introducing air during Cabernet Sauvignon wine fermentation at different scales (100-10,000 L) seems to alleviate the negative sensory characteristics of wines caused by too low reduction potentials, such as hydrogen sulfide (Nelson et al., 2023). In a fermentation study of Shiraz wines, it was found that within the aeration scope studied, not only reductive odors were reduced, but red fruit characteristics were also increased (Day et al., 2021). When the wine is exposed to air, the oxygen concentration is about ~8 mg/L at room temperature. Winemaking operation increased the oxygen content by a range of 0.41 to 2.07 mg/L, with bottling causing the largest increment in dissolved oxygen content (Hajjaj, 2016). Further, another study showed that bag-in-box filling contributed the most to oxygen enrichment (2.47 mg/L for red wines; 2.22 mg/L for white wines) (Catarino et al., 2014). In a study of Riesling wines, a strong correlation was found between headspace volume and color change ($p < 0.0001$). The absorbance values at 420 nm increased with the headspace volumes of the bottle: 0.082 (0 mL), 0.091 (10 mL), 0.102 (20 mL), and 0.115 (30 mL) after 6 months of storage (n = 3) (Morozova et al., 2015). Results of a 400-day period of monitoring SO_2 in Chardonnay, Cabernet Sauvignon and Merlot wines from Bag in box (varied in supplier, film ply construction, EVOH content, and style/construction of the wine tap, details not given) showed that within the first 30 days of storage, the loss of SO_2 was likely due to the headspace volume, which was approximately 3.95 ± 1.3 mg O_2/L in all combinations of wines and packages. After 30 days, the rate of oxygen exposure becomes constant and is related to the "oxygen transmission rate (OTR)" through the packages (Sacks et al., 2020).

On the other hand, Godden et al. (2001) suggested that oxygen content at bottling (0.6−3.1 mg/L) could not be used as a predictor of wine browning. In the same study, four types of wine closures, including the screw-cap, natural cork, technical cork (natural cork with a synthetic component), and synthetic cork were also comprehensively explored. The study showed that screw-cap resulted in the lowest SO_2 loss, followed by technical cork and natural cork, and the highest SO_2 loss in synthetic cork. Loss of SO_2 was highly correlated ($p < 0.001$) with browning, so that wines under the screw-cap had the most sensory protection as far as browning was concerned (Godden et al., 2001). However, the permeability of the closure to air affects the absorption and consumption of oxygen, which in a cascading effect, transforms the wine's hue and sensory traits (Castellanos et al., 2021). A blend of white wine sealed with synthetic corks have the lowest free SO_2 content (18 mg/L) and the highest A_{420nm} value (0.1500) compared to 1+1 cork (technical stoppers with a disc at each end) (25 mg/L free SO_2 and $A_{420nm} = 0.1300$) and micro-agglomerated cork (25 mg/L free SO_2 and $A_{420nm} = 0.1200$) after 48 months post-bottling (Oliveira et al., 2020). What is more, oxygen transfer between the cork and the glass neck of the bottle is also an important factor that should not be overlooked, as it affects the chemical composition of the wine (Karbowiak et al., 2019). It has been proposed that the rate of oxygen entry is slower than the rate of oxygen consumption in bottling wines, and therefore OTR controls the rate of wine oxidation (Danilewicz & Standing, 2018; Sacks et al., 2020). In a study on closure OTR after 10 years of bottle storage on wines, it was shown that OTR correlates with free SO_2 decrease ($R^2 > 0.700$, $p < 0.0001$), dissolved oxygen ($p < 0.001$), color change (absorbance at 420 nm) ($p < 0.001$), 3-sulfanylhexanol ($p < 0.001$) and sotolon ($p < 0.001$). When stored in a closure with higher OTR, the parameters of wine mentioned above become higher. Therefore, these parameters can be used to evaluate the oxidation of wines (Pons et al., 2021). The selected Sauvignon Blanc wines were not oxidized when OTR did not exceed 0.3 mg/year.

Some studies have been done to make wines more stable by controlling the amount of oxygen, these strategies include micro, macro-oxygenation, and aeration techniques. Micro-oxygenation was developed in 1991, France. It can stabilize red wine color through the formation of stable pigments called pyranoanthocyanins, by continuously (from a few minutes up to 7 months) introducing low concentrations of O_2 (2 mg/L/month to 90 mg/L/month) (Gómez-Plaza & Cano-López, 2011; Quaglieri et al., 2017). Macro-oxygenation typically used during fermentation to reduce the amount of reducing odorants (Bekker et al., 2021). Aeration of Shiraz wines during fermentation, with total oxygen exposure ranging from 1.5 to 50 mg/(L h), was effective in improving sensory characteristics. For example, total anthocyanin and phenolic decreased ($p < 0.05$) with higher oxygen exposure in all trials. Acetate acid and ethyl acetate are associated with oxidation. The concentration of acetate acid was only found to reach 0.84 g/L in the long, higher intensity aerations (total oxygen exposure 20.4 and 49.3 mg L^{-1} h^{-1}). In the same trial, ethyl

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acetate reached 81 and 102 mg/L compared to Control trial (48 mg/L for no oxygen exposure treatment) (Day et al., 2021).

4 | ROLE OF TRANSITION METALS: FE AND CU

Transition metals such as iron and copper in wine primarily come from soil, fertilization, the winemaking process, and environmental contamination. They are capable of initiating the oxidation of wine by the activation of oxygen in the range of iron in wine, affecting the quality of the wine (Danilewicz, 2003; Tariba, 2011). Globally, the levels of iron and copper in wine ranged from 0.061 to 50 mg/L and ND (not detected) ~6.82 mg/L, respectively. The iron comes mainly from the soil where the grapes are grown and is absorbed through the roots and eventually into the wine. Copper comes mainly from pesticides (Tariba, 2011).

4.1 | Mechanism of iron-catalyzed oxidation

In acidic aqueous solutions, the two species of iron (ferric and ferrous ions) exist as hexahydrate complexes. The protonation of this hydrate varies according to pH, which leads to a difference in the reduction potential of the Fe couple. Meanwhile, substances such as polyphenols, organic acids, etc. which are stronger ligands than water in wine, displace water in the iron-complexes, further affecting the reduction potential of the iron couple. Therefore, the reduction potential of iron is not only affected by pH, but is also ligand dependent (Danilewicz, 2014). In general, the reduction potential of Fe is considered to be 360 mV in wine at pH 3.5. Furthermore, the ground state electronic configuration of iron allows the reaction between polyphenol and oxygen, which is possible by redox cycling of iron (interconversion of Fe(III) and Fe(II)) (Danilewicz, 2003). In the model wine solution, no significant oxidation reaction occurred in the absence of iron and copper. The addition of 5 mg/L Fe and 0.15 mg/L Cu in model wine resulted in a reduction of 20 mg/L SO_2 within 250 h compared to model wines without metal addition, which indicated that iron and copper initiate the oxidation in model wine (Danilewicz, 2007). The addition of polyphenols to the model wine resulted in a more rapid reduction of SO_2, with a reduction of 90 mg/L free SO_2 at 125 h. No significant oxidation of polyphenol was observed with Cu alone (Danilewicz et al., 2008).

It has been demonstrated that Fe(II) is able to react with O_2 to produce H_2O_2 and Fe(III). This is a thermodynamically favored reaction. In model wine conditions, at pH = 3.6, the potential of Fe is 345 mV and that of O_2/H_2O_2 is 570 mV. The Gibbs free energy for this reaction is negative ($\Delta G = -42.4$ KJ/mol), indicating that Fe(II) is easily oxidized by O_2. Meanwhile, Fe(III) formed by oxidation of Fe(II) will react with polyphenols to produce quinone and Fe(II). Unlike the reaction between Fe(II) and O_2, the reaction of Fe with polyphenols is thermodynamically unfavored. Using Fe(III) with 4-methylcatechol (4-MeC) as an example, ΔG of the reaction is +43.8 KJ/mol. However, the instability of the quinone and the presence of nucleophiles in the wine, facilitate the continuation of the reaction (Danilewicz, 2013). From this point of view, iron redox is influenced by the reduction potential, which is related to the ligand in the wine. It has been demonstrated that the acid composition (i.e., tartaric acid, malic acid, and citric acid) of wine affects the rate and output of the Fenton reaction (Nguyen & Waterhouse, 2022). This is because different acids have different abilities to lower the reduction potential of iron, which affects the potential difference of the redox reaction, and the rate of reaction is therefore different. Further studies of the Fenton reaction have shown that the complex of iron (II) and tartaric acid is an important factor driving oxygen activation (Coleman et al., 2023). In the reacting system, the changes in dissolved oxygen content show distinct initiation, propagation, and termination phases. In further research, Coleman et al. (2023) found that increasing the pH altered the initiation phase and shortened the propagation phase, with a critical pH of 3.5. This may provide a new explanation for the different reaction rates of red and white wines in the face of oxidation (Coleman et al., 2023). There is no study on the effect of different acids on the initial oxidation of wine.

Cu is proposed to facilitate the redox cycle of iron. It has been shown that copper has no effect on the reaction between iron and polyphenols (Nguyen & Waterhouse, 2021). For the oxidation of Fe(II), Cu can reduce the reaction time from 3 days to 6 h (Danilewicz, 2011a; Elias & Waterhouse, 2010). Nevertheless, the mechanism by which Cu catalyzes the reaction of Fe(II) with O_2 is still unclear.

4.2 | Ratio of ferric and ferrous ions

The wine redox potential was once proposed as an indicator of the redox state of wine. However, it was later shown that the redox values of wine were due to the oxidation of ethanol at the electrodes and therefore related only to oxygen exposure (Danilewicz, 2011b). Moreover, redox values for individual components in wine may not exist, since certain redox pairs are not found in wine (e.g., polyphenol and quinone), and the equilibrium state required for redox determination cannot be obtained (Danilewicz et al., 2019).

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TABLE 2 Amounts of two types of iron in different wines (mg/L) (Danilewicz, 2018).

Red Wine	Fe(II)	Fe(III)	White wine	Fe(II)	Fe(III)
Cabernet Sauvignon	3.41	0.09	Pinot Grigio-1	1.97	0.10
Chianti	2.78	0.08	Pinot Grigio-2	1.47	0.08
Côte du Rhône	2.19	0.09	Chardonnay	0.43	1.97
Bordeaux	1.73	0.28	Sauvignon blanc-1	0.41	1.83
Merlot	1.59	0.43	Sauvignon blanc-2	0.62	0.90
Shiraz	2.03	0.69	Soave	1.13	1.16
Tempranillo	2.27	0.38			

It has been proposed that the two states of iron, ferric and ferrous ions, were somehow related to the redox state of wine (Danilewicz, 2018).

Theoretically, 10 mg/L of Fe(II) reacts with 2.86 mg/L of O_2, but only 1.6 mg/L is consumed in the model wine, so not all of the Fe(II) oxidized by O_2 (Danilewicz, 2011a). Based on the previously discussed redox mechanism of iron involvement in wine oxidation, the Fe(II):Fe(III) concentration ratio should be determined by the relative reaction rates of Fe(II) with oxygen and that of Fe(III) with polyphenols (Danilewicz, 2016). The rates of the two reactions were determined separately and it was found that the rate of oxygen consumption was much lower than the rate of Fe(III) reduction (in the presence of SO_2), which means that the oxidation of Fe(II) is the rate-limiting reaction for the wine oxidation (Nguyen & Waterhouse, 2021). A plausible interpretation of this finding is that the oxidation of Fe(II) is inhibited in the presence of Fe(III), thereby preventing its progression (Danilewicz, 2013). However, if both reactions are simultaneously considered, the rates of iron oxidation and reduction are the same when the reaction reaches the equilibrium, that is, when the ratio of two iron species becomes constant (Danilewicz, 2021).

The ratio of iron varies from wine to wine, as shown in Table 2. In a model wine solution, the addition of polyphenols changed the Fe(III):Fe(II) ratio from 2.08:1 to 2.6:1. Oxygen exposure reduces Fe(II) content and thus affects the Fe ratio (Danilewicz, 2016). The Fe(II) concentration was 64.7% at an initial oxygen concentration of 0.29 mg/L. However, when exposed to air, which was saturated with oxygen (~8 mg/L), the Fe(II) concentration decreased to 26.5% after 47.5 h (Danilewicz, 2016). The closure of the bottle affects the rate of ingress of oxygen (Echave et al., 2021). That is why wines closed with screw caps have more Fe(II) content compared to those sealed with natural corks or bag-in-box (Danilewicz, 2016). Similarly, in further research Danilewicz (2018) observed high Fe(II) content in screwcap, technical, and plastic closures and relatively low in natural corks and wine boxes. pH had little effect on the ratio of iron. Therefore, it is suggested that further research should explore how the composition of the wine (polyphenol) affects the ratio of Fe, and also the type of closure should be considered.

4.3 | Are iron and copper additive or synergistic?

Danilewicz (2007) determined the consumption of free SO_2 as an indicator of wine oxidation. When the model wines had no iron or copper, no significant oxidation reactions occurred and the rate of oxidation was very close to 0, which demonstrates the importance of transition metals for the oxidation of polyphenol or SO_2. With the addition of 5 mg/L iron or 0.15 mg/L copper, respectively, the free SO_2 concentration was reduced by about 10 mg/L compared to the control group (no iron or copper trial) at 100 h. However, when iron and copper were added simultaneously, 80 mg/L of free SO_2 was consumed. The two metals were used together resulting in a greater rate of free SO_2 depletion. However, as discussed in the SO_2 section, the reduction in total SO_2 can be assessed for wine oxidation because partially bound SO_2 requires time to complete the progress of the oxidation (Carrascón et al., 2018).

Cu significantly promotes the catalytic effect of Fe, in other words, the synergistic effect between iron and copper was demonstrated by adding Cu and comparing the time required for half of the consumption of oxygen (Danilewicz & Wallbridge, 2010). In wine containing 1.2 mg/L Fe and 0.05 mg/L Cu, the addition of 0.1 mg/L Cu dramatically reduced the time to half oxygen consumption from 45 days to 8.5 days, and to 4.7 days with the addition of 0.3 mg/L Cu. This effect is consistent with 5 mg/L Fe and 0.3 mg/L Cu. One research states that iron improves browning more than copper and that the two are additive effects. The different conclusions here may have arisen because of differences in experimental design. Danilewicz and Wallbridge (2010) adopted model wines and real wines to study the effects of Fe and Cu on O_2 or SO_2 consumption, respectively, whereas Berg and Akiyoshi (1956) used real wines to explore the effects of Fe and Cu on wine browning. Reactions involved in wine browning are more complex and

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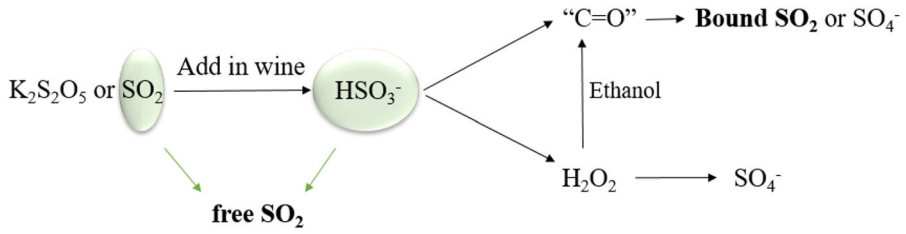

SCHEME 3 The role of SO_2 in initial non-enzymatic oxidation of wine.

there may be polymerization reactions of pigments that are not catalyzed by Fe and Cu leading to browning (Zhao et al., 2023). Hence more consistent studies should be continued to explore the effects of Fe and Cu on oxidation, and extra attention to the difference between model and real wine.

5 | ANTIOXIDANT: SO_2

Sulfur dioxide (SO_2) is widely used in wine production due to its excellent antimicrobial and antioxidant properties. Currently, the limits for total SO_2 in red and white wines are 150 and 200 mg/L, respectively, according to EU regulations (EU Commission, 2019). It has long been thought that SO_2 is able to react directly with oxygen, thus protecting wines from oxidative damage. However, recent decades of research have shown that SO_2 cannot react directly with oxygen due to obstruction of the electronic structure, making it important to explore the mechanism of the antioxidant effect of SO_2 (Danilewicz, 2007). Moreover, due to the potential allergenicity/toxicity of SO_2, the interest in finding antioxidants that can replace SO_2 has gained significant attention, which should also be based on the understanding of the antioxidant mechanism of SO_2 (Giacosa et al., 2019). Many attempts have been made to find alternatives to SO_2, such as ascorbic acid. But ascorbic acid produces a yellowing of the wine, in order to prevent this, the assistance of SO_2 is needed (Barril et al., 2016). With the concept of sustainability and health in mind, extracts from grapes or other plants have been used to evaluate as an alternative to SO_2. A recent study has shown that the use of unripe grape extract and chitosan in Sangiovese wine can provide comparable protection against SO_2. However, more experiments should be carried out in white wines, which are more susceptible to oxidation (Fia et al., 2023).

5.1 | The form and autoxidation of SO_2 in wine

There are two forms of SO_2 in wine: free and bound SO_2. Bisulfite (HSO_3^-), as a primary species of SO_2 in a wine-like solution, plays the role of nucleophile, antioxidant, and enzyme inhibitor (Scheme 3). Its conjugate acid exists in rapid equilibrium with molecular SO_2, which is an important antimicrobial. HSO_3^- and molecular SO_2 together are known as free SO_2. HSO_3^- can reversibly bind to components in wine to form bound SO_2, which includes aldehydes, ketones, and anthocyanins. According to the adduct dissociation equilibrium constant formed, the bound SO_2 can be further subdivided into strongly bound and weakly bound SO_2 (Waterhouse et al., 2016). Even strongly bound SO_2 like acetaldehyde-bisulfite adduct hydrolyzed to replenish free SO_2 when there is a shortage of free SO_2. Hence there should have been a stoichiometric relationship between the consumption of total SO_2 and oxygen (Sacks et al., 2020).

The autoxidation of SO_2 is a transition metal-catalyzed process that involves a free radical chain reaction (Zhou et al., 2018). For the time range studied, the rate of reaction between SO_2 and O_2 was almost 0 in the absence of transition metals. This could be explained by the fact that the reaction proceeds in violation of Pauli's exclusion principle, which is due to the difference in electronic structure, being in the triplet and singlet ground states, respectively (Danilewicz, 2003). The addition of 5 mg/L of iron and 0.15 mg/L of copper resulted in a decline in free SO_2, while ethanol was oxidized by peroxomonosulfate radical ($SO_5^{\cdot-}$) formed by the autoxidation of SO_2 (Danilewicz, 2007). SO_2 is not an antioxidant at this point, but rather, when used alone, it increases the oxidizing power of oxygen. In the presence of Fe(III) and in the absence of polyphenols, the oxidation of SO_2 was very slow, with a loss of only 3.8 mg/L of free SO_2 in 29 h. In contrast, the loss of free SO_2 increased to 17.7 mg/L with the addition of polyphenols. However, it is proposed that auto-oxidation of SO_2 does not exist in wine because polyphenols can intercept the pathway of SO_2 auto-oxidation by scavenging free radicals (Danilewicz, 2007).

5.2 | Reaction mechanism of SO_2 as an antioxidant

SO_2 reacts with quinones and H_2O_2 formed during wine oxidation and protects the wine from oxidative damage.

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The ability of SO_2 to react with quinone is demonstrated by other nucleophiles such as BSA and azide (Danilewicz, 2011a). The reaction of polyphenols with oxygen is thermodynamically unfavorable in a wine-like environment, but SO_2 increases the rate of polyphenol oxidation by reacting with quinone, the product of the above reaction (Danilewicz et al., 2008). From this perspective, SO_2 promotes the oxidation of polyphenols. Simultaneously, SO_2 reduces the quinone back to the polyphenol either partially or almost entirely, depending on the type of polyphenol (Danilewicz & Wallbridge, 2010). For example, the amount of (+)-catechin remained virtually unchanged before and after oxidation (96% reduced), whereas 20% of the (-)-epicatechin was oxidized. This different result might be related to the type or structure of the polyphenol. By reacting with aldehydes or ketones, SO_2 prevents the formation of pigments and protects aroma substances; therefore, SO_2 reduces the loss of thiol, which is a family of wine aroma constituents (Ma & Waterhouse, 2018; Nikolantonaki et al., 2014).

Although the direct reaction with oxygen is blocked by Pauli's exclusion principle, SO_2 is still able to react with H_2O_2, the product of the reduction of O_2. H_2O_2 is a strong oxidant and could be used for accelerated oxidation experiments (Celotti et al., 2022). By reacting with H_2O_2, SO_2 prevents the oxidation of ethanol by the Fenton reaction. In the case of (+)-catechin and (-)-epicatechin, SO_2 was capable of reacting with all H_2O_2 produced via O_2 reduction, and no formation of bound SO_2 was observed. It is proposed because it prevents the production of acetaldehyde through the Fenton reaction (Danilewicz & Wallbridge, 2010).

5.3 | Ratio in the reaction of SO_2 with oxygen

As mentioned before, total SO_2 should be considered when discussing reaction ratios with oxygen. The addition of different concentrations of 4-MeC or (+)-catechin to oxygen-saturated model wine and results indicated that the rate of oxidation of SO_2 increased with an increase in polyphenol concentration. This observation demonstrated that the oxidation of SO_2 depends on the concentration of polyphenols. The proposed mechanism for the reaction of SO_2 with H_2O_2 and quinone, rather than directly with oxygen, was supported (Danilewicz, 2007; Danilewicz & Wallbridge, 2010). In model wine, the ratio of SO_2 to O_2 reaction is close to 2:1 (Danilewicz et al., 2008).

In the preliminary investigations of the reaction between SO_2 and O_2 in wine, the anticipated 2:1 ratio was not achieved. Instead, when accounting for the total SO_2 content, the ratio only reached 1.73:1. Furthermore, when focusing solely on free SO_2, the ratio was even lower at 0.8:1 after 89 h of reaction. This was inconsistent with the proposed mechanism whereby SO_2 reacts with H_2O_2 formed by the reduction of O_2 and quinone formed by the oxidation of polyphenols, respectively (Danilewicz, 2015). With high Fe(II) content in the model at the beginning of the reaction, SO_2 reacts with H_2O_2, in a ratio close to 1:1. This suggests that the rapid reaction of SO_2 and H_2O_2 in the model can prevent ethanol from being oxidized by inhibiting Fenton reaction oxidation to produce acetaldehyde. As the reaction proceeds, the polyphenol starts to oxidize, and quinone is captured by SO_2, at which time the reaction ratio of SO_2 and oxygen returns to 2:1 (Danilewicz, 2011a). The reaction ratio between SO_2 to O_2 in real wine is less than 2:1, probably because of the time of monitoring, and some of the reactions have not yet reached equilibrium. Danilewicz and Standing (2018) found that in some real wines the initial ratio of SO_2:O_2 was indeed 1:1, but if given enough time (10-19 days, varying from wine to wine) the ratio reached 2:1, especially, when a sufficient amount of SO_2 not present in wine. It could be explained by the conversion of SO_2 form. When SO_2 combines with quinone, some of the adduct dissociates to form sulfate and polyphenols, depending on the type of polyphenol. However, this part of the bound SO_2 eventually forms sulfate, which means SO_2 was fully oxidized. Therefore, if the antioxidant action of SO_2 is in progress, the measured SO_2:O_2 turns out to be less than 2:1. When free SO_2 is completely consumed, bound SO_2, such as SO_2-acetaldehyde, replenishes free SO_2 by dissociation (Tachtalidou et al., 2022).

However, wines have a complex composition, and it has been suggested that not only SO_2 can react with oxidation products, but other substances can also reduce the SO_2 to O_2 ratio by reacting with oxygen, though their observations were made over a period of fewer than 10 days, and some of the bound SO_2 has not yet been fully oxidized (Carrascón et al., 2018). In some wines it was found that when the wine was saturated with air several times, the SO_2 consumption at the first saturation was significantly lower and the SO_2:O_2 molar ratio was only 0.759 at this time, which the authors explained as perhaps some amino acids reacting instead of SO_2 (Carrascón et al., 2018). Taking this aspect into consideration, the next step of investigation pertains to determining whether the nucleophilic reactivity of SO_2 is more potent than that of the compounds present within the wine.

6 | EFFECTS OF OXIDATION ON WINE QUALITY

The quality of wine is to some extent related to consumer preferences. Taste and appearance, in other words, the

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sensory experience, are important indicators of consumer choice of wine (Charters & Pettigrew, 2007). Cabernet Sauvignon wines with varying levels of oxidation (by 0, 25, and 50 mL L^{-1}month^{-1} oxygen exposure) were preferred differently by Australian consumers. 40% of consumers preferred moderately oxidized wines (25 mL L^{-1} month^{-1} oxygen exposure), followed by 31% of consumers who preferred highly oxidized wines (50 mL L^{-1}month^{-1} oxygen exposure), and 29% who preferred non-oxidized wines (Parpinello et al., 2012). The results suggest that moderate oxidation (25 mL L^{-1} month^{-1}) affects the olfactory complexity of wines, thus positively influencing consumer choice. This result is in line with the study of varietal wines, where usually consumers preferred blended varietal wines because of the higher sensory complexity compared to single varietal wines (Wang & Spence, 2019). The impact on color is readily apparent, as oxidation can lead to the browning of white wines and the deepening of red hues in red wines. In terms of sensory properties, oxidation introduces subtle nuances that can enhance or detract from a wine's profile. Positive effects include the development of complex aromas such as nuttiness and caramel, contributing to the bouquet. However, excessive oxidation can result in undesirable traits, such as a loss of fruitiness, a flat palate, and the emergence of off-flavors resembling sherry or vinegar (Liu et al., 2023). It is crucial to note that the manifestation of these favorable and unfavorable aromas in wine is variably influenced by grape varietals. In a study, Guo et al. (2022) observed that dry Cabernet Sauvignon wines undergo a decline in specific aroma compounds during aging. Interestingly, this transformation is accompanied by a favorable enhancement of key elements such as furfuryl alcohol, furfural, and 5-methylfurfural, all contributing to the emergence of desirable aged aromas. Notably, benzyl alcohol, responsible for the creation of a delightful nutty aroma, also experiences a positive influence throughout the aging process. Additionally, in the intricate interplay of chemical reactions and sensory attributes, acetaldehyde emerges as a crucial contributor (Culleré et al., 2007).

Bueno et al. (2010) observed that the sensory attributes of white wines undergo a gradual transformation during oxidation, exhibiting rates of change that can vary by up to threefold. On the contrary, for red wines, oxidation tends to induce swift sensory alterations, typically resulting in rapid differentiation of the sample, followed by a subsequent "stabilization" of the rate of change, oxidation rate k value from 0.171-192.3. In the study by Escudero et al. (2000), it was observed that a youthful white wine, following spontaneous oxidation, exhibited a pronounced off-flavor evocative of cooked vegetables. The analysis revealed a notable rise in methional concentration in wines augmented with both methionol and methionine. This implies that methional could be generated through the direct peroxidation of methionol or via Strecker degradation of methionine, potentially mediated by *o*-quinones formed during the oxidative processes in wine. Similarly, Mayr et al. (2015) observed the increase in concentration of methional during oxidative study of 14-year old white wine using synthetic closures, natural corks, and screw caps at 15°C.

Ferreira's investigation into the browning process and volatile compound evolution in oxygen-exposed white wines (Macabeo and Chardonnay grapes) revealed significant alterations in fatty acids and fermentation esters. These compounds underwent notable changes due to oxidative storage, and these transformations were linked to the equilibrium of acid-ester hydrolysis. Volatile phenols exhibited diverse behavior, including varying trends in phenol, 4-ethylphenol, eugenol, and the disappearance of 4-vinyl-guaiacol (Ferreira et al., 1997). Moreover, the browning capacity was found to be dependent on the phenolic composition, including flavonols and hydroxycinnamic acids, as well as other parameters like pH, SO_2, Fe, and storage temperature (Fernandez-Zurbano et al., 1995). In a subsequent study, Fernández-Zurbano et al. (1998) found that hydroxycinnamic acids and esters decreased significantly during the oxidation of eight white wines. Additionally, a correlation ($r > 0.567$) was identified between the flavanol contents of the wines and the extent of browning at the conclusion of the oxidation process. Notably, the phenols displayed similar behavior during storage at both 20 and 50°C.

The study by Bueno et al. (2018) on red wine oxidation revealed that acetaldehyde accumulation in aged wines is linked to SO_2 content, epigallocatechin, and polymerization degree, but inversely related to aldehyde reactive polyphenols (ARPs). Strecker aldehydes increase proportionally with amino acid precursors, notably in aged wines, except for phenylacetaldehyde. The study highlighted the pivotal role played by ARPs in the accumulation of aldehydes, emphasizing the distinct reactivity patterns of phenylacetaldehyde. In a seminal investigation conducted by Carrascón et al. (2018), the kinetics of oxygen and SO_2 consumption in air-saturated red wine were thoroughly examined. The findings revealed that oxygen consumption rates (OCRs) are faster with higher copper and epigallocatechin contents and with higher absorbance at 620 nm and slower with higher levels of gallic acid and catechin terminal units in tannins. It was also found that SO_2 was poorly consumed during the initial saturation due to the low availability of free SO_2 caused by a high anthocyanin/tannin ratio, as well as by a polyphenolic profile that is deficient in epigallocatechin and rich in catechin-rich tannins.

Ferreira et al. (2015) studied the kinetic modeling of oxygen consumption by 15 Spanish red wines that were oxidized along five consecutive air saturation cycles at 25°C. The results indicated that the variability of

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oxygen concentration at saturation at the beginning of each cycle was independent of the wine and gave an estimated value of 0.18 mg of oxygen/L. These variabilities can be associated with small random differences in the exact time of measurement of the first saturation point and differences in wine viscosities affecting the rate of degassing of microbubbles. Furthermore, they found that initial oxygen consumption rates were negatively correlated ($p < 0.05$) with the chemical composition (such as gallic acid, phenolic acid, catechin, and total flavanols, etc.) of the wines before oxidation. A highly significant negative correlation was observed ($R^2 = 0.67$, significant at $p < 0.001$) between the amounts of SO_2 consumed in the first saturation and the initial oxygen consumption rates. Similarly, a previous study by Ferreira et al. (2014) mentioned that the oxidation of 16 Spanish red wines during storage for 6 months at 25°C, under different levels of oxygen (0–56 mg/L), led to a sensory-relevant increase in Strecker aldehydes, 1-octen-3-one, and vanillin. Furthermore, the formation rates of Strecker aldehydes such as methional (r = 0.62), phenylacetaldehyde (r = 0.56), 2-methylpropanal (r = 0.43), 2-methylbutanal (r = 0.41), and 3-methylbutanal (r = 0.76) were correlated with the combined SO_2 levels during oxidation.

Mislata et al. (2020) investigated the influence of oxidation on the aromatic composition and sensory attributes of Rioja red aged wines. The wine samples underwent oxidation for one month in darkness at a controlled temperature of 23 ± 2°C. The findings revealed a substantial decrease in total fermentative aromas (40% loss) and a corresponding increase in oxidative aromas (85%) following the one-month storage period. Notably, esters, fatty acids, acetates, and alcohols exhibited significant reductions of 47%, 52%, 19%, and 12%, respectively. The pronounced decline in esters and fatty acids is attributed to the extensive oxidation occurring under the specified conditions. Esters, commonly associated with fruit aromas, proved highly susceptible to oxidation. Specific aroma compounds, such as 3-methyl-2,4-nonanedione and 2-phenylacetaldehyde, were identified in higher concentrations, while 3-methylbutanal, (E)−2-nonenal, and (E)−2-octenal were found in lesser amounts, emerging as crucial sensory active compounds. Similarly, certain aromatic compounds, including 2,3-butanedione, 3-hexenol, β-citronellol, and geraniol, exhibited decreased concentrations, while a substantial increase was observed in compounds indicative of wine evolution, such as 2-methylpropanal, 3-methylbutanal, methional, and phenylacetaldehyde, following the oxidation of red wines (Tania et al., 2011).

Dai, Zhong et al. (2022) investigated the intricate roles and interactions between *Saccharomyces cerevisiae* and lactic acid bacteria (LAB) concerning acetaldehyde accumulation during red wine micro-oxygenation (Mox) treatment. The findings revealed the pivotal role of Mox in sustaining *S. cerevisiae* populations, thereby deferring the onset of spontaneous malolactic fermentation. Notably, *S. cerevisiae* exhibited a propensity for acetaldehyde production during Mox, with strain-dependent variations. Production rates exhibited a decline throughout the treatment, and post-treatment, acetaldehyde levels experienced a subsequent decrease. It was elucidated that the acetaldehyde stemming from chemical oxidation continued to accrue during Mox. In alignment with the research conducted by Han et al. (2019), the concentration of acetaldehyde at the bottling stage emerged as a critical determinant influencing the phenolic compound profile of Cabernet Sauvignon wines after 1 year of Mox. The impact was particularly pronounced on anthocyanins, followed by flavonols, flavonoids, and hydroxycinnamic acids, while benzoic acids remained unaffected. The study also emphasized that higher acetaldehyde levels and increased oxygen ingress resulted in elevated levels of heterocyclic acetals derived from glycerol.

7 | CONCLUSION

This review paper presents a comprehensive examination of the intricate mechanisms underpinning the initial phases of non-enzymatic wine oxidation, elucidating the roles played by polyphenols, oxygen, iron, copper, and sulfur dioxide. Furthermore, it sums up the impact of quinone on wine composition, the relationship between O_2 content and oxidation, investigates the interplay between Fe and Cu, and assesses the consistency of the SO_2:O_2 reaction ratio with the proposed oxidation.

It can be observed that the current discussion of the wine oxidation mechanism is mostly based on a modeling approach, which facilitates a clearer elaboration of the possible reaction processes in wine and allows for the comparison of reaction rates. Model wines, while providing valuable insights can only represent real wines to a certain extent. Future studies should emphasize more on obtaining results that are consistent between model and real wines. For this purpose, innovative analytical methods may be needed to identify intermediates in the oxidation process and to understand more fully the non-enzymatic oxidation mechanisms in wine.

AUTHOR CONTRIBUTIONS

Guanghao Wang: Conceptualization; investigation; funding acquisition; writing—original draft; formal analysis; visualization; methodology; writing—review and editing; data curation; supervision; resources. **Yogesh Kumar**: Writing—review and editing; investigation; conceptualization; methodology; writing—original draft.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflict of interest.

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