

Short communication

Determination of the iron state in ferrous iron containing vitamins and dietary supplements: Application of Mössbauer spectroscopy

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Abstract

Determination of the iron state in commercially manufactured iron containing vitamins and dietary supplements is important for evaluation of pharmaceuticals quality. Mössbauer (nuclear gamma-resonance) spectroscopy was used for analyzing the iron state in commercial pharmaceutical products containing ferrous fumarate ($\text{FeC}_4\text{H}_2\text{O}_4$), ferrous sulfate (FeSO_4), ferrous bisglycinate chelate (Ferrochel[®]) and ferrous iron (hydrolyzed protein chelate). Mössbauer parameters and the iron states were determined for iron compounds in the studied pharmaceuticals. Various ferric and ferrous impurities were found in all of the commercial products. The quantities of ferric impurities exceeded the FDA limitation of 2% in products containing ferrous fumarate. The quantities of ferric impurities exceeded 58% and 30% in products containing ferrous bisglycinate chelate and ferrous iron (hydrolyzed protein chelate), respectively. The presence of ferrous and ferric impurities was not related to the ageing of the vitamins and dietary supplements. Two pharmaceutical products contained major iron compounds, the Mössbauer parameters of which did not correspond to the ferrous fumarate or ferrous bisglycinate chelate claimed by the manufacturer.

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1. Introduction

Iron is one of the vitally important metals. Iron ions are found in the active sites in hemoglobin, myoglobin, cytochromes, catalase and many other proteins that realize oxygen and electron transport and enzyme functions. Therefore, disorders of iron metabolism are among the most common diseases of humans [1]. For instance, iron deficiency causes anemia and other pathological changes in the body, and anemia remains an important public health problem [2]. To prevent and treat iron deficiency, some iron containing pharmaceutical products are used. These medicines may be oral iron containing vitamins and dietary supplements [3–8] or injectable iron containing pharmaceuticals [9–11]. The knowledge of the iron states in these products is very important because the valence/spin state of iron may be related to the effect and toxicity of medicines. For instance, fer-

rous iron compounds are more readily bioavailable than ferric ones for oral iron containing supplements [12,13]. The toxic side effects of oral medicines were more severe with ferric salts because the absorption of ferric iron was relatively slower than ferrous based formulations [14]. In general, the quality of iron-containing pharmaceuticals may also depend on both the chemical state of iron in the main iron compound and the presence of iron containing impurities. In accordance with the FDA requirements (Food and Drugs, Sec. 172.350) ferrous fumarate ($\text{FeC}_4\text{H}_2\text{O}_4$), which is widely used in oral iron-containing vitamins and dietary supplements, should not contain more than 2% of ferric iron. Hindustan Basic Drugs, one of the ferrous fumarate manufacturers, announced at their web site [15] that premium grade ferrous fumarate contained lower than 1% of ferric compounds. The V.V. Turakhia & Group of Companies announced the presence of 1.17% of ferric iron in their ferrous fumarate [16]. Some other ferrous fumarate manufacturers, such as Jost Chemicals and Kedia Chemicals Industries Ltd., indicated at their web sites [17,18] that the presence of ferric iron was not higher than 2%. Thus, an analysis of the iron state, iron

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compounds and their quantities in iron containing medicines is useful for the quality control of these pharmaceuticals.

It is well known that Mössbauer (nuclear gamma-resonance) spectroscopy [19–21] is the most sensitive technique for the analysis of iron state. Mössbauer spectroscopy permits the observation of the hyperfine splitting of the nuclear energy levels as well as changes of energies of the ground and excited states for Mössbauer isotopes, for instance, ^{57}Fe , in the absorption or emission spectrum of gamma-rays. Mössbauer parameters isomer shift (δ) and quadrupole splitting (ΔE_Q) give information about the iron electronic structure, valence and spin state of iron while absorption areas of subspectra (S) give information about relative content of various iron containing species in the sample. Mössbauer spectroscopy has been shown to be a useful technique for various biomedical researches, in particular, for analysis of pharmaceutical products containing Mössbauer isotopes [22–25].

A number of studies of ferric iron–dextran and iron–polysaccharide complexes, which were developed or produced as medicines for the treatment of iron deficiency anemia, demonstrated the potential of Mössbauer spectroscopy to characterize the iron state, find the differences of the iron hyperfine parameters and observe ferrous impurities in ferric pharmaceuticals [26–33]. Ferrous iron containing medicines were also studied by Mössbauer spectroscopy [14,25,34–36] and various ferrous and ferric impurities were found by Mössbauer spectroscopy in medicines containing ferrous sulfate and ferrous fumarate [14] (7–9% of ferric compounds in the samples containing ferrous fumarate were attributed to ferric fumarate). Approximately 8% of ferric iron was also found in ferrous fumarate (FemironTM) in ref. [34] while two ferrous sulfates did not contain ferric impurities. These values of ferric impurities in ferrous fumarate exceeded the FDA limit. However, there were no further studies of ferrous iron containing pharmaceuticals. Therefore, a new study of ferrous iron containing medicines for the control of iron state was begun [35,36]. Here the results of a Mössbauer spectroscopic study of the iron state and iron impurities in various

vitamins and dietary supplements containing ferrous fumarate, ferrous sulfate and ferrous chelates are described.

2. Materials and methods

Various industrial samples of multiple vitamins and dietary supplements with iron in the form of ferrous fumarate, ferrous bisglycinate chelate (Ferrochel[®] from Albion Laboratories, USA), ferrous iron (hydrolyzed protein chelate) and ferrous sulfate listed in Table 1 were studied. All studied vitamins contained 18 mg of iron per tablet while dietary supplements contained 6 mg of iron (Nutralite), 8.3 mg of iron (Your Life[®] Maximum Pak[®]), 27 mg of iron (Women's Ultra Mega[®]) and 50 mg of iron (Feosol[®]) per tablet. Samples were used as powder prepared from about 1/2 of each product tablet. The effective thickness of the vitamin samples varied from 2 to 6 mg Fe/cm². Samples of My Favorite[®] Multiple and Your Life[®] Maximum Pak[®] contained 1000 and 200 mg of Ca, respectively, that led to the decrease of the absorption effect especially for My Favorite[®] Multiple. The effective thickness of the Your Life[®] Maximum Pak[®] sample was about 2 mg Fe/cm². The sample of Feosol[®] was prepared from small black granules containing ferrous sulfate extracted from the capsule of the product. The effective thickness of the Feosol[®] sample was about 7–8 mg Fe/cm².

Mössbauer spectra were measured with the constant acceleration computerized precision spectrometer that was a part of the multi-dimension parametric Mössbauer spectrometer SM-2201 [37]. The noise of velocity signal of the spectrometer was 1.5×10^{-3} mm/s, the drift of the zero point velocity was $\pm 2.6 \times 10^{-3}$ mm/s, the non-linearity of the velocity signal was 0.01%, the harmonic distortion factor was 0.005% for the frequency band in the range 0–1120 Hz. These characteristics of the driving system indicate the high stability and sensitivity of spectrometer. Vitamin spectra were measured over the periods from several hours to one day, depending on the absorption effect. There were no changes in the velocity resolution during all periods of spectral measurement. The

Table 1
Pharmaceutical products

Sample	Manufacturer	Expiry date	Announced iron compound
One Daily Plus Iron	Western Family Foods, Inc., USA	04.1992	Ferrous fumarate ($\text{FeC}_4\text{H}_2\text{O}_4$)
Vita Fresh [®]	Vita-Fresh Vitamin Co., Inc., USA	09.1998	Ferrous fumarate ($\text{FeC}_4\text{H}_2\text{O}_4$)
My Favorite [®] Take One TM	Natrol, Inc., USA	12.2000	Ferrous fumarate ($\text{FeC}_4\text{H}_2\text{O}_4$)
Centrum [®]	Whitehall–Robins Healthcare, USA	03.2001	Ferrous fumarate ($\text{FeC}_4\text{H}_2\text{O}_4$)
Maximum One Daily	Leiner Health Products, Inc., USA	04.2001	Ferrous fumarate ($\text{FeC}_4\text{H}_2\text{O}_4$)
Essential Balance [®]	Nature Made Nutritional Products, USA	08.2001	Ferrous fumarate ($\text{FeC}_4\text{H}_2\text{O}_4$)
Multiple Vitamins With Iron I	Walgreen Co., USA	07.2002	Ferrous fumarate ($\text{FeC}_4\text{H}_2\text{O}_4$)
Multiple Vitamins With Iron II	Walgreen Co., USA	01.2004	Ferrous fumarate ($\text{FeC}_4\text{H}_2\text{O}_4$)
Daily-Vitamin With Iron	Windmill TM Health Products, USA	11.2002	Ferrous fumarate ($\text{FeC}_4\text{H}_2\text{O}_4$)
Therapeutic M	Walgreen Co., USA	02.2003	Ferrous fumarate ($\text{FeC}_4\text{H}_2\text{O}_4$)
Nutralite	Amway Corp., USA	04.2004	Ferrous fumarate ($\text{FeC}_4\text{H}_2\text{O}_4$)
My Favorite [®] Multiple	Natrol, Inc., USA	12.2000	Ferrous bisglycinate chelate (Ferrochel [®] from Albion Laboratories, USA)
Women's Ultra Mega [®]	General Nutrition Corp., USA	10.2003	Iron (hydrolyzed protein chelate)
Your Life [®] Maximum Pak [®]	P. Leiner Nutritional Products, Inc., USA	03.1992	Ferrous sulfate (FeSO_4)
Feosol [®]	SmithKline Beecham Corporation, USA	11.1995	Ferrous sulfate (FeSO_4)

line widths of the Mössbauer spectra of the standard absorber of sodium nitroprusside, measured for the control of spectrometer velocity resolution and reproducibility, were in the range of $(0.247\text{--}0.254) \pm 0.029$ mm/s for all measurements. The $(0.5\text{--}0.3) \times 10^9$ Bq $^{57}\text{Co}(\text{Cr})$ source was used at room temperature.

Mössbauer spectra of the vitamins and dietary supplements were measured at room temperature in transmission geometry with moving absorber. Measurements were made between 2002 and 2004 to study fresh and outdated medicines. Samples of Essential Balance[®] were measured twice in 2002 and 2004. Mössbauer spectra of all of the samples were computer fitted with the least squares procedure using Lorentzian line shape. Mössbauer hyperfine parameters (δ and ΔE_Q) as well as line width Γ , subspectrum area S , absorption effect and statistical

criteria χ^2 , were determined. The values of isomer shift are given relative to $\alpha\text{-Fe}$ at 295 K.

3. Results

Mössbauer spectra of outdated and fresh pharmaceutical products containing ferrous fumarate are shown in Figs. 1 and 2, respectively. These spectra were fitted with various quantities of components and better fitting was determined using lower χ^2 value and the physical sense of parameters. For instance, values of χ^2 were 3.96 for one quadrupole doublet fit and 1.12 for two quadrupole doublets fit for the spectrum of Maximum One Daily (Fig. 1d). The results of the better fitting procedures are given in Table 2. Mössbauer spectra of Vita Fresh[®], Maximum One Daily, Centrum[®], One Daily Plus Iron,

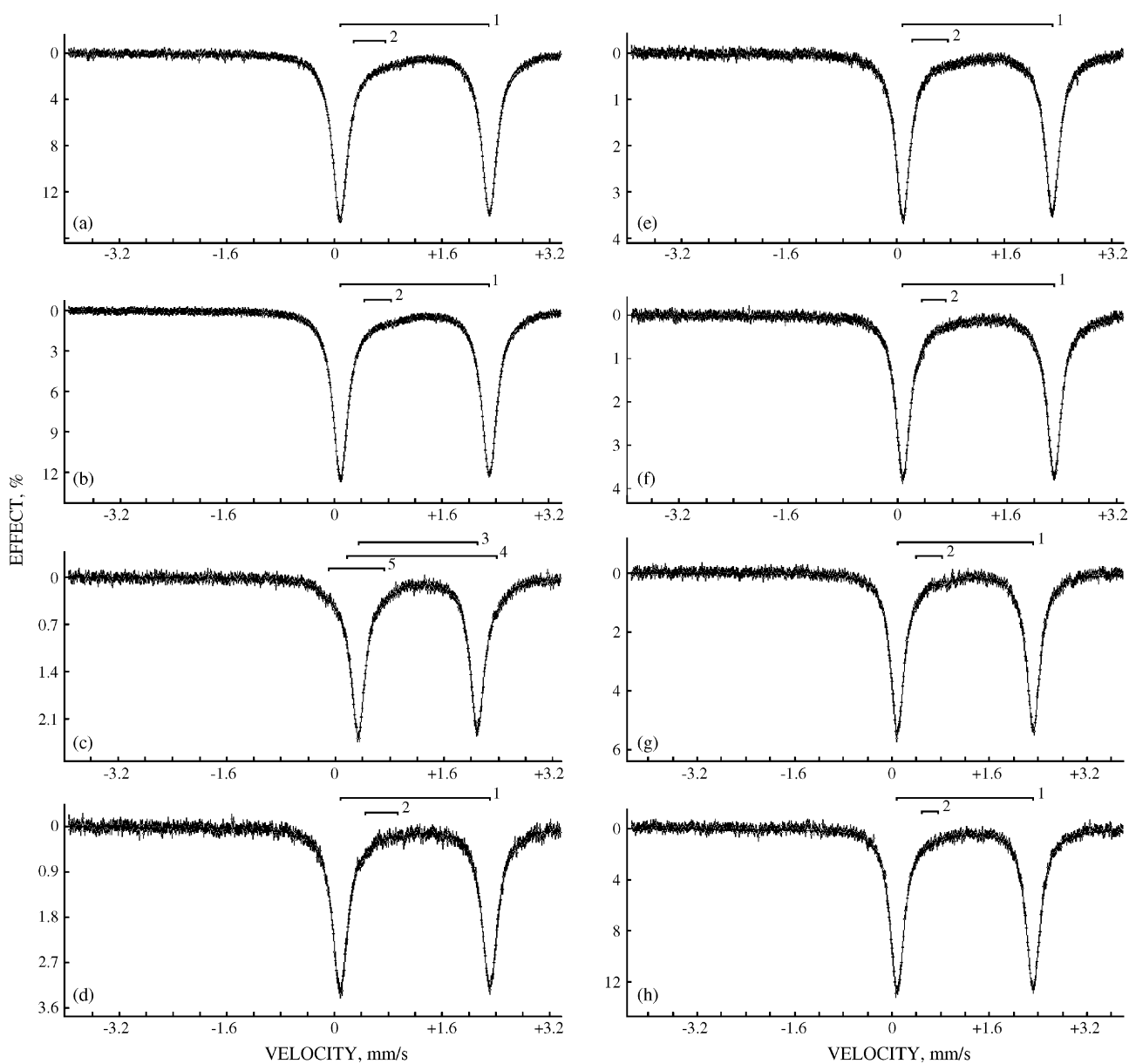


Fig. 1. Mössbauer spectra of outdated vitamins containing ferrous fumarate: (a) One Daily Plus Iron, (b) Vita Fresh[®], (c) My Favorite[®] Take One[™], (d) Maximum One Daily, (e) Centrum[®], (f) Essential Balance[®] (2002), (g) Essential Balance[®] (2004) and (h) Daily-Vitamin With Iron. (1) Ferrous fumarate, (2) ferric impurity, (3) unknown ferrous compound, (4) ferrous impurity and (5) ferric impurity. $T = 295$ K.

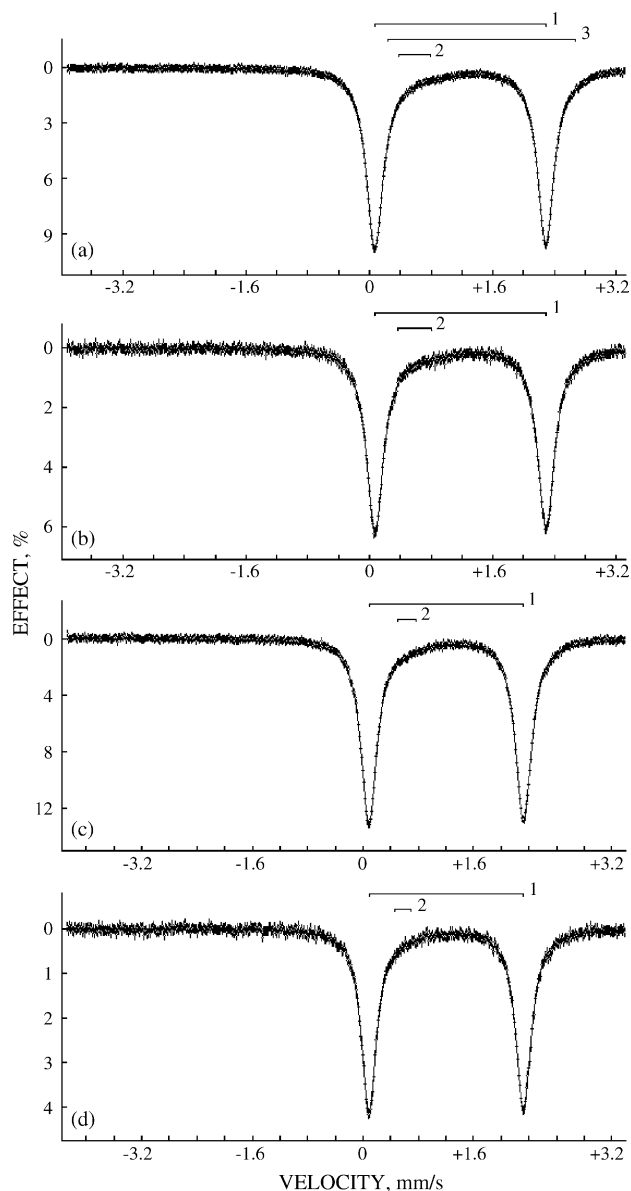


Fig. 2. Mössbauer spectra of fresh vitamins containing ferrous fumarate: (a) Multiple Vitamins With Iron I, (b) Therapeutic M, (c) Multiple Vitamins With Iron II and (d) Nutrilite. (1) Ferrous fumarate, (2) ferric impurity and (3) ferrous impurity. $T = 295$ K.

Essential Balance[®] (2002 and 2004), Nutrilite, Therapeutic M, Multiple Vitamins With Iron II and Daily-Vitamin With Iron contained a major component 1 with the same hyperfine parameters, within the experimental error, that corresponded to the high spin ferrous compound related to ferrous fumarate. The first Mössbauer study of pure ferrous fumarate made 35 years ago demonstrated parameters which were $\delta = 1.16 \pm 0.01$ mm/s and $\Delta E_Q = 2.18 \pm 0.01$ mm/s [38]. The next study of pure ferrous fumarate (from ICN) showed parameters $\delta = 1.24$ mm/s and $\Delta E_Q = 2.20$ mm/s [34]. These parameters were close to the present results. The minor component 2 of spectra shown in Figs. 1 and 2 had slightly different hyperfine parameters that corresponded to the high spin ferric compound. This component may be considered as an impurity or result of the oxidation of

ferrous fumarate. The previous Mössbauer study of medicines containing ferrous fumarate [14] showed the same hyperfine parameters for ferrous fumarate while a minor component was approximated with singlet line related to ferric fumarate. Mössbauer parameters for ferrous fumarate obtained in ref. [34] also were the same as in ref. [14] but with a minor component of the ferric compound (8%) that was approximated with quadrupole doublet. However, the Mössbauer hyperfine parameters for the ferric impurity obtained in ref. [34] ($\delta = 0.37$ mm/s and $\Delta E_Q = 0.88$ mm/s) were significantly different than those obtained for component 2 in the present study. It should be noted that the values of Γ for ferric impurities were broader in some cases, for instance, for samples of Centrum[®], One Daily Plus Iron and Multiple Vitamins With Iron I. This fact may be a result of the presence of more than one ferric compound or the distribution of similar ferric compounds, however, the fit of the Mössbauer spectra with more components was not successful.

The Mössbauer spectrum of Multiple Vitamin With Iron I (Fig. 2a) consisted of three components. Components 1 and 2 were the same as in the above mentioned vitamin spectra. Mössbauer hyperfine parameters of the other minor component 3 corresponded to the high spin ferrous compound and differed from those of ferrous fumarate. However, the Mössbauer spectrum of My Favorite[®] Take OneTM (Fig. 1c) appeared to be different from other vitamin spectra. This spectrum consisted of three components. The minor component 4 had Mössbauer parameters that were close to ferrous fumarate (ΔE_Q was the same within the experimental error while δ was slightly higher). The major component 3 had Mössbauer parameters that corresponded to the unknown high spin ferrous compound with a lower ΔE_Q value than that of ferrous fumarate. Another minor component 5 had hyperfine parameters that corresponded to the high spin ferric compounds, however, the ΔE_Q value was significantly higher while δ was significantly lower than those of ferric compounds 2. Refitting of this spectrum with other initial parameters showed some changes of the Mössbauer parameters for ferrous and ferric impurities: $\delta = 1.17 \pm 0.014$ mm/s, $\Delta E_Q = 2.401 \pm 0.014$ mm/s and $\delta = 0.479 \pm 0.014$ mm/s, $\Delta E_Q = 0.497 \pm 0.014$ mm/s, respectively. Relative areas of spectral components were almost the same as in Table 2 while χ^2 slightly increased (1.162 in comparison with 1.156 for results from Table 2). Both fits showed the presence of the same quantities ($\sim 10\%$ and $\sim 6\%$) of high spin ferrous and ferric impurities in the sample of My Favorite[®] Take OneTM. However, the Mössbauer hyperfine parameters obtained in the first fitting of the spectrum of My Favorite[®] Take OneTM given in Table 2 for ferric compound 5, were close to those obtained for the ferric impurity observed previously [34] ($\delta = 0.37$ mm/s and $\Delta E_Q = 0.88$ mm/s). This uncertainty is a result of the difficulties in fitting very weak peaks overlapping with high intensity peaks in the spectrum.

Mössbauer spectra of outdated and fresh pharmaceutical products containing ferrous chelates are shown in Fig. 3. The better fit of these spectra showed the presence of three components. Mössbauer parameters with a better fit are given in Table 3. The major component 3 of the spectrum of My Favorite[®] Multiple had parameters that corresponded to the high spin ferric

Table 2

Mössbauer parameters of pharmaceutical products containing ferrous fumarate measured at 295 K

Samples	Γ (mm/s)	δ (mm/s)	ΔE_Q (mm/s)	S (%)	Compound
One Daily Plus Iron	0.276 ± 0.029	1.191 ± 0.014	2.221 ± 0.014	92.7	Ferrous fumarate
	0.558 ± 0.029	0.522 ± 0.014	0.477 ± 0.014	7.3	Ferric high spin
Vita Fresh®	0.274 ± 0.029	1.204 ± 0.014	2.220 ± 0.014	94.4	Ferrous fumarate
	0.474 ± 0.029	0.648 ± 0.014	0.403 ± 0.014	5.6	Ferric high spin
My Favorite® Take One™	0.409 ± 0.029	1.274 ± 0.014	2.194 ± 0.014	9.9	Ferrous high spin
	0.248 ± 0.029	1.215 ± 0.014	1.751 ± 0.014	84.1	Ferrous high spin
	0.336 ± 0.029	0.317 ± 0.014	0.818 ± 0.014	6.0	Ferric high spin
Maximum One Daily	0.281 ± 0.029	1.195 ± 0.014	2.226 ± 0.014	95.8	Ferrous fumarate
	0.365 ± 0.029	0.700 ± 0.014	0.495 ± 0.014	4.2	Ferric high spin
Centrum®	0.261 ± 0.029	1.198 ± 0.014	2.222 ± 0.014	92.4	Ferrous fumarate
	0.819 ± 0.029	0.488 ± 0.014	0.526 ± 0.014	7.6	Ferric high spin
Essential Balance® (2002)	0.258 ± 0.029	1.185 ± 0.014	2.210 ± 0.014	96.0	Ferrous fumarate
	0.297 ± 0.029	0.533 ± 0.014	0.351 ± 0.014	4.0	Ferric high spin
Essential Balance® (2004)	0.268 ± 0.032	1.204 ± 0.016	2.233 ± 0.016	95.1	Ferrous fumarate
	0.370 ± 0.032	0.609 ± 0.016	0.425 ± 0.016	4.9	Ferric high spin
Daily-Vitamin With Iron	0.281 ± 0.032	1.202 ± 0.016	2.233 ± 0.016	95.8	Ferrous fumarate
	0.508 ± 0.032	0.620 ± 0.016	0.271 ± 0.016	4.2	Ferric high spin
Multiple Vitamins With Iron I	0.259 ± 0.029	1.179 ± 0.014	2.218 ± 0.014	91.1	Ferrous fumarate
	0.298 ± 0.029	1.457 ± 0.014	2.435 ± 0.014	2.9	Ferrous high spin
	0.581 ± 0.029	0.585 ± 0.014	0.409 ± 0.014	6.0	Ferric high spin
Therapeutic M	0.264 ± 0.029	1.183 ± 0.014	2.219 ± 0.014	95.3	Ferrous fumarate
	0.405 ± 0.029	0.586 ± 0.014	0.445 ± 0.014	4.7	Ferric high spin
Multiple Vitamins With Iron II	0.280 ± 0.032	1.205 ± 0.016	2.237 ± 0.016	96.4	Ferrous fumarate
	0.414 ± 0.032	0.641 ± 0.016	0.278 ± 0.016	3.7	Ferric high spin
Nutrilite	0.268 ± 0.032	1.206 ± 0.016	2.235 ± 0.016	97.6	Ferrous fumarate
	0.238 ± 0.032	0.583 ± 0.016	0.237 ± 0.016	2.4	Ferric high spin

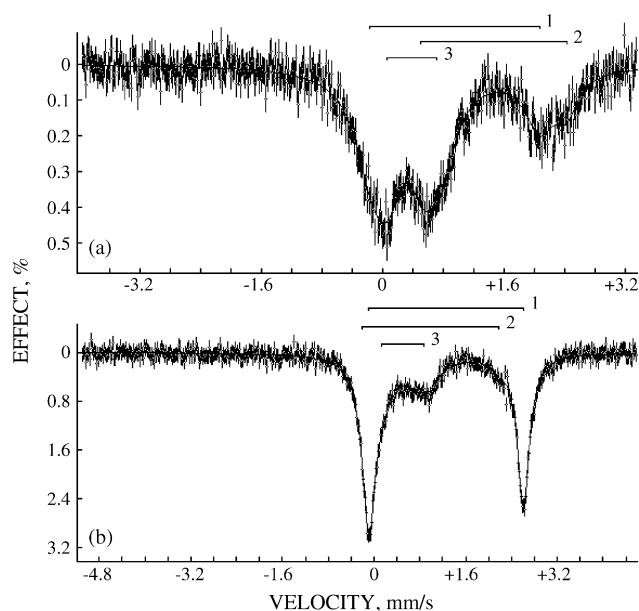


Fig. 3. Mössbauer spectra of pharmaceutical products containing ferrous chelates: (a) My Favorite Multiple® and (b) Women's Ultra Mega®. (1 and 2) Ferrous compounds and (3) ferric compound. $T = 295$ K.

compound. Other components (1 and 2) had parameters that corresponded to two different high spin ferrous compounds. The major component 1 and the minor component 2 of the spectrum of Women's Ultra Mega® had Mössbauer parameters that corresponded to the different high spin ferrous compounds while Mössbauer parameters of the component 3 corresponded to the high spin ferric compound. Unfortunately, there was no literature data on the Mössbauer study of pure Ferrochel® and iron (hydrolyzed protein chelate).

Mössbauer spectra of outdated dietary supplements containing ferrous sulfate are shown in Fig. 4. These spectra were better fitted using two components with parameters given in Table 4. Mössbauer parameters of the major component 1 in both spectra corresponded to the high spin ferrous compound with the same δ values within the experimental error and almost the same ΔE_Q values. The minor components 2 had different Mössbauer parameters that corresponded to the high spin ferric compounds. Earlier studies of medicines containing ferrous sulfate [14] showed the presence of three components: two high spin ferrous compounds (56–65% of FeSO_4 and 25–36% of $\text{FeSO}_4 \cdot \text{H}_2\text{O}$) and one high spin ferric compound (6–10% of $\text{Fe}_2(\text{SO}_4)_3$). Mössbauer hyperfine parameters of FeSO_4 obtained in ref. [14] were close to those of the major component of the spectra of Your Life® Maximum Pak® and Feosol® while Mössbauer parameters of the minor ferric compound were different from those

Table 3
Mössbauer parameters of pharmaceutical products containing ferrous chelates measured at 295 K

Samples	Γ (mm/s)	δ (mm/s)	ΔE_Q (mm/s)	S (%)	Compound
My Favorite [®] Multiple	0.493 ± 0.029	0.957 ± 0.014	2.234 ± 0.014	28.0	Ferrous high spin
	0.371 ± 0.029	1.473 ± 0.014	1.920 ± 0.014	13.5	Ferrous high spin
	0.559 ± 0.029	0.387 ± 0.014	0.656 ± 0.014	58.5	Ferric high spin
Women's Ultra Mega [®]	0.266 ± 0.038	1.254 ± 0.019	2.695 ± 0.019	63.5	Ferrous high spin
	0.255 ± 0.038	0.975 ± 0.019	2.388 ± 0.019	5.3	Ferrous high spin
	0.557 ± 0.038	0.495 ± 0.019	0.747 ± 0.019	31.2	Ferric high spin

Table 4
Mössbauer parameters of dietary supplements containing ferrous sulfate measured at 295 K

Samples	Γ (mm/s)	δ (mm/s)	ΔE_Q (mm/s)	S (%)	Compound
Your Life [®] Maximum Pak [®]	0.307 ± 0.029	1.309 ± 0.014	2.749 ± 0.014	96.0	Ferrous sulfate
	0.459 ± 0.029	0.500 ± 0.014	0.577 ± 0.014	4.0	Ferric high spin
Feosol [®]	0.355 ± 0.029	1.288 ± 0.014	2.712 ± 0.014	98.3	Ferrous sulfate
	0.289 ± 0.029	0.302 ± 0.014	0.212 ± 0.014	1.7	Ferric high spin

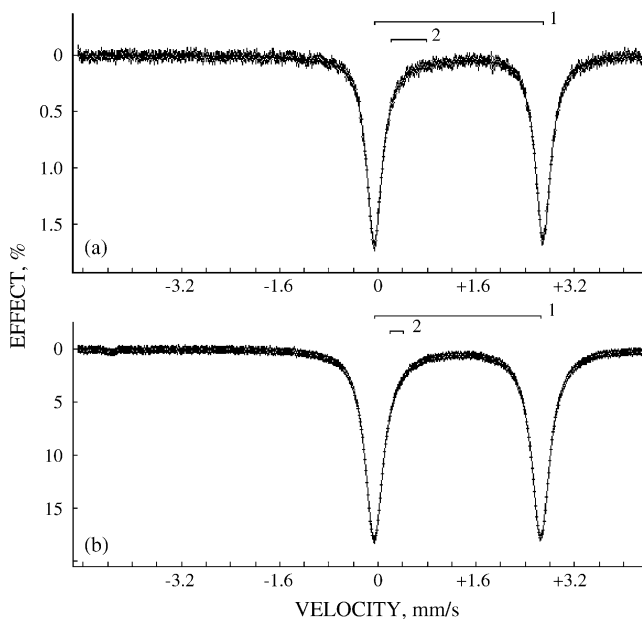


Fig. 4. Mössbauer spectra of dietary supplements containing ferrous sulfate: (a) Your Life[®] Maximum Pak[®] and (b) Feosol[®]. (1) Ferrous sulfate and (2) ferric impurity. $T = 295$ K.

observed in Your Life[®] Maximum Pak[®] and Feosol[®]. Moreover, the abundance of the ferric impurity in Your Life[®] Maximum Pak[®] and Feosol[®] was found to be significantly lower than that in the previous study [14]. Mössbauer parameters of ferrous sulfates (Eckerd and SmithKline Beecham Co.) studied previously [34] were close to our data, however, the authors related these parameters to ferrous sulfate monohydrate. They did not find any ferric impurities in these samples.

4. Discussion

Mössbauer parameters for all outdated vitamins containing ferrous fumarate were the same for ferrous fumarate and slightly

varied for ferric impurities except My Favorite[®] Take One[™] (in case of the first fitting). The quantity of ferric impurities was in the range 4.0–7.6% and did not depend on the age of the outdated vitamins. The results of two measurements of the vitamins Essential Balance[®] with an expiry date of 1 year before the first measurement were the same for ferrous fumarate while the ferric impurity had slightly different parameters. These results demonstrate the stability of ferrous fumarate over a long period of time; however, the presence of the ferric impurity (that may be a result of the manufacturing process) exceeded the FDA limit. The sample of Multiple Vitamins With Iron I contained an additional ferrous impurity. This small quantity of ferrous impurity may also be a result of the manufacturing process, but not product ageing. The quantity of the ferric impurity was 6.0% and exceeded the FDA limit. Mössbauer parameters for the fresh vitamins were the same for ferrous fumarate. The quantity of ferric impurities in these samples was less than in other vitamins and varied from 2.4% to 4.7%. However, these values also exceeded the FDA limit. In contrast to other vitamins, the sample of My Favorite[®] Take One[™] contained a major ferrous compound that was different from ferrous fumarate found in other samples. The measurement of the Mössbauer spectrum of this sample was made a year after the expiry date. Therefore, this ferrous compound cannot be considered as a result of ageing.

The Mössbauer spectra of ferrous chelates demonstrated the presence of three different iron compounds. The sample of My Favorite[®] Multiple contained a major component related to an unknown ferric compound (58.5%) instead of the claimed ferrous iron in the iron bisglycinate chelate [39]. This unusual fact may be the result of chemical transformation of the initial ferrous bisglycinate chelate during vitamin preparation due to instability of this compound because measurement was made a year after expiry date. Moreover, the presence of two different minor ferrous compounds may also be the result of instability of the ferrous bisglycinate chelate. Therefore, an additional study of stability of the iron state in Ferrochel[®] is required. The sample of

Women's Ultra Mega[®] contained a major ferrous compound that may be related to the iron (hydrolyzed protein chelate). However, the presence of 31.2% of ferric compound in the fresh sample indicated the instability of iron (hydrolyzed protein chelate). Thus, pharmaceutical products containing ferrous chelates as announced by manufacturers actually contained large amount of ferric compounds.

Mössbauer spectra of Your Life[®] Maximum Pak[®] and Feosol[®] containing ferrous sulfate showed the presence of ferrous sulfate in the form of the monohydrate in accordance to previous work [34] and only small quantities of ferric compounds in spite of the analysis being conducted a long period after the expiry date. This result indicates a high degree of stability of the ferrous sulfate in these dietary supplements.

5. Conclusion

The results obtained demonstrate the presence of several ferrous and ferrous impurities in industrial samples of iron containing vitamins and dietary supplements. The quantities of ferric impurities exceeded the FDA limit for ferrous fumarate. Moreover, it was found that two products from Natrol, Inc., USA (My Favorite[®] Take One[™] and My Favorite[®] Multiple) contained major iron compounds the Mössbauer parameters of which did not correspond to the ferrous fumarate and ferrous bisglycinate chelate, claimed by the manufacturer. Therefore, Mössbauer spectroscopy can be a useful tool of the control over the iron state in iron containing pharmaceuticals.

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