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RAMAN SPECTROSCOPIC TECHNIQUES FOR MEAT ANALYSIS: A REVIEW

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Keywords: *Raman spectroscopy, nondestructive analysis, meat quality, meat products*

Abstract

Raman spectroscopy (vibrational spectroscopy) proved to be an effective analytical approach in the field of geology, semiconductors, materials and polymers. Over the past decade, Raman spectroscopy has attracted the attention of researchers as a non-destructive, highly sensitive, fast and eco-friendly method and has demonstrated the unique capabilities of food analysis. The use of Raman spectroscopic methods (RSMs) to assess the quality of meat and finished products is rapidly expanding. From the analysis of one sample, you can get a large amount of information about the structure of proteins, the composition of fatty acids, organoleptic parameters, autolysis and spoilage indicators, authentication of raw materials, technological properties. An important advantage of the method is the comparability of the results obtained with the data of traditional analytical methods. Traditional methods of determining the quality of meat are often time-consuming, expensive and lead to irreversible damage to a sample. It is difficult to use them in production conditions directly on the meat processing lines. Technological advances have made it possible to develop portable Raman spectroscopes to use directly in production. The article presents the basic principles of Raman spectroscopy, systematizes the results of the use of RSMs for the analysis of meat quality from different types of slaughter animals and provides tools for analyzing the data of the obtained spectra. Raman spectra have many dependent variables, so chemometric assays are used to work with them. Literature analysis has shown that currently there is no unified database of meat spectra in the world, standardized protocols for conducting research and processing the obtained results. In Russia, the use of RSMs is a new, promising and relevant area of research in the field of meat quality.

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Introduction

With the growth in global import and export of agri-food products, the questions of food safety have received increased attention worldwide.

Meat is the main protein source and has a great physiological value for humans; its consumption is growing every year [1]. In 2020, the global meat consumption was 324 million metric tons, which is three times higher than 50 years ago [2]. In Russia, per capita consumption of meat was about 76–77 kg in 2021; a slight increase is possible in 2022 [3]. With a growth in meat consumption, its quality is becoming an increasingly important factor influencing a consumers' decision [4].

For meat quality assessment, two main approaches are used: subjective and objective. Subjective methods include sensory evaluation, which involves visual and eating experiences [5]. Their disadvantage resides in poor repeatability, dependence on taster's experience and difficulties in quantitative interpretation of results. Objective methods include various laboratory tests that evaluate physical and chemical properties of meat, including electrophoresis [7], enzyme-linked immunosorbent assay (ELISA) [8], mass-spectrometric methods [9], gas chromatography-mass spectrometry [10], high performance liquid chromatography (HPLC) [11,12] and methods based on the polymerase chain reaction (PCR) [13,14,15]. Although PCR and ELISA are the most specific and sensitive methods, they require expensive equipment and highly qualified specialists, which restricts their use. Chromatographic methods usually have low repeatability. These methods give accurate results, but a sample is damaged or destroyed, and the procedure, especially sample preparation, requires, as a rule, large amounts of time and resources. This hinders significantly their use for automated analysis directly in production [16].

Therefore, the development of rapid and non-destructive detection methods is necessary to ensure the population health, analysis of meat quality and safety.

Over the last decade, many complex studies associated with quantitative assessment of characteristics of carcasses and meat of slaughter animals were carried out using methods of imaging and spectroscopy [17,18], as well as tools for assessment and analysis of images and new algorithms for effective prediction of quality indicators in meat raw materials [19,20].

Much attention is given to the spectroscopic methods in visible and near-infrared range (VIS–NIRS and NIRS), hyperspectral imaging (HSI) and Raman spectroscopy. In this paper, we consider a possibility of using Raman spectroscopy for analysis of quality indicators of meat raw materials from different species of slaughter animals.

Raman spectroscopy is spectroscopy that allows identification of vibrational modes of molecules and is a non-destructive method of analysis. When photons collide with molecules, three different types of scattering occur: Rayleigh scattering, anti-Stokes Raman scattering and Stokes Raman scattering. Raman scattering is caused by the fact that photons give energy to molecules (Stokes scattering) or receive energy from molecules (anti-Stokes scattering) [21]. Due to this exchange of energy, shifts between the energetic levels in molecules are caused. Raman scattering (RS) spectra represent both structural and qualitative information about a substance [22,23].

Raman spectroscopic methods (RSMs) demonstrated a significant potential in analysis of various indicators in agricultural products such as milk, eggs, nuts, vegetable oils, fruit and vegetables, grain (Figure 1) [24].

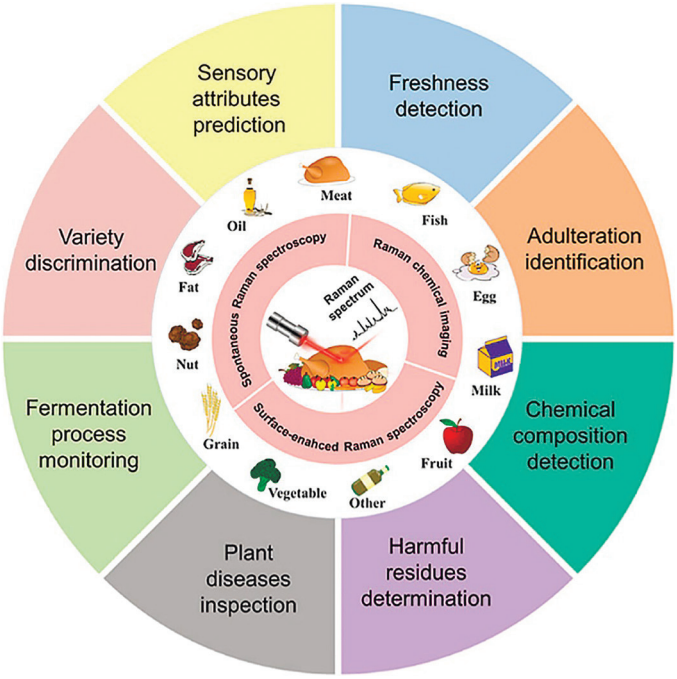

Figure 1. Application of Raman spectroscopy in analysis of agricultural products [24]

Vibrational spectroscopy attracts attention as an alternative to traditional methods for assessment of meat quality indicators [5,25,26,27]. Its advantages include minimal sample preparation, fingerprint spectrum (unique spectra of molecules of different substances), high sensitivity, rapid acquisition of data, non-destructive control, environmental friendliness.

Contrary to infrared spectroscopic methods, the Raman effect is observed in the scattered light from a sample and not in a spectrum of light absorption by a sample. With that, heavy molecules, such as water molecules, scatter Raman radiation worse, which makes Raman spectroscopy less sensitive to the moisture content both in a sample and in the environment, in which the measurement is performed. This fact is extremely important in analysis of food and, first of all, meat, dairy and fish products.

The paper presents a review of the potential of using Raman spectroscopy in tandem with approaches of chemometric modeling in analysis of meat raw materials and finished products.

Objects and methods

Design of the study:

The systematic review was carried out according to PRISMA (Preferred Reporting Items for Systematic Reviews and Meta-Analyses) [28].

The strategy for searching publications is presented in Figure 2.

Inclusion criteria were as follows:

1. Correspondence to the theme of the systematic review by one of three modalities: Raman spectroscopy, non-destructive analysis of meat products, use of Raman spectroscopy for meat quality analysis.
2. Original research published in a peer-reviewed journal.
3. Presentation of data about methods of statistical and/or chemometric analysis
4. Publication is dedicated to the application of Raman spectroscopy for analysis of meat raw materials and finished meat products including detection of structural changes in proteins, intramuscular fat, pH, drip losses during storage, detection of fatty acids, raw material falsification.

Exclusion criteria were as follows:

1. Studies that envisage the use of alternative types of spectroscopy for analysis of meat samples (for example, IR-spectroscopy, UV-visible spectroscopy).
2. Studies that envisage the use of RSMs to determine microelement composition in samples.
3. Studies that envisage the use of RSMs to study food products of the agro-industrial complex not relevant to meat raw materials.
4. Studies that envisage the use of RSMs to study inorganic materials.
5. Studies that envisage the use of RSMs to study cells and tissues (of animals and humans), as well as microorganisms.

A search of relevant scientific publications was carried out in Russian and foreign electronic databases (Web of Science, U. S. National Library of Medicine (pubmed.ncbi.nlm.nih.gov), Russian Scientific Electronic Library (elibrary.ru), Russian National Public Library for Science and Technology) in Russian and English for a period of 2007 to 2022. A special attention was paid to publications issued over the last five years.

During the last 15 years, 401 studies dedicated to various investigations of meat raw materials by the spectroscopic methods were published.

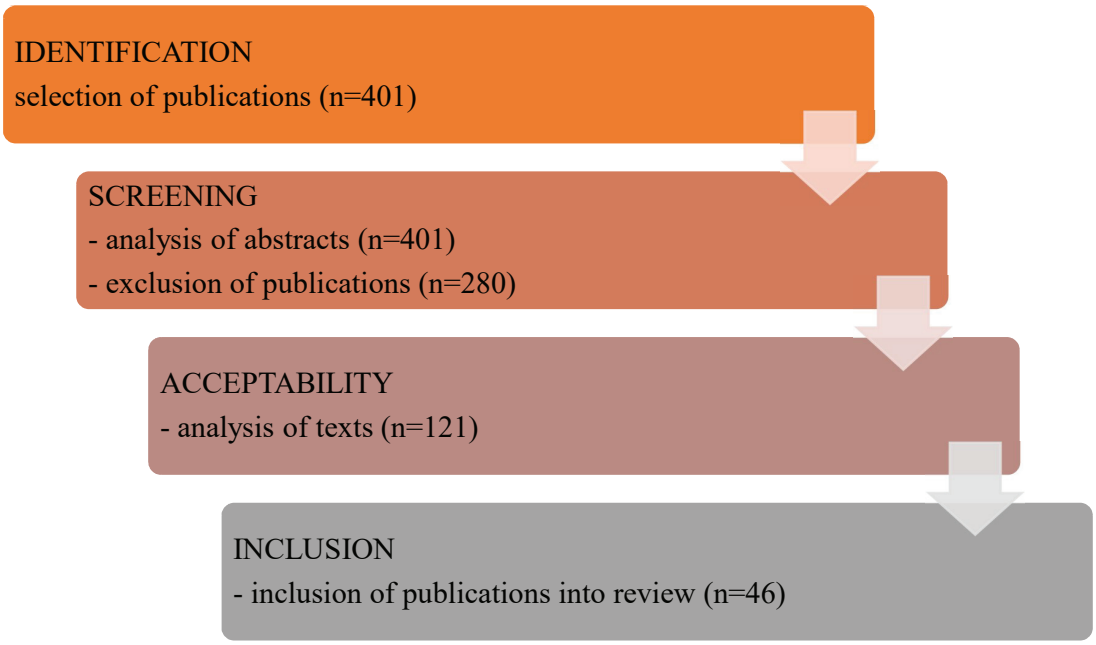

Figure 2. The strategy for selection and inclusion of publications into the systemic review

At the first stage, the titles of the papers obtained as a result of the search were analyzed. Part of publications was excluded as not corresponding to the inclusion criteria. Then, analysis of abstracts of the selected papers was carried out, on which basis the second exclusion was performed. At the next stage, the following information was taken from each publication included in the review: author(s), publication year, country; aim and methods of investigation; testing of the statistical hypothesis; description of the methodology of the experiment; the obtained results. Detailed analysis of each publication included into the review was conducted based on the specific elements of investigation questions and the aim of the review by double data extraction (two independent researchers worked on the review).

All obtained data were used for analysis and systematization of the results.

History of discovering Raman spectroscopy

Inelastic scattering of light was predicted by A. Smekal as far back as 1923. He assumed that light has the quantum structure and that, after scattering, monochromatic light would have both the original frequency and frequencies of higher and lower wavelengths [29]. However, in practice, inelastic scattering was not observed until 1928. The Raman effect is called in honor of one of its discoverers, the Indian scientist C. V. Raman, who together with K. S. Krishnan observed this effect in organic liquids in 1928 (Figure 3). In 1930, C. V. Raman was awarded a Nobel Prize in physics for this invention. Russian scientists G. Landsberg and L. Mandelstam observed the similar effect in inorganic crystals independently of them. Raman spectra in gases were observed for the first time by F. Rasetti in 1929 [30].

Between 1930 and 1934, the American physicist of the Czechoslovak origin G. Placzek developed theoretically the "effect of Raman scattering". The experiments were

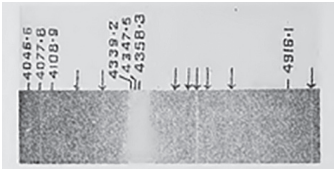

Figure 3. The first Raman spectrum of benzol published by C. V. Raman and K. S. Krishnan in 1928 [31]

carried out using the mercury arc as the main light source with photodetectors that were replaced with spectrophotometric detectors. During the years after its discovery, Raman spectroscopy was used for creation of the first catalog of molecular vibrational fingerprints. However, enormous effects were needed to obtain the Raman spectrum because of the essentially weak sensitivity of the method. Thus, the use of Raman spectroscopy decreased, in particular, after the development of commercial IR spectrophotometers in the 1940s. Raman spectroscopy again attracted the attention in 1960, when laser appeared. This source of monochromatic light simplified the detection tool and increased the sensitivity of the method. The use of laser as a source of monochromatic light stimulated the development of Raman spectroscopy as a valuable analytical method [32].

Principle of Raman spectroscopy

The principle of the Raman effect is based on the inelastic process of light scattering between the incident light and a substance under irradiation. During the interaction between light and a sample, the incident light interacts with the molecules and distorts the electron cloud forming a "virtual level". The "virtual level" is instable; thus, photons are scattered immediately to a relatively stable state. When photons return to the initial level of energy (ground level), there is no energy transfer between the incident light and scattered light, and photon frequency and wavelength are not changed (Figure 4). This elastic collision process is referred to as Rayleigh scattering. On the other hand, when

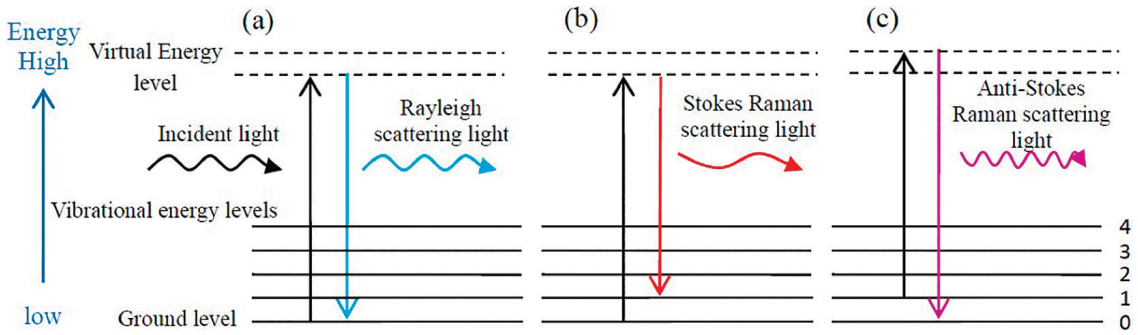

Figure 4. Diagram of the Rayleigh and Raman scattering processes:
(a) Rayleigh scattering, (b) Stokes Raman scattering and (c) anti-Stokes Raman scattering [33]

photons move to a new energy level that is different from the initial one, the energy transfer occurs (that is, a photon loses or acquires a certain amount of energy), which leads to a downward or upward shift in the energy of laser photons. This provides information about a substance under investigation [33].

Raman scattering can be divided into two types: Stokes Raman scattering and anti-Stokes Raman scattering. In Stokes Raman scattering, photons are excited from the initial energy level and move to a higher energy level. As a result, the scattered light has a lower frequency than the incident light. In anti-Stokes Raman scattering, photons are excited from the initial energy level and move to a lower energy level. In this case, the scattered light has a higher frequency compared to the incident light [33].

Transitions that have large Raman intensities often have weak IR intensities and vice versa. When a bond is strongly polarized, a small alteration in its length, which occurs, for instance, during vibration, will have only a small effect on polarization. Thus, vibrations that are associated with polar bonds (for example, C-O, N-O, O-H) are relatively weak Raman scatterers. However, such polarized bonds carry their electrical charges during the vibrational motion (unless neutralized by symmetry factors), which leads to a larger alteration in the net dipole moment during vibration, generating a strong IR absorption band. On the contrary, comparatively neutral bonds (for example, C–C, C-H, C=C) undergo large changes in polarizability during vibration. Nevertheless, there is no similar effect on the dipole moment; therefore, vibrations that involve mainly this type of bonds are strong Raman scatterers, but they are weak in the IR range [34].

Raman shifts are usually expressed in wavenumbers that have the inverse length since this value is directly related to energy. For conversion of a spectral wavelength into wavenumbers of a shift in the Raman spectrum, the following equation can be used:

$$\Delta\tilde{\nu} = \left(\frac{1}{\lambda_0} - \frac{1}{\lambda_1}\right), \qquad (1)$$

where

$\Delta\tilde{\nu}$ is the Raman shift expressed in a wavenumber;

λ_0 is the excitation wavelength;

λ_1 is the Raman spectrum wavelength.

An inverse centimeter (cm^{-1}) is the most frequently used measurement unit for expression of a wavenumber in Raman spectra. As a wavelength is often expressed nanometers (nm), the equation given above can be scaled for this conversion of units:

$$\Delta\tilde{\nu}(cm^{-1}) = \left(\frac{1}{\lambda_0(nm)} - \frac{1}{\lambda_1(nm)}\right) \times \frac{(10^7\,nm)}{(nm)}. \qquad (2)$$

Modern Raman spectroscopy almost always envisages the use of lasers as a source of light excitation. As lasers became available only more than three decades after the discovery of the effect, C. V. Raman and K. S. Krishnan used a mercury lamp and photographic plates for spectra recording [31]. It required hours or even weeks to obtain early spectra because of weak light sources, low sensitivity of detectors, as well as weak Raman scattering cross sections of most materials. To choose certain regions of wavelengths for excitation and detection, different color filters and chemical solutions were used. Nevertheless, a wide central line that corresponded to Rayleigh scattering of the excitation source still dominated in the photographic spectra [35].

Usually, Raman scattering is very weak, which was a problem for spectra collection for a long time. There was a need for methods that could separate weak inelastic scattering from intensive Rayleigh scattering. To this end, holographic gratings and dispersion came into use. Initially, photomultipliers were used as detectors; however, this method of collection was time consuming [36].

Technological achievements appeared in the 1980s made Raman spectroscopy much more sensitive. This was facilitated by the development and invention of modern radiation detectors such as charge-coupled devices — CCD detectors. The development of the method was also strongly affected by the appearance of reliable, stable, low-cost lasers [37].

The principle scheme of the modern Raman spectroscope is presented in Figure 5.

Modifications of Raman spectroscopy

The term Raman spectroscopy usually refers to vibrational Raman radiation with the use of laser wavelengths that are not absorbed by a sample. No less than 25 modifications of Raman spectroscopy were developed including surface-enhanced Raman spectroscopy, resonance Raman

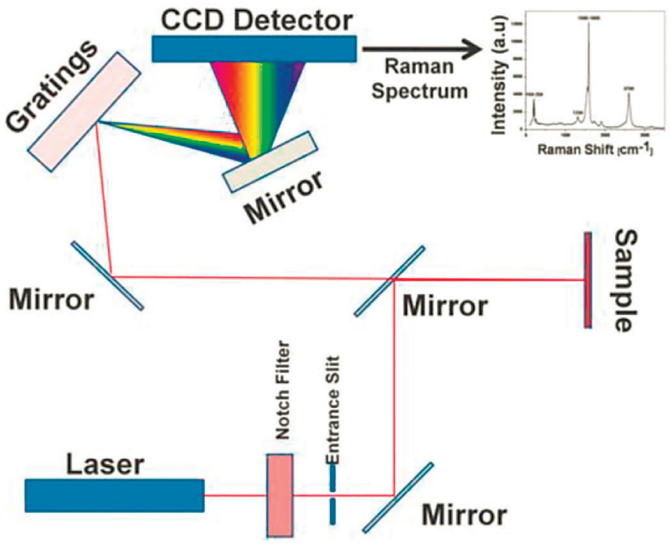

Figure 5. Principle scheme of the modern Raman spectroscope [36]

spectroscopy, polarized Raman spectroscopy, stimulated Raman spectroscopy, transmission Raman spectroscopy, spatially offset Raman spectroscopy and hyper-Raman spectroscopy [30,36]. As a rule, the aim is an increase in sensitivity (for example, surface-enhanced Raman spectroscopy), improvement of spatial resolution (Raman microscopy) or acquisition of a very specific information (resonance Raman scattering).

Spontaneous (or far-field) Raman spectroscopy

Spontaneous Raman spectroscopy or normal Raman spectroscopy includes Raman spectroscopic methods based on Raman scattering with the use of normal far-field optics. There are several variants of normal Raman spectroscopy regarding the excitation-detection geometry, combination with other methods, application of specific (polarization) optics and specific selection of excitation wavelengths to enhance resonance:

- Correlative Raman imaging [38]
- Resonance Raman spectroscopy (RRS) [39]
- Angle-resolved Raman spectroscopy [40]
- Optical tweezers Raman spectroscopy (OTRS) [41]
- Spatially offset Raman spectroscopy (SORS) [42]
- Raman optical activity (ROA) [43]
- Transmission Raman spectroscopy [44]
- Micro-cavity substrates [45]
- Remote Raman spectroscopy [46]
- X-ray Raman scattering [47]

Enhanced (or near-field) Raman spectroscopy

In enhanced Raman spectroscopy, the enhancement of Raman scattering is attained due to the enhancement of the local electric field by the optical near-field effect (for instance, localized surface plasmons). The examples include:

- Surface-enhanced Raman spectroscopy (SERS) [48, 49]
- Surface enhanced resonance Raman scattering (SERRS) [50]
- Tip-enhanced Raman spectroscopy (TERS) [51,52]
- Surface plasmon polariton enhanced Raman scattering (SPPERS) [53]

Non-linear Raman spectroscopy

In non-linear Raman spectroscopy, the enhancement of the Raman signal is attained due to the non-linear optical effects achieved, as a rule, by mixing two or more wavelengths that are emitted by spatially and temporally synchronized pulsed lasers. The examples include:

- Hyper Raman spectroscopy [54]
- Stimulated Raman spectroscopy (SRS) [55]
- Inverse Raman spectroscopy [56]
- Coherent anti-Stokes Raman spectroscopy (CARS) [57]

There is also morphologically directed Raman spectroscopy (MDRS), which combines the methods of automated particle imaging and Raman microspectroscopy into an integrated platform that allows detecting the chemical and morphological characteristics of individual components in a multi-component sample [58].

The main Raman spectroscopic methods used in analysis of biological objects are presented in Table 1 [21].

Table 1. Raman spectroscopic methods used in analysis of biological objects [21]

Method	Characteristics	Advantages	Drawbacks	Application
Coherent anti-Stokes Raman spectroscopy (CARS)	Non-linear approach using multiple laser frequencies; generated strong anti-Stokes signal reveals vibrational coherence	Increased signal (10^3–10^6); high sensitivity; 3D imaging	Non-resonant background can dominate over weak resonant signals	Imaging of cells and tissues; diagnosis of cancer
Confocal Raman microspectroscopy	Adding a confocal microscope allows tissue depth measurement. A pinhole is used in the spectrometer for stray light rejection	High sensitivity; high lateral and depth resolution; 3D imaging	Diffraction-limited resolution	Imaging of cells and tissues; diagnosis of cancer
Drop coating deposition Raman spectroscopy (DCDRS)	Small volume of a liquid sample is dropped onto the flat substrate and dried	Small volumes (2–10 µl) of liquids are needed	Not fully free from the "coffee ring" effect	Analysis of biofluids; quantification of protein
FT-Raman spectroscopy	System that uses Fourier transformation and the Michelson interferometer	High throughput; high resolution; fluorescence-free	Low scattering intensity; limited to IR measurements; detector noise limited	Plant materials

Table 1. End

Method	Characteristics	Advantages	Drawbacks	Application
Kerr-gated Raman spectroscopy	Linear method that uses the repeated laser pulses and the Kerr gate (capture Raman light temporally — up to 3 picoseconds)	Depth measurement up to several millimeters; fluorescence rejection; high sensitivity	Not fully fluorescence-free; better performance in combination with shifted excitation	Depth profiling of human tissue
Polarized Raman spectroscopy (PRS)	Polarized light with the specific electric field vector obtains spectral information only from specific vibrational modes according to their orientation in reference to the incident beam	Information about the molecular structure and orientation	Inapplicable to the majority of samples; loss of the spectral information; time consuming	Orientation of collagen structures; plant photosystems
Raman Optical Activity (ROA)	Use of right- and left-circularly polarized incident light, which allows detecting the optical activity of discrete molecular vibrations.	Structural information from specific conformations of chiral molecules	Circular intensity differences are very small; vibrational coupling in signals can hamper accurate band assignment	Analysis of biopolymers
Resonance Raman spectroscopy (RRS)	Uses the "resonance effect" when the laser frequency coincides with (or is close to) the frequency of the electronic transition of a sample or compound under investigation	An increase in a signal of up to 6 orders of magnitude	Susceptible to fluorescence interference	Photosystems of plants; Analysis of human tissues
Shifted excitation Raman difference spectroscopy (SERDS)	Non-linear approach, in which two spectra at slightly different laser frequencies are obtained and a difference spectrum is created by subtracting the two; hence, eliminating background fluorescence	Fluorescence rejection; increased sensitivity	Difference spectra are reconstructed with the use of peak fitting; error-prone	Living cells and tissues of animals and humans
Spatially offset Raman spectroscopy (SORS)	For illumination of the the sample surface, continuous low intensity laser beams are used. Spectra are then derived at different distances from the surface. A scaled subtraction between these spectra shows changes indicative of the underlying layers	Depth measurements up to several millimeters	Comparatively weak signal	Diagnosis of cancer Chemical analysis upon physical impacts
Surface enhanced spatially offset Raman spectroscopy (SESORS)	SERS and SORS approaches are combined, enabling detection of SERS nanoparticles added to turbid samples	Detection of the SERS signals up to 50 mm below the sample surface	Addition of nanoparticles is needed	Depth measurements of samples
Stimulated Raman scattering (SRS)	Non-linear approach with the use of wave pumping and scattered Stokes radiation, which are tuned to a specific frequency representative of molecular vibrations. The transmitted intensity is proportional to the biochemical components	Not susceptible to the effect of fluorescence and the nonresonant background; high sensitivity (1 in 10^6 photons); high spatial resolution	Proneness to interference from strong Raman scatterers; restricted to measuring one Raman peak per acquisition	Imaging of cells and tissues
Surface enhanced Raman scattering (SERS)	Surface plasmon resonance of the metal surface with nanoscale roughness is used, which significantly increases the electric field upon excitation by a laser. Upon adsorption on a biomolecule, nanoparticles lead to a significant enhancement of Raman scattering	Enhanced signal (10^3–10^{10}); resolution is lower than the diffraction limit; fluorescence quenching; low limit of detection; molecular labeling	Lack of reproducibility; band intensity of high frequency modes can be reduced; molecular selectivity to nanoparticle adherence	Detection of single molecules; analysis of living cells; diagnosis of cancer; identification of bacteria; plant materials
Surface enhanced resonance Raman scattering (SERRS)	RRS and SERS approaches are combined with the use of the laser frequency in resonance with a biomolecule in question and introduction of the SERS active substrate	Enhancement up to 10^{15}; incremental benefits of both SERS and RRS	Increased complexity of the experiment	Detection of biomolecules Analysis of protein
Tip enhanced Raman spectroscopy (TERS)	Tip of the atomic force microscope, which is coated with SERS active metal, is used. Upon placement in close proximity to a sample, it leads to enhanced scattering	Tip-dependent spatial resolution; low limit of detection; fluorescence quenching; resolution is lower than the diffraction limit	Increased complexity of the experiment; sample is heated at the tip apex	Microbiology; biochemical imaging
Total internal reflection Raman spectroscopy	Sample is placed in contact with a reflective prism, through which a laser beam is reflected, generating an evanescent wave that penetrates the sample below	Specified penetration depth	Surface sensitivity is reduced	Plant materials
Transmission Raman	Raman scattered light is collected on the opposite side of laser illumination	Depth measurements up to 30 mm; appropriate for non-transparent materials	Interference from surface molecules	Diagnosis of cancer

Application of Raman spectroscopy in the meat industry

Recently, Raman spectroscopy has received much attention. Many authors confirm that Raman spectroscopy is of great interest in assessment of meat composition and quality [59,60,61]. However, it has to be taken into account that Raman spectra have many dependent variables; therefore, it is necessary to use methods of multivariate analysis. The most often used method of multivariate analysis for this technique is partial least-squares regression (PLSR) analysis [62]. Several authors additionally use chemometrics to extract representative information from Raman spectra of meat and analyze the relation between the molecular structure and different radical groups to determine and assess meat quality (Figure 6) [63,64].

Data of Raman spectroscopy correlate with results obtained using the traditional control methods (water binding capacity, detection of texture, content of dimethylamine, peroxide value and fatty acid composition) and can be used for meat quality assessment. Raman spectroscopic methods (RSMs) give structural information about changes in meat proteins and lipids occurring during storage [65].

Raman spectroscopy is an effective and non-invasive method for studying alterations in the protein secondary structure, analysis of amide I (1650–1680 cm^{-1}) and amide III (1200–1350 cm^{-1}) regions, C–C groups (940 cm^{-1}) and modifications of local muscle proteins (tryptophan residues, bands of aliphatic amino acids) [22]. Herrero [22] used Raman spectroscopy to reveal structural changes in isolated myofibrillar and connective tissue proteins due to the addition of various compounds and an effect of freezing and storage in the frozen state. It was found that RSMs are a tool for in situ monitoring of protein structural changes in meat during storage in the frozen state and prediction of functional and organoleptic properties of raw materials [22].

Chemical substances, such as glycogen, glucose, lactate and cortisol, are predictors of meat quality; however, their detection on the meat surface by conventional Raman spectroscopy is restricted due to a low concentration. Ostovar Pour et al. [66] used spatially offset Raman spectroscopy (SORS) to detect spectral bands of glycogen, lactate, glucose and cortisol in beef muscle tissue (5 mm below the surface). The chemometric analysis performed by the authors revealed clearly the separation of peaks of metabolites into four groups under investigation [66].

Later on, Ostovar Pour et al. [67] studied the potential of spatially offset Raman spectroscopy (SORS) in discrimination between beef cuts (rump, Scotch fillet, round, chuck, tenderloin, and T-bone). The obtained results showed differences in the structure-sensitive bands from the amide I and III regions, cysteine, glutamic acid, and phenylalanine [67].

Cama-Moncunill et al. [68] investigated the potential of RSMs with subsequent chemometrics to predict Warner-Bratzler shear force (WBSF), intramuscular fat (IMF), pH, drip losses and cooking losses. Regression models PLS were developed based on the spectra recorded in thawed *longissimus thoracis et lumborum* muscle frozen 2 days after slaughter. Except pH, models demonstrated pronounced performance in calibration (coefficient of determination R^2 was in a range of 0.5 to 0.9) and promising predictive capability: WBSF (root-mean square error of prediction (RMSEP) was in a range of 4.6 to 9 N,) IMF (RMSEP from 0.9 to 1.1%), drip losses (RMSEP from 1 to 1.3%) and cooking losses (RMSEP from 1.5 to 2.9%).

Yang et al. [69] studied pH, meat color and microbial counts in beef steaks stored at 4 °C for 21 days using two different packaging methods: vacuum packaging (VP) and modified atmosphere packaging (MAP). The PLSR models demonstrated that Raman spectroscopy was able to predict total viable counts (TVC) and lactic acid bacteria (LAB) counts that were measured 21 days after slaughter (TVC in VP: $R^2_{cv} = 0.99$, RMSEP = 0.61; TVC in MAP: $R^2_{cv} = 0.90$, RMSEP = 0.38; LAB in VP: $R^2_{cv} = 0.99$, RMSEP = 0.54; LAB in MAP: $R^2_{cv} = 0.75$, RMSEP = 0.60). The obtained results showed a possibility of using Raman spectroscopy to rapidly detect meat spoilage.

Combination of Raman spectroscopy with the chemometric method for quantification of myoglobin proportions (deoxymyoglobin and oxymyoglobin) is presented in [70]. The optimal results were obtained with the prediction model "random frog-partial least squares (projection into latent structure)" (RF-PLS) for both

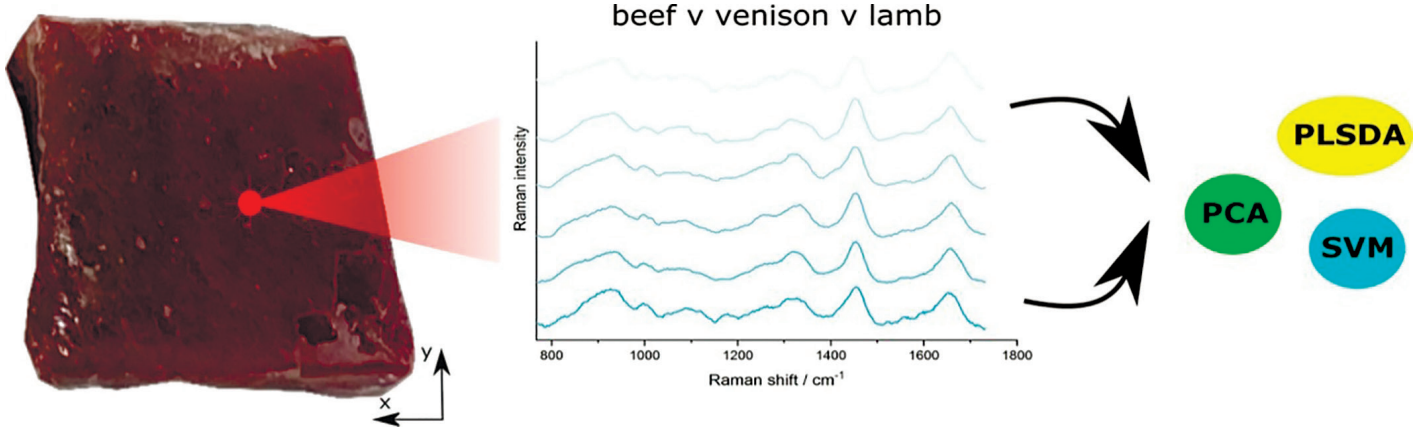

Figure 6. Scheme of Raman spectroscopy and analysis of the obtained results [64]

deoxymyoglobin (Rp = 0.8936; RMSEP = 2.91) and oxymyoglobin (Rp = 0.9762; RMSEP = 1.23).

Boyacı et al. [71] used the Raman spectroscopic method coupled with chemometrics to detect beef falsification with horsemeat (n = 49). Processing of data from the collected Raman spectra was carried out using principal component analysis (PCA). All meat samples were correctly classified by their origin. In addition, different concentrations (25%, 50%, 75%, w/w) of horsemeat in the beef samples were also determined using the created model system.

The results of the studies demonstrate that Raman spectroscopy in combination with the chemometric method of data processing can be used to determine an origin of meat from different species of slaughter animals over a very short time of analysis (30s) without a need for sophisticated chromatography, immunological or genetic methods of analysis [72,73].

The potential of Raman spectroscopy combined with three chemometric methods for differentiation of red meat samples (beef, lamb and venison; n = 90) is shown in [64] (Figure 7). PLSDA (partial least squares discriminant analysis) and SVM (support vector machines) classifications were used for creation of classification models, while PCA was used for the exploratory research (Figure 8). The results obtained with the linear and non-linear kernel SVM models demonstrated sensitivity of more than 87% and 90%, respectively. The PLSDA model showed accuracy of 92% and 81% in determining lamb and 88% and 79% in determining beef for both the training and test sets, respectively.

Zhao et al. [74] used RSMs to predict organoleptic properties of beef samples (n = 72) (Figure 9). The best results of prediction were achieved when a Raman frequency range of 1300–2800 cm^{-1} was used. The prediction performance of the PLSR models was moderate to high for all organoleptic indicators (R_{CV}^2 = 0.50–0.84; RMSECV = 1.31–9.07) and especially high for flavor characteristics (R_{CVS}^2 = 0.80–0.84, RMSECVs = 4.21–4.65).

Raman spectroscopy is widely used in studying quality of meat from different species of slaughter animals, as well as chicken, including analysis of raw fat characteristics [75,76], detection of boar taint in pork [77], determination of organoleptic properties of raw materials [78], pH [79], spoilage [80] and identification of meat from different animal species [72,81] (Table 2).

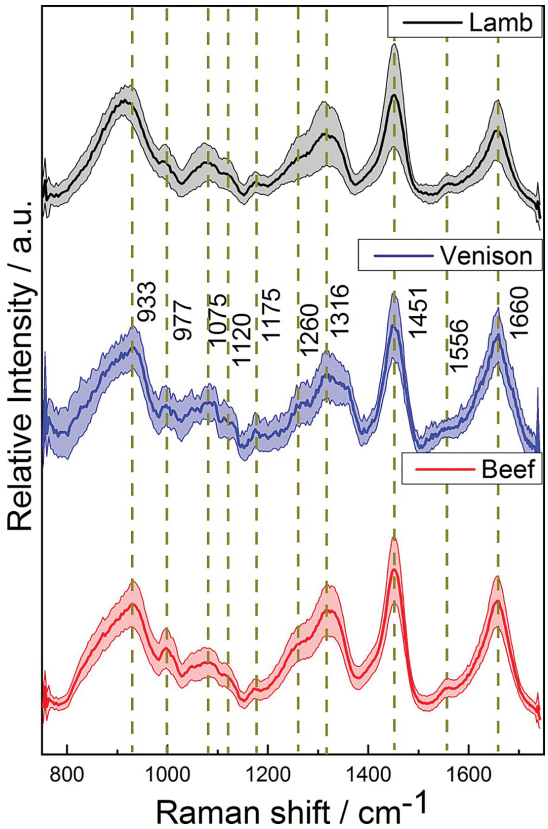

Figure 7. Mean Raman spectra of beef, venison and lamb [64]

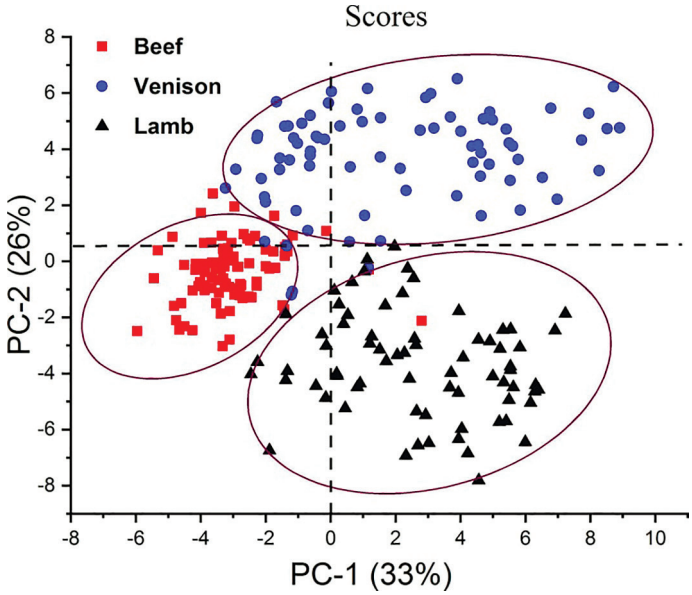

Figure 8. Separation of beef, venison and lamb samples using PCA [64]

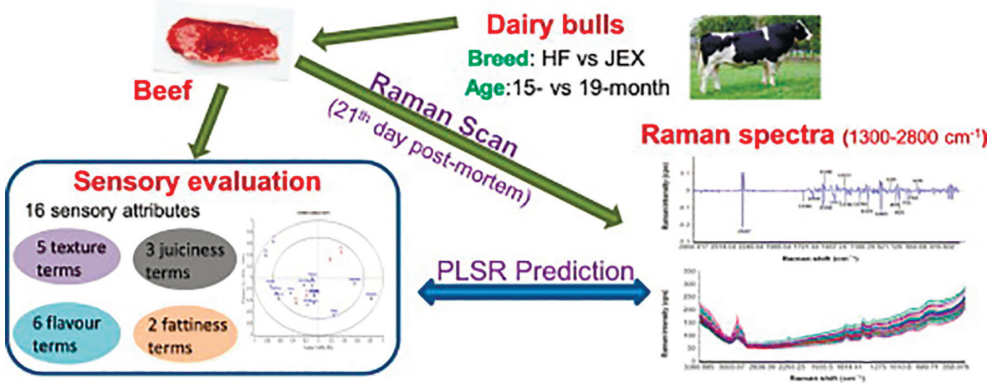

Figure 9. Scheme of research using RSMs and PLSR [74]

Table 2. Use of Raman spectroscopy for meat raw material analysis

Sample	Indicator	Algorithm of data analysis	Results	Source
Chicken	Protein structure	one-way ANOVA*	Upon addition of sodium bicarbonate, an increase in hydrophobic interactions as a result of protein unfolding and exposure of aliphatic residues was established. It was concluded that sodium bicarbonate can be used for reduction of the sodium chloride content.	[82]
Pork backfat	Fatty acids	PLS*	Correlation of spectra with parameters of the total fatty acid composition and most of the individual fatty acids (R_{CV}^2 = 0.78–0.90)*	[83]
Pork after heat treatment	Temperature control during heat treatment	PLS-DA* PCA	Detection of processing temperature of below or above 65 °C (accuracy of 97.87% and 97.62%, respectively)	[84]
Pork	pH	PLS-DA*, PCA	It is possible to predict pH values by spectra, (RMSECV = 0.13 for pH after 45 min. and RMSECV = 0.21 for pH after 24 hours)	[79]
Pork (longissimus lumborum)	Drip losses and pH	PLSR*	It is possible to use Raman spectroscopy for rough screening of drip losses and pH (R_{CV}^2 = 0.75 for drip losses and R_{CV}^2 = 0.72 for pH)*	[85]
Beef	Tenderness	PLSR*	Tough and tender samples can be identified with the accuracy of 70–88%	[25]
Beef	Falsification	PLSDA*	Detection of falsification with the efficiency rate of 86.6% and 79.8% for the training and test sets, respectively	[86]
Beef	Texture	PCA*, PLSR*	Prediction of tenderness, chewiness and firmness with R^2 = 0.81, 0.80 and 0.81*, respectively	[87]
Beef	Organoleptic characteristics	PLSR	R^2 = 0.63–0.89* for the same breed and 0.52–0.89 for the same age	[74]
Beef	Saturated fatty acids	PCA*	Differences between Australian grass-fed and grain-fed beef by average spectra of carcasses indicating different fatty acid content	[88]
Beef	Physico-chemical indicators	PLS-DA*, PLSR	All samples were correctly classified using PLS-DA*; with that, correct identification was achieved for 86.7% of samples from different muscles. The PLSR* models that used Raman spectra of the 3rd day after slaughter had better prediction performance compared to the models that used Raman spectra of the 7th and 14th days	[89]
Beef	Organoleptic indicators (juiciness and tenderness)	PLSR*	Correlation between predicted and observed values of juiciness and tenderness of 0.42 and 0.47, respectively	[90]
Beef, venison and lamb	Identification of meat from different animal species	PCA*, PLS-DA*, SVM*	Models providing accuracy of more than 80% (PLSDA*) and 92% (SVM*) for identification of unknown meat samples (test set)	[64]
Beef tallow, pork lard, chicken fat, duck oil	Fatty acid analysis: unsaturated fatty acids and total fatty acids	Linear correlation	Fat classification using Raman peak ratio. An indicator "oil gauge (OG)" was proposed as a standard trait for fat classification	[91]
Lamb (*m. Longissimus lumborum*)	Intramuscular fat content and major fatty acid groups	PLSR* and linear regression	Prediction of PUFA (R^2 = 0.93)* and MUFA (R^2 = 0.54)*, as well as SFA levels adjusted with regard to the IMF content (R^2 = 0.54)*	[92]
Lamb	Technological properties (Warner-Bratzler shear force, cooking losses)	PLSR*	For shear force R^2 = 0.79* and R^2 = 0.86*, for cooking losses R^2 = 0.79* and R^2 = 0.83* for two models between observed and predicted values	[93]
Lamb (*m. Longissimus lumborum*)	Technological properties	PLSR*	R_{CV}^2 = 0.06* between observed and cross validated predicted values	[26]
Lamb (*m. Semimembranosus*)	Technological properties	PLSR*	R_{CV}^2 = 0.27* between observed and cross validated predicted values	[94]
Lamb (*m. Semimembranosus*)	Indicators of meat freshness during storage and after freezing/thawing	PLSR*	R_{CV}^2 from 0.33 to 0.59 for various indicators between observed and predicted values. Possibility to identify carcasses with deviations during autolysis	[95]

* PLS — partial least squares (projection into latent structure); PLSR — partial least squares regression (PLS-regression); ANOVA — analysis of variance; PLS-DA — partial least squares discriminant analysis; PCA — principal component analysis; SVM — support vector machines; R^2 — coefficient of determination; R_{CV}^2 — coefficient of determination in cross-validation

Andersen et al. [96] compared the results of Raman, near infrared (NIR) and fluorescence spectroscopy for analysis of pH and porcine intramuscular fat (*m. Longissimus lumborum*) (n = 112) 4–5 days after slaughter. The results of Raman spectroscopy showed R_{CV}^2 in a range of 0.49 to 0.73 for all examined indicators (upon PLSR). Near infrared and fluorescence spectroscopy demonstrated limited possibilities for quality analysis (R_{CV}^2 was in a range from 0.06 to 0.57 and from 0.04 to 0.18, respectively).

Later on, Andersen et al. [85] carried out research on prediction of drip losses using RSMs. Their results revealed that Raman spectroscopy can be used for rough screening of drip losses and pH in pork and that a sampling site is important for successful predictions. PLSR models were created using spectra from each of two samples of *m. Longissimus lumborum* (ventral and dorsal) individually or averaged spectra from both samples. The best results were observed for the models that used the sample from the ventral part of the muscle: $R_{CV}^2 = 0.75$, RMSECV = 1.27%, the ratio of prediction to deviation (RPD) = 2.0 for drip losses measured by the method EZ-DripLoss, and $R_{CV}^2 = 0.72$, RMSECV = 0.05 and RPD = 2.0 for pH.

Using a Raman spectrometer, samples of dry-cured ham from Iberian pigs were analyzed (n = 110). Four commercial categories were used in the study: pure-bred Iberian acorn-fed pigs, crossbred Iberian acorn-fed pigs, crossbred free-range feed-fed Iberian pigs and crossbred Iberian feed-fed pigs [97]. The results presented by the authors demonstrate that RSMs can be used as a rapid screening tool for quality verification of commercial dry-cured Iberian ham. LDA (linear discriminant analysis) chemometric models obtained using a Raman signal enabled classifying pigs according to the breed and feeding regime.

Tomasevic et al. [27] studied a possibility of using Raman spectroscopy for species identification of beef and pork in frankfurters. To this end, five different sausage recipes that included beef and pork were investigated. Linear discrimination analysis in combination with PCA and PLS was used for data analysis. The results showed high sensitivity of the models for beef sausages: 91.67% and 100%. The authors concluded that Raman spectroscopy can be used as a non-invasive method for rapid authentication of frankfurters.

Beattie et al. [98] used a combination of the Raman method with the method of multivariate analysis and neural networks achieving accuracy of 96.7% (with PCLDA) to 99.6% (with PLSDA) in classification of chicken, beef, lamb and pork fat. This possibility of identification was confirmed by others scientists worked with RSMs and used principal component analysis for successful classification of fat samples [81] (Figure 10 and Figure 11). The authors proposed RSMs as a useful tool for detecting falsifications in the meat industry, which will facilitate alleviation of consumers' concerns about meat they eat [98].

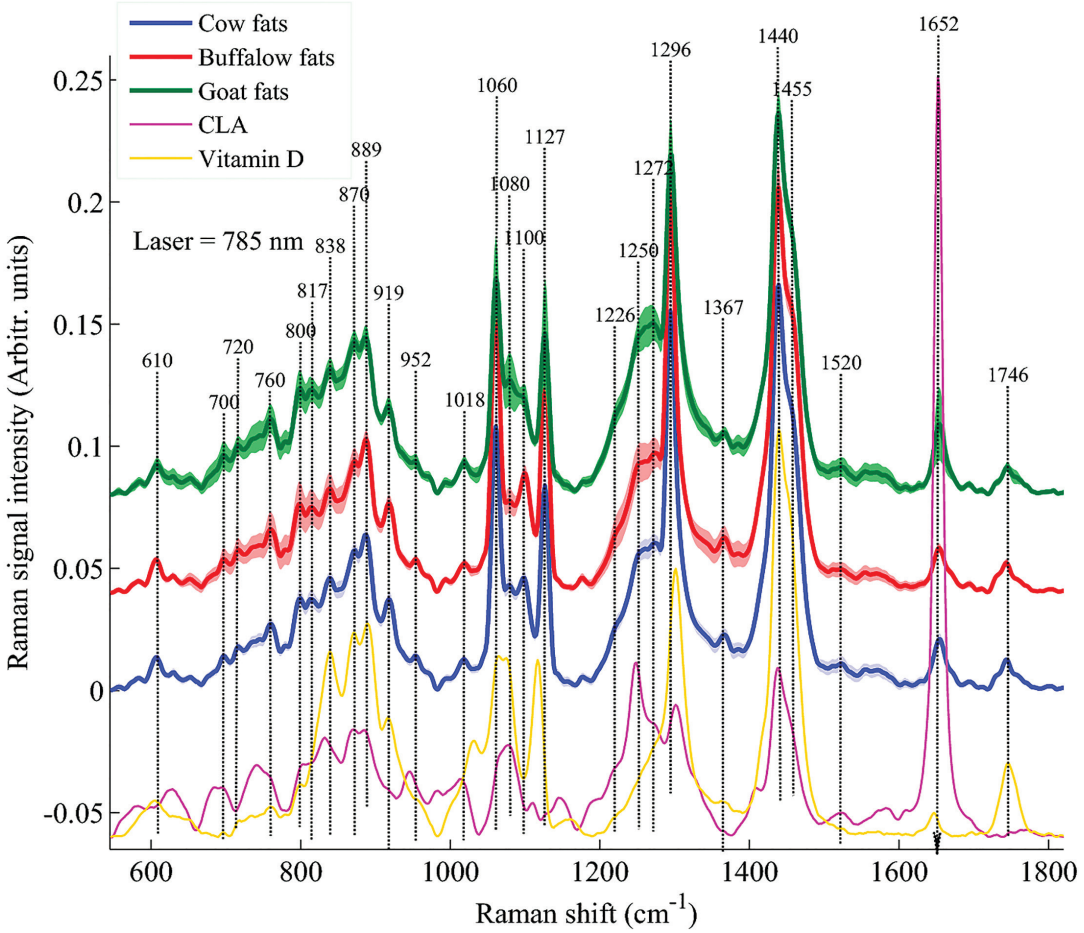

Figure 10. Raman spectra of 13 samples of beef fat, 18 samples of buffalo fat and 16 samples of goat fat, vitamin D and CLA (conjugated linoleic acid) obtained using laser with a wavelength of 785 nm [81]

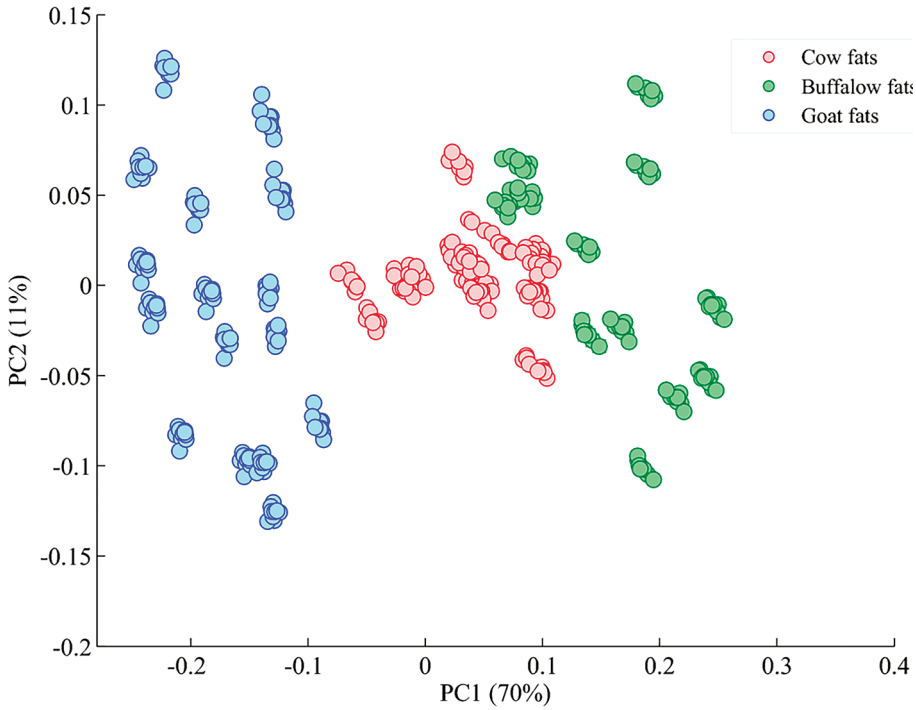

Figure 11. PCA analysis of Raman spectra of goat, beef and buffalo fat samples obtained using laser with a wavelength of 785 nm [81]

The prediction results for several technological characteristics of lamb quality such as Warner-Bratzler shear force (WBSF), color, cooking losses and pH using the method of Raman scattering are presented in [26,93,94,95]. The most interesting results were obtained when studying shear force. Prediction of this indicator in the samples of *m. Longissimus thoracis et lumborum* showed low accuracy (R_{CV}^2 = 0.06 between observed and cross validated predicted values) in [26], in contrast to the study by Schmidt H. et al. [93], where coefficients of determination of 0,79 and 0.86 were obtained for this characteristic (measurement in two muscle sites after freezing and thawing). When predicting shear force in *m. Semimembranosus*, a reduction in root mean square error by 12.9% and 7.6% was observed during aging for one and five days, correspondingly [94]. Raman spectroscopy was not able to predict this indicator in analysis of the *m. Semimembranosus* samples after freezing and thawing [95].

Traditional methods for determination of fat content and fatty acid composition are based, as a rule, on methods of extraction with a solvent and gas chromatography, and require the use of dangerous chemical solvents and thorough sample preparation, are expensive, labor intensive and result in irreversible damage to a sample. These drawbacks make them unsuitable for using in production conditions of meat plants [99]. Raman spectroscopy showed good results in measuring concentrations of the main fatty acid groups, such as PUFA, MUFA and SFA, as well as intramuscular fat [92]. The use of Raman scattering to assess the fatty acid content has a significant practical advantage as it does not require extraction and purification processes.

Lee, J.Y. et al. [91] classified four animal fats (beef, pork, chicken and duck fat) using Raman spectroscopy in combination with simple calculation of the intensity ratios of the Raman signals at the vibrational modes that corresponded to unsaturated fatty acids and total fatty acids.

When developing spectroscopic equipment for assessment of meat quality and composition, a special attention is being given to RSMs, as these methods do not require long and labor intensive sample preparation, are rapid and easy to perform (analysis can be done within several seconds). A trend towards promotion of the real-time automated control and quality control directly in production is seen worldwide [63]. At present, portable Raman spectroscopes with a robust water-proof casing for sensor protection have been developed for the use in the meat industry [100].

Bauer et al. [25] used a portable Raman system with a wavelength of 671 nm to evaluate tenderness of beef (n = 175) aged at minus 1°C and 7°C for 14 days. The correlation between Raman spectra and Warner-Bratzler shear force with the use of PLS gave cross-validated predictions of WBSF for both storage temperatures with the coefficients of determination Rcv^2 = 0.33–0.79. It was found that tough and tender samples could be distinguished with accuracy of 70–88%.

Fowler, S.M. et al. [90] studied sensory characteristics (juiciness and tenderness) of beef loins (*m. Longissimus lumborum*) (n = 45) using a portable Raman spectroscope with a wavelength of 671 nm before and after freezing. It was established that the spectroscopic device could determine juiciness and tenderness with the correlation between the predicted and observed values (ρ) of 0.42 и 0.47, respectively. The main changes were observed in fatty acid concentrations, protein hydrophobicity and collagen orientation.

Conclusion

Comprehensive evaluation of meat raw materials by organoleptic characteristics, internal constituents and external factors, as well as the application of the developed high performance quality control systems to real meat processing lines in production are still topical. Researches focus their attention primarily on methods of non-destructive quality control.

The unique analytical possibilities of Raman spectroscopy are demonstrated. The collected data presented in this review show that the use of Raman spectroscopy makes it possible to predict quality indicators of meat raw materials with a high degree of certainty and obtain a large volume of information from an object without its destruction.

The main principles of Raman spectroscopy, used equipment and tools for analysis of obtained spectra are described. RSMs have been successfully used to determine the meat chemical composition, including the content of moisture, fat, fatty acids and protein, pH, indicators of freshness, organoleptic and technological indicators, as well as to reveal raw material falsification.

Raman spectroscopy is an alternative method for rapid identification of the meat chemical composition. Contrary to the traditional methods, it does not require the complex sample preparation, use of chemical reagents or highly qualified personnel. The use of portable spectroscopes allows doing research directly on a technological line.

On the other hand, Raman spectroscopy, like other new technologies, requires further research:

Nowadays, there is no unified database of meat raw material spectra, as well as protocols with optimal conditions (laser wavelength, power, exposure time) of spectra recording; therefore, time is necessary for analysis of obtained results.

Obtained spectra often contain a significant amount of excessive data, which can slow down the speed of their real-time processing.

Developed predictive models are mainly based on a single indicator and, therefore, their use for multivariable prediction is hampered.

The cost of the equipment is very high and it is necessary to develop inexpensive spectroscopes for routine investigations.

Therefore, RSMs can replace several traditional methods for analysis of physico-chemical, biochemical and technological indicators of quality of meat raw materials and products, and a huge work lies ahead for their wide application.

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