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水果内部品质近红外光谱无损检测研究进展

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摘要 近红外(NIR)光谱分级技术正越来越广泛地应用于水果的产后加工和质量评判中。介绍了水果内部品质光学特性检测原理;分析了规则反射、透射和漫反射 3 种光特性测量方法在水果内部品质不同需求检测中的适用性;化学计量学方法是近红外光谱分析技术的一个重要部分,对一些新的预处理方法和回归算法作了介绍;探讨了水果状态对光谱影响及修正方法,如温度补偿、大小修正等;并阐述了水果的糖度、酸度、硬度等定量检测和褐变、黑心、水心、损伤等定性判别的国内外最新研究进展;分析了近红外技术在水果品质检测和控制方面的应用前景。

关键词 应用光学;近红外光谱;无损检测;化学计量学;光谱预处理;水果;内部品质

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Review of Nondestructive Measurement of Fruit Quality by Means of Near Infrared Spectroscopy

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Abstract An overview of near infrared (NIR) spectroscopy for use in measuring quality attributes of horticultural produce is given. Nondestructive quality inspection and sorting technique based on NIR technique is widely used in post-harvest processing and quality control. Different spectrophotometer designs and measurement principles are compared, and novel techniques for the estimation of light absorption and scattering properties of fruit tissue are reviewed. Chemometrics is an essential part of NIR spectroscopy, and some available preprocessing and regression techniques, including nonlinear ones, are discussed. The effects of orchard state on spectrum, and correction method, such as temperature compensation, are addressed. Most applications of NIR spectroscopy have focused on the nondestructive measurement of soluble solids content of fruit where typically a root mean square error of prediction of Brix can be achieved, and other applications involving texture, acidity or disorders of fruit have also been reported. Finally, the application prospects of this technology were analyzed.

Key words applied optics; near infrared spectroscopy; nondestructive inspection; chemometrics; pretreatment; fruit; interior quality

1 引 言

自从 1985 年美国农业部的 Birth 课题组用近红外(NIR)光谱分析技术检测果蔬品质以来,经过 20 多年的发展,社会认知程度不断提高,检测技术层出不穷,检测理论日趋成熟,检测仪器早已从实验室走出,实际应用逐步扩大,并由在线检测向便携式发展,检测目标有从产后管理向产中管理延伸趋势;检测项目由当初的单一糖度(SSC)指标到如今的苹果等果实内部褐变、水心、淀粉、浅层损伤,柑橘局部失水、浮皮等多项同时检测;检测品种由桃等薄皮中

小型果实向西瓜等厚皮大型果实迈进。通过近红外光谱分析技术实现了品牌经营,提高了果品的竞争力和附加值。

国内在 863,科技攻关,科技支撑,国家自然科学基金等项目的支持和市场引导下,已有数个高等院校、科研院所以及部分企业相继开展了相关研发工作。毕卫红、傅霞萍等^[1~3]已就此专题分别撰写了综述论文,在应义斌、刘燕德等^[4,5]无损检测综述论文中也涵盖了这部分内容,众多学者也进行了专项研究,近红外技术越来越倍受世人关注。

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为了更好地总结过去,展望未来,在参考国内外专业文献的基础上,结合本文作者多年的科研工作,特撰写本文。

2 水果理化特点

光谱不但反映果品的生化特性,同时也反映果品的物理特点,了解果品的理化属性,有利于加深对光谱的认识与理解。

果品的成分相对单一,成分间的相互作用不十分敏感。水是果品中最丰富的组成成分,占果品重量的85%~90%。水分对近红外光吸收强烈,时常覆盖了其他成分信息,易产生干扰。多数果品的品质由成分指标糖酸比和质地指标硬度等来评价,果品的糖度与酸度和质构因品种不同而各异。

例如苹果的可溶性固形物质量分数约占11%~15%,总糖质量分数约为9%~14%。苹果的糖分含量高,信号强,适宜光谱的采集与分析;而总酸含量则较少,约占0.2%~1.6%质量分数,相对糖度而言预测难度增大。苹果的密度为0.835~0.862 g/cm³,轻于水,这有利于光的透射。果品的成分不论是径向还是轴向均呈不均匀状态分布,整体数值常取平均代之。

生理病害也是果品品质评价的重要指标。例如苹果的内部褐变、水心,鸭梨的黑心,柑橘的枯水病等。其他还包括挤压、磕碰等浅层物理损伤等。

3 光谱采集方式

采集光谱首先要考虑光源种类、透反射方式、波长范围、仪器类型等多种因素。

近红外光源布置形式有单光源、多光源之分,常采用卤素灯、发光二极管(LED)和激光三种发光技术。卤素灯技术成熟、价廉物美,需散热装置^[6,7]。LED灯发热少、节能,可使仪器结构简单,降低成本^[8]。激光可省略滤光片,可使特征吸收波长更加准确无误。

光谱通过漫反射或透射或漫透射方式进行采集,如图1所示^[9]。透射和漫透射的优缺点是:1)可以测量果实整体;2)可以测量厚皮果品;3)可以检测果实内部特征;4)只限于易透光物料;5)需要配置高灵敏度、高动态范围检测器。漫透射和透射适宜苹果内部水心、褐腐病、鸭梨内部褐变等果皮较厚的果实。漫反射的优缺点是:1)适合多种果品;2)只能获取一个方向且为果皮附近果肉信息;3)

不能测量柑橘之类的厚皮果实;4)在选果线上,近红外线照射位置一定,而果实大小和人工放在输送装置上的果实位置,将使测定位置产生偏差,从而影响测定精度。漫反射适宜检测果皮较薄的桃、梨、苹果等果实的糖酸度。

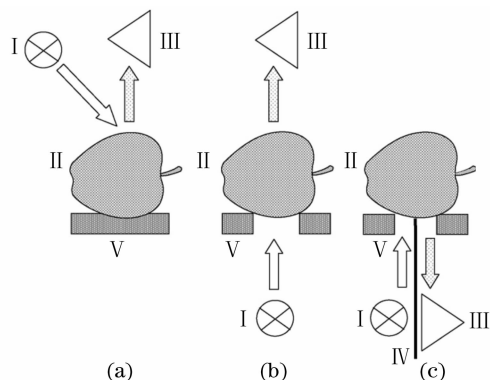


图1 采集方式。(a)漫反射;(b)透射;(c)漫透射;(I)光源;(II)水果;(III)分光器/检测器;(IV)挡光板;(V)承载台

Fig. 1 Setup for the acquisition of (a) reflectance, (b) transmittance, and (c) diffuse transmission. (I) light source, (II) fruit, (III) monochromator/detector, (IV) light barrier, and (V) support

波长范围大致可分为特征波长、短波(760~1100 nm)、中波(1100~1800 nm)、长波(1100~2400 nm)和全波(760~2400 nm)等五种。特征波长用于特定成分、质构的测量分析;短波常结合透射或漫透射方式,其测量对象多为单个物料的深层特征,以获取内部或深层信息为主,例如苹果水心、黑心等,近期开发研制的果品专用仪器基本以此为主流^[6,7];长波常与漫反射方式并用,以获取表层信息,如苹果、桃等糖度,在果品品质近红外检测学术论文中,时常结合化学计量学方法进行研究^[10]。

滤光片型、光栅单色器型、傅里叶变换干涉仪型、声光调谐滤波器型(AOTF)和多通道检测器型为广泛使用的五种类型仪器,滤光片型和光栅单色器型多用于专用仪器,傅里叶变换干涉仪型常见于实验室的通用仪器;检测方式涉及在线检测、现场检测、离线或实验室检测。在线检测、现场检测的目的多为填补技术空白,而离线或实验室检测因其效率高,多为代替现有技术。

4 水果状态与光谱处理方法

近红外光谱技术应用于水果品质检测时,光谱信息受到物料状态影响严重,如成分分布、温度、大

小、品种、产地、收获年份、成熟度等。为了准确、快捷地获得被检目标含量值,很多学者深入研究了各种修正方法,如平均测量法降低水果成分分布不均的影响;温度补偿法修正水果个体间温度差异的影响等。对于不同影响因素采取不同处理方法。

4.1 水果成分分布与光谱处理

果品的糖酸度呈不均匀分布。例如,苹果的果核处的糖度最低,阳面表皮侧最高。沿半径方向上,糖度变化近似二次曲线形状,通过拟合方法可用 $y = ax^2 + bx + c$ 方程表示,从果柄部分到花萼部分苹果糖度逐渐增大^[11]。温州蜜橘果顶部糖度大于赤道部大于果梗部,表皮部大于中心部。酸度则是赤道部大于果梗部大于果顶部,中心部大于表皮部,橘瓣间糖度最大相差1%。甜瓜的内部囊侧糖度大于表皮,糖度最大相差2%。西瓜赤道部位的糖度大于果梗大于果顶,心部大于外部。

对于苹果而言,可通过三种方式消除因成分分布不均产生的影响:1)漫反射光谱与苹果表面糖度。沿赤道分别相隔90°测量4个点的漫反射光谱和Brix值,然后取平均进行建模;2)漫反射光谱与苹果整体糖度;3)透射光谱或漫透射与苹果整体糖度。甜瓜则取果底(花痕)处的漫透射光谱和果肉糖度进行相关分析。桃则常采集与缝合线成90°部位的反射光谱与糖度值代表桃的整体。

4.2 水果温度与光谱处理

近红外光谱易受物料温度影响,因为只要温度发生改变,即使物料的化学成分不变,水的吸收强度也要变化。为此,将温度等同于一个未知的成分值考虑,通过建立温度修正模型加以解决。人为地改变物料温度并测量其相应的光谱,继而建立包含温度变化在内的检测模型即可。由表1多元回归分析的结果可知,无温度修正时,温州蜜橘糖度检测偏差较大,进行温度修正后,基本消除了偏差^[12],表1中SEP为预测标准误差。大場聖司^[13]检测甜瓜糖度时,在多元回归方程中采用了波长839 nm后,可以精确测量物料温度各异的甜瓜糖度。Roger等^[14]采用外部参数正交化偏最小二乘法(external parameter orthogonalisation of PLS, EPO-PLS)处理苹果光谱数据,修正温度对苹果糖度模型的影响,修正后模型预测偏差降到0.3°Brix以下。Ann Peirs等^[15]研究了苹果近红外光谱(900~2000 nm)的反射比与温度的关系,如图2所示,并研究了温度对可溶性固形物模型的影响,采取了相关温度补偿措施,建立混合模型其预测平方根误差RMSEP=0.77°Brix。

表1 温度修正前后温州蜜橘糖度预测结果

Table 1 Prediction results of satsuma SSC before and after temperature compensation

Temperature / °C	Before compensation		After compensation	
	SEP	Bias	SEP	Bias
21	0.49	-0.33	0.41	-0.02
26	0.44	0.05	0.39	-0.01
31	0.50	0.20	0.46	-0.03

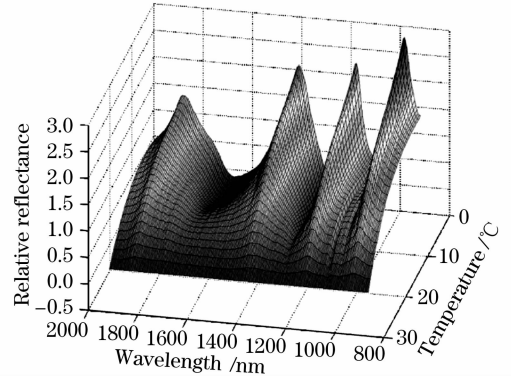


图2 温度对苹果近红外反射光谱的影响图

Fig. 2 Average relative temperature influence on the NIR reflectance spectrum of 'Jonagold' apples

4.3 水果大小与光谱处理

当采用透射方式进行检测时,不能忽略果实大小对光谱的影响,因为果实越大光谱越向上漂移,即使实施2阶导处理,也无法消除果实大小的影响,如图3所示。可将每个果实的光谱换算成同一直径果实的光谱,作为消除果实大小影响的方法^[12],即用每个果径除各个测量波长处的吸光度值。由图4可知,与糖度无关而与果实直径相关的波长844 nm是水的吸收带。通过各个物料的2阶导光谱被各个物料的这个波长2阶导吸光度值相除,可以得到不被果实大小影响的正规化2阶导光谱,如图4所示。

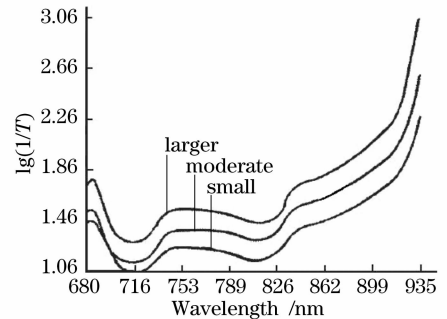


图3 蜜橘透射光谱

Fig. 3 Transmittance spectra of satsuma

4.4 品种、产地、收获年份、成熟度与光谱处理

水果品种繁多,如我国苹果、梨等达到十种甚至是几十种。同类水果品种间差异较大,一般宜采用

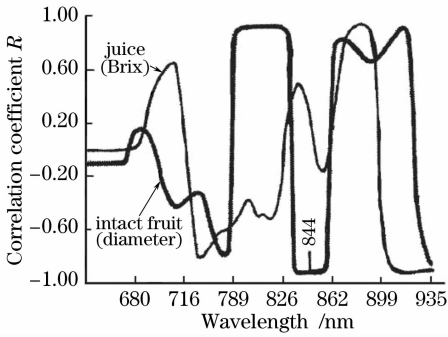


图4 糖度、果径 2 阶导光谱的相关系数图

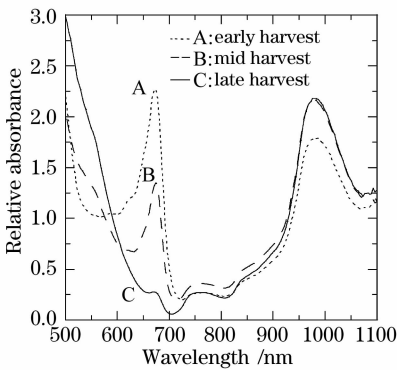
Fig. 4 Spectral COR of SSC, diameter after 2nd derivative

图5 苹果不同时期的近红外吸光度光谱

Fig. 5 Typical absorbance spectra of apples from early harvest (A), mid-harvest (B) and late harvest (C)

分类建模。V. Andrew McGlone 等^[16]采用可见近红外漫反射法研究了‘Royal Gala’苹果采前及采后贮藏时期的品质指标,光谱如图5所示。Ann Peirs 等^[17]研究了苹果自然特性对可见近红外模型预测采摘期精确性的影响,如品种、产地、收获年份、成熟度等。结果表明,近红外光谱与成熟度有一定相关关系,其 $R_p > 0.94$, $RMSEP < 7.7$, 且成熟度不只是与果皮颜色相关,且受到其内部特性综合影响。

5 定量分析

在水果近红外检测研究中,定量分析最早也最多。定量分析一般用于评价水果内部成分含量,如糖度、酸度、硬度等及其维生素含量等。定量分析涉及光谱采集模式、光谱预处理、波段选择、建模方法、模型评价等。

光谱采集模式主要有透射(Transmission)、反射(Reflectance)、漫反射(Interactance)以及透反射(Transflectance)。对于反射又提出连续波(Continuous wave, CW)、近红外反射和时间分辨(Time-resolved, TRS)近红外反射方法。近红外光

谱数据预处理方法主要有平滑(Smoothing),包含卷积平滑(Savitzky-Golay filter, S-G Smoothing)和求导滤波平滑(Norris derivative filter)。微分(Derivative),包含一阶微分(first-order Derivative, 1st D)和二阶微分(second-order Derivative, 2nd D)。标准归一化(Standard normal variate, SNV)、多元散射校正(Multiplicative signal correction, MSC)、小波变换(Wavelet transform, WT)、正交信号校正法(Orthogonal signal correction, OSC)、净分析物预处理法(Net analyte preprocessing, NAP)及多种方法联合应用。有效建模波段选择方法主要有相关系数法(Correlation coefficient, COR)、无信息变量消除法(Removing uncertain variables, RUV)、间隔偏最小二乘法(interval Partial least squares, iPLS)、反向间隔偏最小二乘法(Backward interval Partial least squares, BiPLS)、正向间隔偏最小二乘法(Forward interval partial least-squares, FiPLS)、移动窗口偏最小二乘法(Moving window partial least-squares, MWPLS)、遗传算法(Genetic algorithm, GA)、独立分量分析方法(Independent component analysis, ICA)等。

建模方法主要有多元线性回归(Multiple linear regression, MLR)、逐步多元线性回归(Stepwise multiple linear regression, SMLR)、主成分回归(Principal component regression, PCR)、偏最小二乘法(Partial least squares, PLS)、核函数偏最小二乘法回归(kernel Partial least squares, kernel PLS)、混合线性分析(Hybrid Linear Analysis, HLA)和人工神经网络(Artificial neural network, ANN)等。

在表2中列出了近红外光谱技术在水果品质检测中的应用,包括水果品种、检测对象、采谱方式、波段范围、预处理方法及预测标准偏差。其中RMSEP为最优模型预测结果,当给出值为预测误差(SEP)和偏差(bias)时根据公式 $RMSEP^2 = SEP^2 + bias^2$ 转换。

6 定性分析

定性分析常用于水果的内部品质判别,如苹果的水心、褐腐病、果肉褐变及梨黑心病等;浅层损伤和内部缺陷,如苹果碰伤、柑橘局部失水等以及水果产地、品种鉴别等。

国内外研究者对苹果的水心、褐腐病、果肉褐变和梨的黑心检测进行了大量研究,根据检测目标特

表 2 近红外光谱技术在水果品质检测中的应用

Table 2 Applications of NIR spectroscopy to measure SSC, firmness and other qualities of fruits

Fruit Species	Acquisition mode	Spectral range /nm	Pretreatment and calibration means	RMSEP	References
1. SSC or sugar content /($^{\circ}$ Brix)					
Apple					
Fuji	Interactance	812~2357	1st D, PLS	0.452	Liu ^[18]
	Reflectance	909~2632	BiPLS, FiPLS, PLS	0.732	Zou ^[19]
	Reflectance	1300~2100	None, HLA	0.485	Zhang ^[20]
	Reflectance	630~1030	S-G Smoothing, PLS	0.941	Dong ^[21]
	Interactance	1000~2500	MSC, S-G Smoothing, GA, PLS	0.797	Wang ^[22]
	Reflectance	909~2632	SNV, WT, iPLS, PLS	0.411	Zou ^[23]
	Reflectance	1300~2100	OSC, NAP, PLS	0.492	Zhao ^[24]
	Reflectance	909~2632	ICA, PLS	0.436	Zou ^[25]
Delicious	Reflectance	800~1690	1st D, WT, kernel PLS	0.441	Nicolai ^[26]
	Reflectance	810~999	COR, MLR	1.06 *	Ventura ^[27]
Idared	Interactance	400~1100	SNV, PLS	0.940	Zude ^[28]
Gala	Reflectance	400~1800	None, PCR	0.279 *	Park ^[29]
	Interactance	600~1000	Smoothing, 2nd D, PLS	0.72	McGlone ^[16]
Jonagold	Reflectance	810~999	COR, MLR	1.10	Ventura ^[27]
Pear					
Conference	Reflectance	780~1700	CW, TRS, PLS	0.44	Nicolai ^[30]
Xueqing	Reflectance	800~2632	GA, PLS	0.395	Ying ^[31]
Crystal pear	Transmission	643~928	Smoothing, 2nd D, PLS	0.464	Zhang ^[32]
Mandarin					
Satsuma	Reflectance	350~2500	MSC, PLS, PCR	0.162	Gómez ^[33]
Orange	Reflectance	570~1048	1st D, SNV, PLS	0.461	Cayuela ^[34]
		1100~1850			
Citrus	Transmission	500~1000	1st D, 2nd D, MSC, SNV, PLS, PCR	0.538	Lu ^[35]
Mango	Interactance	400~1100	2nd D, PLS	0.67 *	Subedi ^[36]
	Reflectance	1100~2500	Smoothing, 1st D, MLR, PLS	0.70	Saranwong ^[37]
	Reflectance		Smoothing, 1st D,	1.18	
Kiwifruit	Interactance	300~1100	2nd D, MSC, SNV	0.93	Schaare ^[38]
	Transmission		PLS, PCR	0.89	
	Interactance	400~1100	2nd D, PLS	0.39	McGlone ^[39]
Peach					
Honey peach	Interactance	800~2500	Smoothing, PLS	0.534 *	Liu ^[40]
Dabaitao	Interactance	800~2500	SNV, MSC, 1st D, 2nd D, PLS	0.336	Ma ^[41]
2. TA/PH					
Apple					
Fuji	Interactance	1100~2357	1st D, PLS	0.068	Liu ^[18]
Gala	Interactance	500~1100	Smoothing, 2nd D, PLS	0.052	McGlone ^[16]
Jonagold	Reflectance	380~1650	None, PCR, PLS	0.068	Lammertyn ^[42]
Pear					
Xueqing	Reflectance	800~2632	COR, PLS	0.019	Liu ^[43]
Xueqing	Reflectance	800~2632	GA, PLS	0.020	Ying ^[44]
Mandarin					
Mandarin	Reflectance	350~2500	MSC, PLS, PCR	0.115	Gómez ^[33]
Orange	Reflectance	570~1048	1st D, SNV	0.331	Cayuela ^[34]
		1100~1850	PLS		

(续表)

Fruit Species	Acquisition mode	Spectral range /nm	Pretreatment and calibration means	RMSEP	References
3. Firmness (N)					
Apple					
Jonagold	Reflectance	380~1650	None, PCR, PLS	2.49 [*]	Lammertyn ^[42]
Delicious Gala	Reflectance	400~1800	None PCR	4.91 [*]	Park ^[29]
Pear					
Xueqing	Interactance	800~2630	1st D, 2nd D, SMLR, PCR, PLS	4.26	Fu ^[45]
Conference	Reflectance	780~1700	CW, TRS, PLS	0.4	Nicolai ^[30]
Kiwifruit	Interactance	400~1100	2nd D, PLS	7.8	McGlone ^[39]
Mandarin	Reflectance	350~2500	MSC, PLS, PCR	8.313	Gómez ^[33]
Watermelon	Interactance	650~950	S-G Smoothing, PCR, PLS	0.589	Tian ^[46]
4. V_c (mg/100 g)					
Orange	Interactance	833~2500	1st D, MSC, WT, PLS	3.9	Xia ^[47]
Citrus	Interactance	833~2500	WT, PLS	3.9	Xia ^[48]

* : SEP value.

性,多采用可见/近红外透射光谱法。韩东海等^[49]采用光密度差分法对水心苹果等级进行了分析,采用810 nm和760 nm处光强值能较好判别轻度和重度水水果;韩东海等^[50]还利用可见/近红外连续透射光谱技术(650~900 nm)对苹果内部褐变进行了研究,选择715 nm,750 nm和810 nm 3个特征波长进行了褐变果判别分析,实验结果表明,样品的正确判别率达到95.65%;C. J. Clark 研究组^[51]通过不同的采集方式和回归方法,利用透射法(300~1140 nm)检测苹果内部褐变,结果显示当果轴水平,在光源与检测器呈一定角度的条件下采集光谱,用PLS建模时效果最佳($R = 0.91$, $RMSEP = 7.9$);V. Andrew McGlone等^[52]采用两种光谱扫描系统在线检测苹果褐变,其分级传送速度达到500 mm/s,且预测模型具有较高精度($RMSEP < 4.1\%$),可用于商业化;王加华等^[7]采用可见/近红外能量光谱法快速判别苹果褐腐病、水心,图6为水

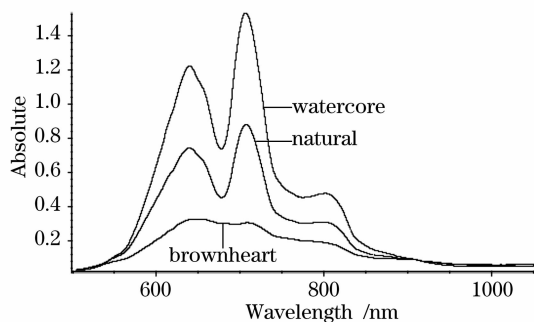


图6 水心、正常、褐腐苹果绝对能量平均光谱
Fig. 6 Average spectra of absolute energy of watercore, natural and brownheart apples

心苹果、褐腐病和正常苹果的能量光谱图,采用多种分析方法建立模型,褐腐病苹果判别率都为100%,水心苹果判别率最高达到96.7%,为在线分选提供了一种新思路;Han等^[53]采用近红外透射法检测鸭梨黑心,如图7所示。图7为不同程度黑心鸭梨的透射光谱图,采用全光谱的马氏距离判别法可将黑心梨完全剔除;Fu等^[54]做了近红外透射法(400~1028 nm)和漫反射法(Si: 670~1110nm; InGaAs: 800~2630 nm)检测雪青梨黑心的比较研究,发现透射法优于漫反射法,其判别率达到91.2%。

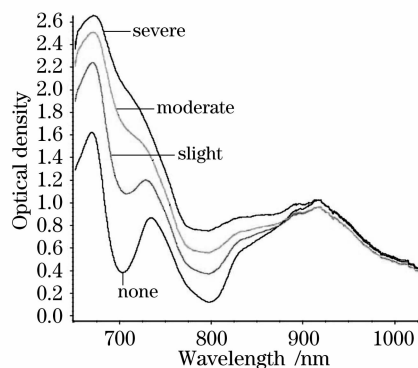


图7 不同程度黑心鸭梨的透射光谱图
Fig. 7 Transmitted spectrum of pears with different degrees of brown core

水果在采摘、运输、分选、包装过程中,由于碰撞或挤压不可避免地会造成损伤,损伤检测也广受关注。损伤主要为浅层损伤,多采用近红外漫反射法进行检测。J. Xing等^[55]研究了苹果损伤类型对近红外检测的影响,损伤主要有碰伤和压伤,其模型相关系数 CR^2 分别为0.74和0.68,碰伤判别率比压伤

判别率高;J. Xing等^[56]还采用可见/近红外反射法(400~1700 nm)研究‘Golden Delicious’苹果损伤后不同贮藏时间的检测精度,其研究结果表明,1天后的检测精度为83.67%,21天后的判别准确率高达98.03%;J. Xing等^[57]研究了Jonagold苹果损伤的近红外检测,比较了选用不同波段及预处理方法进行建模时的判别准确度,结果显示经MSC处理后精度为91.53%,波段选择影响较小;同时J. Xing等^[58]采用近红外漫反射技术评价苹果碰伤后的软化指数,检测苹果新鲜碰伤。

近红外光谱结合化学计量学算法也可用于水果的品种鉴别。Li等^[59]采用可见/近红外光谱法判别草莓品种,不同化学计量学算法用于数据处理及建模,如主成分分析(PCA)结合人工神经网络(ANN),识别率达到95%。Xie等^[60]采用可见/近红外光谱结合化学计量学方法判别转基因西红柿,其马氏距离法(Discriminant analysis, DA)和偏最小二乘判别法(Partial least-squares discriminant analysis, PLSDA)的判别率分别为96.95%和100%。赵杰文等^[61]研究了支持向量机(Support vector machine, SVM)在苹果分类的近红外光谱模型中的应用,结果显示不同产地苹果分类模型的回判识别率为87%,预测识别率为100%。李晓丽等^[62]采用主成分和多类判别分析方法结合可见/红外光谱鉴别水蜜桃品种,蜜露水蜜桃、大白桃水蜜桃和红仙玖水蜜桃判别率都为100%。

7 未来趋势

在采后处理方面。提高农业生产效率、降低农业劳动强度需要现代化技术。运用了现代无损检测技术的果品自动化分选设备,可以实现每一个果品内外品质的快速无损检测、分级。作为水果采后处理的一项新技术,不论国内外均已开始普及应用,今后还将向多技术糅合方向发展。

在产中管理方面,便携式近红外水果品质检测仪的问世,为水果的产中管理提供了强有力的支持。通过树上未成熟期水果糖度的监测,可以把握果品品质生长趋势、预测收获期,结合树势信息可以指导翌年的整枝、剪枝、施肥等生产。由此可见,在线检测和便携式检测是水果生产中的主要发展趋势。

在科研方面,利用近红外技术提高梨的内部黑心、苹果等初期浅层褐变正确判别率和果品硬度定量计算准确性尚有一定的难度。

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