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Abstract: This article reviews various methods that could be used to determine the geographic origin of cultured pearls, potentially allowing a consumer to trace them back to the farm. Chemical marking using different substances is possible due to the porosity of the nucleus and nacre. It is also possible to affix a logo marker to the nucleus that can later be imaged using X-radiography. In addition, radio-frequency identification chips are today so small that they can be housed within the nucleus of a cultured pearl. Also discussed is the potential of using trace-element chemistry to differentiate mollusc species and pearling regions. Carbon and oxygen isotopes could also be useful given that they reflect the waters in which a cultured pearl grew, and DNA testing may offer options in the future.

Keywords: cultured pearl branding, cultured pearl traceability, LA-ICP-MS, RFID chips, shell and cultured pearl DNA



Introduction

Branded jewellery products are more successful than non-branded goods (Kapferer and Bastien, 2009). There is continued demand from jewellery consumers for branded goods and increasing desire for traceability of products (Conroy, 2007; Ganesan *et al.*, 2009). Cultured pearls are an interesting case study where some products are branded (e.g., *Figure 1*), but traceability to source is something that is difficult to verify independently at present. A cultured pearl strand with a branded tag does not provide a clear guarantee of origin for the end consumer, given that individual cultured pearls can easily be exchanged or strands re-strung. At the same time, there is a growing interest in tracing cultured pearls through the supply chain, so that an end consumer knows which farm their cultured pearls came from. Producers who operate responsibly are investigating ways of marking their cultured pearls so that provenance can be guaranteed to the end consumer.

Any method used to trace cultured pearls must largely be invisible so as to maintain the commercial value of the end products. Cultured pearls are produced both with a nucleus (e.g., Akoya, South Sea and Tahitian) and without a nucleus (e.g., Chinese freshwater beadless products); for general reviews, see for example Gervis and Sims (1992) and Southgate and Lucas (2008). Different labelling/traceability approaches may be required for these two types of cultured pearls, based on their internal structure. This article reviews a wide range of methods — chemical, physical and biological — that potentially could be used in tracing cultured pearls through the supply chain.

Chemical marking

Pearls consist of fine polycrystalline calcium carbonate (CaCO₃) crystals and traces of organic matter. The mother-of-



Figure 1: A branded necklace of South Sea cultured pearls (12 mm in diameter) produced by Atlas Pearls in northern Bali and West Papua (Indonesia). Photo courtesy of Atlas Pearls, Claremont, Western Australia.



Figure 2: Cross-section of a 'chocolate' beaded cultured pearl. The lightcoloured bead (i.e., nucleus) and the darker overgrowth are clearly visible. It is evident in the enlarged image at the bottom right that the brown colour has been artificially added. This demonstrates the porosity of a cultured pearl and its potential for absorbing chemically doped or colour-doped solutions. The colour has penetrated approximately 0.5 mm. Photo by H. A. Hänni.

pearl (also called nacre) surface of pearls is made up of aragonite tablets. A pearl's porous structure means that it has a good potential for absorbing chemically doped or colour-doped solutions. A good example of this are dyed cultured pearls (e.g., Figure 2), which can be found in many different colours (Hänni, 2006; Strack, 2006). In a similar way, cultured pearls from selected producers could be marked using a colourless doped solution - that is unique to a pearl producer - after harvest. If chemically doped, these pearls could later be identified in a gemmological laboratory using EDXRF spectroscopy (Hänni, 1981). However, the applicability of this approach is limited given that EDXRF spectroscopy is not in widespread use in the jewellery industry.

Alternatively, rather than marking the cultured pearl after harvest, one could mark the nucleus before insertion using a specific solution. However, if the nacreous overgrowth is too thick, it may not be possible to identify the chemical signal from the nucleus. Another approach would be to remove a tiny amount of nucleus material from a drilled cultured pearl for chemical analysis.

The authors have experimented with the diffusion of fluoroamine (NH₂F) into a cultured pearl, something a pearl farmer could easily do. The subsequent detection of fluorine could then be linked back to that farm. Fluorine is a relatively light element that is not detectable by EDXRF spectroscopy, but is best analysed by nuclear magnetic resonance (NMR). However, NMR is cost-intensive and the instrument's sample chamber is typically smaller than the diameter of a cultured pearl.

If only a limited number of pearl farms are involved in such chemical marking of their cultured pearls, it could be viable to supply each of them with different cost-effective and nontoxic chemicals that could be detected in a gemmological laboratory.

Labelling the nucleus or the surface of a cultured pearl

Initial experiments using physical labels affixed to a cultured pearl nucleus were carried out in 2010 by author HAH. Thin (0.05 mm) rings consisting of gold wire were affixed to several Mississippi shell nuclei (the nucleus material commonly used in the pearl industry) and used to produce cultured pearls. The aim was to investigate the possible rejection of labelled nuclei by the molluscs and to see whether this gold label (or the associated adhesive) would influence cultured pearl growth. Results after six months showed that the labelling materials (gold and glue) had no influence on cultured pearl production and this spurred further efforts

to investigate the production of nucleus logos.

Any such logo marker must be extremely thin, be composed of noble metal (and therefore be resistant to corrosion) and have the same convex shape as the nucleus to ensure that the resulting cultured pearl is also round. However, the production of such round metal labels, generally 3–4 mm wide and 0.05 mm thick, is relatively expensive. Different label production techniques were tested, such as galvanic production, pressing, etching and cutting with a



Figure 3: Silver logo labels (3 mm in diameter) for a pearl farm. These can be affixed onto the bead prior to insertion and later be used to trace a beaded cultured pearl back to its farm. Photo by H. A. Hänni.

laser or water jets; these are widely used techniques in manufacturing (Schultze and Bressel, 2001). The water jet technique was most precise for cutting the contours of the logo, but still considered too expensive.

Several dozen logo tags (e.g., *Figure 3*) were affixed to shell nuclei and sent to different marine farms to be tested in cultured pearl production. After the usual 12–18 month growth period, these 'tagged' cultured pearls were harvested and successfully examined with X-radiography *(Figure 4).* Due to the position of the logo in the peripheral part of a cultured pearl, there is only a statistically small chance of the logo being damaged during drilling.

The production of such logo markers is relatively expensive, even if produced in large quantities. In addition, these cultured pearls need to be tested using X-rays, which is relatively unfeasible for a jeweller. (X-rays used for medical purposes, such as in dentistry, are not strong enough to visualize all required details within a cultured pearl of, e.g., 10 mm.) Nevertheless, for beaded cultured pearls that use a nucleus (e.g., Akoya, South Sea and Tahitian), this method is an option. For beadless cultured pearls (e.g., Chinese freshwater cultured pearls), the introduction of a label together with the saibo (donor mantle tissue) would have the disadvantage of positioning the logo in the centre of the cultured pearl, resulting in a high likelihood of damage during the drilling process.

Another approach is to mark the surface of the cultured pearl rather than the nucleus. This could involve either laser engraving with a unique number (similar to laser inscriptions on diamonds) that can later be used to identify its source or embossing a hologram onto the surface of the cultured pearl that can be read with a suitable reader. Both of these methods are currently being investigated in French Polynesia ('Redonner ses Lettres...', 2013; 'Le Tahiti Pearl Consortium Disparaît', 2013). These methods are slightly destructive to a cultured pearl's surface and it remains to be seen if they are acceptable to the pearl trade.



Figure 4: X-radiographs of three Tahitian cultured pearls with a branded nucleus. The farm-specific logos are in silver, which has a high density making it quite visible with X-rays. Three cultured pearls are shown in two slightly different orientations in this composite image. The diameter of the cultured pearls is approximately 8 mm and the width of the logos is 3 mm. Image by H. A. Hänni.

RFID – radio frequency identification

Radio frequency identification (RFID) technology has undergone rapid development in the past decade and is now a widely used method in many technology applications (Want, 2006). It is increasingly being employed in jewellery management solutions (Wyld, 2010). Through the miniaturization of RFID chips (transponders in millimetre sizes), the use of electromagnetic frequencies is a feasible option for the tagging/traceability of cultured pearls. Transponders are chips that contain relevant data which can be accessed with an RFID reader. These devices are inexpensive and they could be easily used in jewellery retail stores (June HK Fair Special...', 2013). Information stored on the chips could include the production location, harvest date and details about the pearl farm. Additional information can be added to the RFID chip after a cultured pearl has been harvested, including its quality grade, inventory data and unique identification information that could be useful for theft recovery.

RFID chips have been introduced into commonly used Mississippi shell nuclei, which are currently being piloted by pearl farmers in the Pacific Ocean.



Figure 5: A composite shell bead that has been sliced and polished to show a small RFID chip (3 mm long) embedded within it. The information on such a chip can be accessed using an RFID reader. Photo by H. A. Hänni.



Figure 6: X-ray shadow images of bead nuclei (7.5 mm diameter) consisting of pieces of shell with embedded RFID chips. These are being marketed by Fukui Shell Nucleus Factory. Image by H. A. Hänni.

One nucleus manufacturer (Fukui Shell Nucleus Factory, Hong Kong) has already brought to market nuclei that contain RFID chips (see 'June HK Fair Special...', 2013). *Figure 5* shows such a 'micro-chip embedded nucleus' which, depending on its size, costs US\$2–3 per piece. According to the manufacturer, these nuclei consist of two layers of shell material (i.e., laminated nuclei) and a 3 mm RFID chip that is located 1 mm below the surface of the nucleus (*Figure 5*). *Figure 6* shows an X-ray shadow image of such chipembedded nuclei.

One disadvantage of these nuclei is the relatively high cost of the chips, which would be wasted in cultured pearls of low quality. Also, the 3 mm size of the straight-edged chips is rather large when taking into account that the nucleus has a spherical shape. The size and position

Ha	bitat								S	ultwater						
Sa	mple	Pinctada	maxima (si	lver) shell	Pinctada	maxima (g	old) shell	Pincta	ida radiata	shell	Pinctada m	argaritifera cul	ltured pearl	Pinctada m	argaritifera cu	ltured pearl
So	urce		Indonesia			Philippines		UnitedA	rab Emirate	s (RAK)	Rangi	roa, French Poly	nesia		Fiji	
CaO	wt.%	56.03	56.03	56.03	56.03	56.03	56.03	56.03	56.03	56.03	56.03	56.03	56.03	56.03	56.03	56.03
Na_2O	wt.%	0.91	0.91	0.88	0.84	0.84	0.86	0.97	0.92	0.95	0.75	0.75	0.74	0.90	0.86	0.93
Li	ppm	0.53	0.40	0.40	0.40	0.33	0.23	0.56	0.50	0.56	0.38	0.44	0.40	0.57	0.64	0.69
В	ppm	18.2	20.5	21.2	15.1	15.1	15.6	1.8	1.9	1.7	10.7	10.8	10.6	16.3	14.6	14.0
Mg	ppm	169	180	199	115	115	121	476	392	463	206	198	226	120	98	98
Ь	bpm	34.0	33.0	35.0	6.6	6.5	6.6	73.2	71.5	71.7	13.8	13.2	12.9	13.3	13.5	12.7
К	ppm	71.1	63.6	51.1	131	157	180	104	118	106	60.0	51.2	42.7	82.7	87.2	83.7
Cr	ppm	2.3	1.9	2.1	2.0	1.9	1.8	2.0	1.8	2.0	2.5	2.2	2.2	2.3	2.4	2.2
Mn	bpm	3.4	3.3	3.4	6.2	6.6	6.2	0.19	0.18	0.18	1.3	1.4	1.4	88.5	92.9	56.9
Fe	ppm	18.4	19.6	17.3	18.3	19.1	22.8	15.6	18.1	18.1	26.9	26.7	29.0	25.4	26.8	28.3
Cu	ppm	0.14	0.08	<0.06	0.09	0.11	0.10	0.26	0.35	0.29	0.11	0.10	0.06	0.04	0.08	0.07
Zn	ppm	<0.08	0.34	0.41	0.38	0.44	0.55	0.59	0.73	0.59	0.28	0.21	0.45	<0.05	0.31	0.12
Sr	ppm	1030	1070	1130	1040	1040	1080	802	775	795	166	169	163	964	138	113
Ba	ppm	0.35	0.49	0.43	0.28	0.26	0.30	0.17	0.15	0.23	0.28	0.37	0.30	0.19	0.39	0.23
$^{\mathrm{Pb}}$	ppm	0.12	0.10	0.10	3.0	3.6	14.9	0.13	0.13	0.12	0.06	0.05	0.05	0.05	0.05	0.04
Ag	ppm	<0.003	<0.004	<0.006	<0.005	0.01	0.01	<0.009	<0.009	<0.004	<0.004	<0.004	<0.004	<0.005	<0.005	<0.005
Η	bitat			Saltv	vater							Freshwat	er			
Sa	mple	Pte	ria sterna s	hell	Pteri	ia penguin s	shell	θ	vriopsis she	П		Unio shell		Mi	ng cultured pe	arl
Sc	urce		Mexico		Irian	ı Jaya, Indon	esia		China			Scotland			China	
CaO	wt.%	56.03	56.03	56.03	56.03	56.03	56.03	56.03	56.03	56.03	56.03	56.03	56.03	56.03	56.03	56.03
Na_2O	wt.%	0.97	0.95	0.88	0.67	0.66	0.70	0.28	0.28	0.28	0.25	0.25	0.25	0.34	0.34	0.35
Li	ppm	0.64	0.57	0.47	0.47	0.56	1.14	<0.07	0.12	0.09	<0.07	<0.09	<0.08	<0.03	0.07	<0.03
В	ppm	5.0	5.5	7.3	8.5	8.5	9.7	4.0	4.0	3.5	3.7	4.4	4.4	0.20	<0.13	<0.14
Mg	ppm	132	73	57	165	147	112	23.9	23.5	15.4	18.3	18.7	56.4	14.0	20.0	18.0
Р	ppm	123	122	135	158	150	121	192	183	188	100	113	127	183	191	160
К	ppm	65.7	66.0	65.0	41.8	40.8	50.5	19.6	8.9	5.6	34.6	66.4	95.7	19.0	28.0	6.0
Cr	ppm	2.2	2.0	2.0	2.1	2.2	2.2	2.1	1.7	1.6	2.3	1.4	1.8	2.3	2.6	2.3
Mn	ppm	1.7	1.5	0.8	1.6	1.5	2.2	661	1580	861	519	501	477	133	268	137
Fe	ppm	11.6	13.4	12.8	12.4	12.4	13.7	13.3	12.3	15.1	14.9	16.3	16.3	28.0	29.8	32.4
Cu	ppm	0.10	0.10	0.07	0.06	0.05	0.05	0.17	0.20	0.22	0.33	0.25	0.38	0.25	0.41	0.38
Zn	ppm	0.41	0.40	0.56	0.43	0.33	0.30	0.29	<0.11	0.18	0.67	0.26	0.53	2.3	2.6	1.8
Sr	ppm	1000	908	923	1280	1320	1170	761	877	360	250	260	267	305	446	409
Ba	bpm	0.20	0.17	06.0	1.11	0.96	1.03	247	288	225	469	516	543	57.6	117.3	76.2
Чd	ppm	0.36	0.45	0.40	0.27	0.24	0.29	0.02	0.02	0.02	0.12	0.07	0.14	0.05	0.06	0.13
Ag	ppm	<0.003	0.005	<0.003	<0.002	<0.003	<0.003	<0.006	<0.003	<0.005	<0.004	<0.004	<0.005	<0.02	<0.02	<0.02
* CaC but	= 56.03 w could not 1	rt.% was used	d as an inter	an iodine sta	on the basis - ndard. Be, Al	of the CaCO ₃ , Sc, Ti, V, Cd	, formula for o, Ni, As, Rb,	aragonite and Y, Cd, REEs	d calcite. Ag (La, Ce, Nd,	was measure Tb, Yb and :	ed to identify cu Lu), and Bi wer	ltured pearls dyc e measured at oi	ed black using s r just below the	ilver nitrate. Iod detection limit (ine was analyse sub-ppm). Each	d in all samples sample was
anal	ysed in thi	ee different	spots, corres	ponding to t	he three colu	mns for eact.	i sample.									

Table I: LA-ICP-MS analyses of cultured pearls and shells from various species and locations.*

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of these chips within the nucleus means they may often be damaged during the cultured pearl drilling process. Rapid developments in RFID technology are promising, but we may need to await the further miniaturization of the chips before they become a feasible option for the cultured pearl industry.

Advanced fingerprinting of pearl and shell materials

Laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) has become more widely used in the last few years in geosciences, even in gemmology (e.g., Saminpanya et al., 2003; Abduriyim and Kitawaki, 2006). Many laboratories and researchers now employ it for the chemical characterization of gems because it has a low detection limit and can also detect light elements. With this method it is possible to carry out high-resolution spot analyses, which allows us to take into account possible chemical zoning in gem materials, including cultured pearls. The technique has been used for characterization of cultured freshwater pearls (Jacob et al., 2006) and natural saltwater pearls from Australian Pinctada maxima molluscs (Scarratt et al., 2012). To our knowledge, there are no published LA-ICP-MS data on a wider range of cultured pearls or shell samples from various mollusc species.

For this study, a preliminary LA-ICP-MS investigation of cultured pearls and shell material was undertaken at the University of Bern. The instrumentation used a 193 nm ArF laser, and synthetic glass (SRM612) was used as a standard for calibration before and after each round of measurements. This was also done to ensure the reproducibility of measurements and detect possible impurities in the chamber that might affect subsequent data. The pits produced on the surface of the samples during ablation had a diameter of 160 µm. As such, the technique is quasi-nondestructive.

Table I lists the results for the seven shell samples and three cultured pearls from different locations that were



Figure 7: The Atlas Pearl farms that produced the necklace shown in Figure 1 are located in Bali (shown here) and West Papua, Indonesia. Giving consumers access to the origin of their cultured pearls may create additional value for pearl farmers. Photo by L. Cartier.

analysed. It is clear that further research is required to compile a useful LA-ICP-MS database that might permit origin determination of cultured pearls from different species.

Another possible (and nondestructive) method for chemically fingerprinting gem materials is particle-induced X-ray emission (PIXE), which has been applied to ruby and emerald (Calligaro et al., 1999; Yu et al., 2000). More recently, PIXE was used on cultured pearls (Murao et al., 2013). Other studies have measured oxygen and carbon isotopic values of nacre and cultured pearls in an attempt to identify geographic origin (Yoshimura et al., 2010). However, all these techniques remain academic and expensive, and they presently do not fulfil the requirements for a rapid and cost-effective tracing method for cultured pearls.

A final method that is very new but merits description is DNA fingerprinting of cultured pearls. Oyster shells and pearls have a biological origin and contain small amounts of organic matter between aragonite layers and in the form of organic pockets. A recently published study described how DNA can be extracted from this organic material in cultured pearls in a practically nondestructive manner (Meyer *et al.*, 2013). The DNA can be used to identify the oyster species of the cultured pearl and the authors also proposed that geographic origin determination might also be possible using next generation sequencing (NGS) techniques in the near future. A similar approach has been used for geographic origin and species determination of ivory (Wasser *et al.*, 2004).

Conclusion

The aim of this review is to show the range of currently available methods that potentially could be used to trace cultured pearls through the supply chain. Supply chain accountability and product traceability are becoming increasingly important issues in the jewellery industry. The branding strategies of various producers, wholesalers and jewellery companies would benefit from additional support through an efficient traceability method. Furthermore, there is a potential for responsible pearl farmers (e.g., *Figure 7*) to capture greater value for their products if they can be traced all

the way to the consumer, but the supply chain accountability and provenance need to be guaranteed (Conroy, 2005; Cartier 2012; Cartier and Ali, 2012). As technology continues to evolve, the search for methods to trace cultured pearls through the supply chain should be addressed in collaboration with the gemmological community and the focus should be on developing cost-effective solutions that are feasible for those at all levels of the supply chain (producer, wholesaler, retailer and consumer).

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