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PURGING GRAIN BULKS WITH NITROGEN: PLUG-FLOW AND MIXING PROCESSES OBSERVED UNDER FIELD CONDITIONS

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ABSTRACT

Six experimental, full-scale applications of nitrogen (N_2) to grain bulks were extensively monitored. These trials showed that in practice there are often substantial deviations from the theoretical behaviour expected when a gas, such as N_2, is used to displace air from a particulate commodity, such as cereal grain, in store. In the most efficient form of oxygen (O_2) displacement, plug-flow would be expected in the grain bulk, followed by dilution of the headspace by a free mixing process. There was some dispersion in practice as the purging front transited the bulk; most regions lost O_2 by this disperse plug-flow process. Using rudimentary introduction systems, in practice, some areas of grain bulks were not effectively purged by disperse plug-flow processes but instead they lost O_2 by dilution, leading to inefficient use of purge gas. These areas were apparently associated with low pressure gradients and, possibly, also with high levels of grain dust. Dilution of the headspace generally followed the expected logarithmic fall in O_2 content with time.

Purging efficiencies overall were 73–92%, based on the plug-flow/free mixing model, with N_2 usage of 1.2–2.0 $m^3\,t^{-1}$ achieving 1% O_2 atmospheres throughout.

Deviations from ideal behaviour reduced efficiency somewhat, but they were not sufficient to prevent target atmospheres from being achieved in the cases reported.

INTRODUCTION

Storage under nitrogen (N_2)-based controlled atmospheres (CA's) is one of the few means currently available for providing inexpensive, residue-free pest control in dry bulk grain. In some specialised cases, even using liquid N_2 as an atmosphere source, it is already cost-competitive with phosphine fumigation. Part of the export grain terminal at Newcastle, New South Wales, Australia, has used N_2-based CA's routinely since 1993, successfully treating more than 100,000 t of grain. It is to be expected that with the cheaper atmosphere sources now under trial there will be increased interest in CA use, particularly in situations where safety considerations and local regulations place onerous restrictions on fumigation and those where grain protectants cannot be used, such as for market reasons.

There have been a substantial number of studies published (Banks *et al.*, 1980a; Tranchino *et al.*, 1980; Agboola, 1993; Cassells *et al.*, 1994) which describe the experimental treatment of bulk grain with N_2 atmospheres under full-scale conditions. These studies provide a sound basis for the application of CA's in a wide variety of well-sealed grain storage types in a wide range of locations and systems (Banks *et al.*, 1980b).

These publications have tended to concentrate on successes, not failures. They have included little, if any, discussion of the difficulties encountered. However, in general, failures are more instructive than successes. In this paper we have concentrated particularly on deviations from expected behaviour, as it is these deviations that are most likely to result in such operational problems as increased gas usage, incomplete effectiveness or the need for prolonged exposure.

The data on which this paper is based was gathered during the full-scale proving trials carried out in Australia during the 1970's as part of the development program for commercial use of CA's. Summary details of these trials have already been published (Banks, 1979). Only the purge phase of the operation is discussed here, as it is valid to conclude that if an appropriate atmosphere can be created by a certain N_2 application rate, it can be maintained by a rate of addition that is less than the adequate purge rate. The trials were aimed at creating an insecticidal 1% O_2 atmosphere in a number of different structures, but here, for the purpose of illustration, a 2% O_2 level is often chosen as a successful limit. This is the level regarded as the maximum which is lethal to all species of stored-produce insects (Bailey and Banks, 1975).

EXPECTED BEHAVIOUR

The system under consideration here, the addition of N_2 to a grain bulk, is a specific case of addition of a fluid to a packed bed. The latter is a common process in the chemical industry, and the behaviour of the added fluid and its interaction with the fluid already present in the bed can be understood in chemical engineering terms. The mathematics and terms used by Levenspiel (1976) to describe the progress of a step concentration change through a packed bed are used here as one model to describe the behaviour of a N_2 purge passing through a grain bulk. It is not expected that there will be direct displacement of the interstitial air by N_2 (true plug-flow) because a limited degree of mixing occurs at the N_2-air interface. This may be caused by molecular diffusion, due to partial mixing as the N_2 enters the bulks. The mixing leads to a moving zone where the O_2 concentration is less than that in the existing storage atmosphere but substantially more than that in the incoming gas (<0.01% O_2). The process is known as 'disperse plug-flow'. Limited degrees of dispersion give a sigmoid profile for O_2 concentration against distance within the zone. The fall in O_2 concentration with time as the mixing front passes a given point is also sigmoid. Both these profiles, O_2 level with time at a point and O_2 level with distance at a given time, can be transformed to give a linear trend if the percent concentration of O_2 is expressed as a ratio of the initial concentration, normally 21% O_2. This ratio is treated as a frequency of a Gaussian distribution.

An alternative analysis framework was used by McGaughey and Adams (1989) for the application of burner-produced low O_2 atmospheres to small bins. This was not adopted here as we wished our approach to be consistent with the approach used for describing fluid flow through packed beds.

Substantial deviations from this model were to be expected with increasing dispersion, particularly at low O_2 levels. In the limiting case, where complete mixing occurred, the fall in O_2 concentration from an original concentration c_0 to a new value c is an exponential function of time t, following the equation:

$$c = c_0 \, e^{-kt} \tag{1}$$

where k, the rate constant for the decay is given by:

$$k = \frac{Q}{V} \tag{2}$$

Thus

$$\ln c = \ln c_0 - kt = \ln c_0 - \frac{Qt}{V} \tag{3}$$

and consequently the apparent flow rate, Q, into the region of volume, V, can be found from the slope of the plot of ln c with time.

The rate of upward movement of the N_2 atmosphere can be estimated by observing the time taken ($t_{x\%}$) to reach a set O_2 level (x%) with height z in the storage on a given sampling line. The corresponding apparent input rate (Q) through a cross sectional area (A) in a medium with porosity n associated with observations from a given sampling line is then given by:

$$\frac{dz}{dt_{x\%}} = \frac{nQ}{A} \tag{4}$$

Efficiencies (E_3%, Banks, 1979) are calculated on the basis that the most efficient achievable mode of N_2 purging corresponds to true plug-flow in the grain bulk with complete mixing in the headspace. Thus, the theoretical minimum gas usage V_{th} (V_a = volume added, V_B = bulk volume, V_{HS} = headspace volume and C_0 = concentration of O_2 in air, i.e. 21%) is given by:

$$V_{th} = nV_B + V_{HS} \ln \frac{C_0}{c} \tag{5}$$

and thus,

$$E_3\,\% = \frac{100\,V_a}{nV_B + V_{HS} \ln \dfrac{C_0}{c}} \tag{6}$$

EXPERIMENTAL DATA

The data discussed here was obtained from seven trials carried out in Australia in cylindrical grain storage bins at Bordertown, South Australia (twice); Cunningar, New South Wales (twice); Sunshine, Victoria; Newcastle, New South Wales; and Balaklava, South Australia. Details of the trials, bin construction, dimensions and contents are given in Table 1.

TABLE 1
Dimensions and loads of the test bins

	Bin construction		Bin dimensions				Total
Trial	Walls and roof	Floor	Diam. (m)	Height to eaves (m)	Load (t)	Head-space (m^3)	storage capacity (m^3)
Bordertown 1	welded steel	concrete (flat)	23.1	19.6	7035	693	9049
Bordertown 2	welded steel	concrete (flat)	23.1	19.6	5220	2365	9049
Cunningar 1	concrete	concrete (conical)	10.9	30.5	2040	430	2930
Cunningar 2	concrete	concrete (conical)	10.9	30.5	2040	430	2930
Sunshine[a]	welded	concrete (flat)	13.9	19.0	1809[a]	601	3055
Newcastle	bolted steel	concrete (flat)	8.2	9.4	294	72	421
Balaklava	welded steel	concrete (flat)	23.1	19.6	6474	812	9049

[a]This trial carried out on barley, the others on wheat.

Of the structures used, only the Newcastle bin had been constructed specifically for use with fumigants or modified atmospheres. All the other structures required some sealing work (sealing for doors, removal of ventilators or filling of the wall to floor joint) to increase their level of gastightness. The existing fitments had to be modified to take the pipe work for the introduction of N_2. In the Bordertown 1 trial, a special distribution system was constructed for the N_2 purge. This was designed to give an even application of gas across the floor of the bin. All other trials either used the existing ductwork, which had been fitted for aeration, or introduced the gas directly into the bulk through the bin wall via a shielded inlet, as described by Banks and Annis (1977). In all cases a substantial level of gastightness was achieved, with the bins used at Bordertown 2, Sunshine and Balaklava exceeding the standard for bins suitable for N_2 use given by Banks and Annis (1977).

Liquid N_2 was supplied to the site by cryogenic tanker. The liquid was vaporised and brought to ambient temperature with forced draught heat exchangers before being introduced into the test bins. The input flow of N_2 was measured using either a rotameter-type system (Elliot Automation 101 Metric Series) or an Anubar in-line pitot tube system. The total gas usage was calculated both from weighbridge readings and by integration of the flow measurements. Gas volumes are given at average grain temperature and ambient atmospheric pressure for the site. Concentrations are given in % v/v.

Before grain inloading, the bins were rigged with both 3-mm o.d. semi-rigid nylon tubing, for gas sampling, and copper-constantan thermocouples, for temperature measurement. Each bundle of lines, designated below by a capital letter, contained a number of lines terminating at different heights in the storage, giving a 1–3-m separation of sampling points. These points are designated below by height in the bin. Thus, Sunshine A, 4.0 refers to the sampling point on line-bundle A at 4.0 m above the floor in the Sunshine trial. Heights in the Cunningar trials use the base of the cylindrical wall as zero. The distribution of sampling lines for each trial is shown in Fig. 1.

Gas samples were taken with a small diaphragm pump and analysed for O_2 concentration with Bacharach Model K525 or Beckman Model 715 electrolytic oxygen meters.

Grain was loaded into the test bins under normal commercial conditions. However, the bins were not fully filled as this would have tended to overstress the fixings of the cables supporting the sampling lines where they were attached to the roof. The grain surface was raked flat for the Newcastle trial but was left as it fell in the others. Generally, the larger-than-normal headspace in the bins accentuated the deviation from ideal behaviour and reduced the observed purging efficiency.

RESULTS

Movement of the nitrogen atmosphere

Two modes of O_2 removal from positions within the grain bulk, as detailed below, were noted during the trials. The mode followed was decided by the fit of the trend of O_2 concentration with time after one of two transformations.

Mode 1 — The diffuse plug-flow model. With this mode the O_2 level, transformed as a frequency, was an approximately linear function of time for values between 20.8 and 1% O_2 (e.g. for Gaussian frequencies of 0.05 to 0.99).

Mode 2 — The free mixing model. With this mode the O_2 level was found to be a semilogarithmic function of time for values between 20.8 and 1% O_2.

The fall of O_2 concentration was rapid for most points. Often, this fall from >19% O_2 to <3% O_2 occurred within the sampling interval used, and therefore data adequately defining the shape of the O_2 concentration-time curve was not obtained. Where this was so, it has been assumed that the disperse plug-flow model (Mode 1) is appropriate. However, with increasing dispersion, the rate of O_2 displacement lessened, allowing enough observations to be collected for comparison with those expected from the postulated models. In a few cases neither of these models obviously held.

Typical curves obtained for O_2 displacement following Mode 1 are given in Figs. 2 and 3. Examples of the few instances of curves giving behaviour more consistent with Mode 2 are given in Figs. 4 and 5. Figure 5 also shows the most extreme deviation from the ideal plug-flow behaviour observed in any of the trials. In this case a number of points close to the bin wall purged very slowly (10.2, 12.7 and 15.2 m), remaining at >10% O_2 when the remainder of the points in the grain were <1% O_2. The sampling line with the aberrant points was also equidistant from each arm of the Y-shaped gas introduction duct, giving no horizontal pressure gradient to help disperse the O_2.

In Fig. 5 it can be seen that some points give O_2 reductions consistent with Mode 2, albeit with a very slow rate of change. This rate is independent of N_2 introduction rate. A few other lines show behaviour inconsistent with either mode.

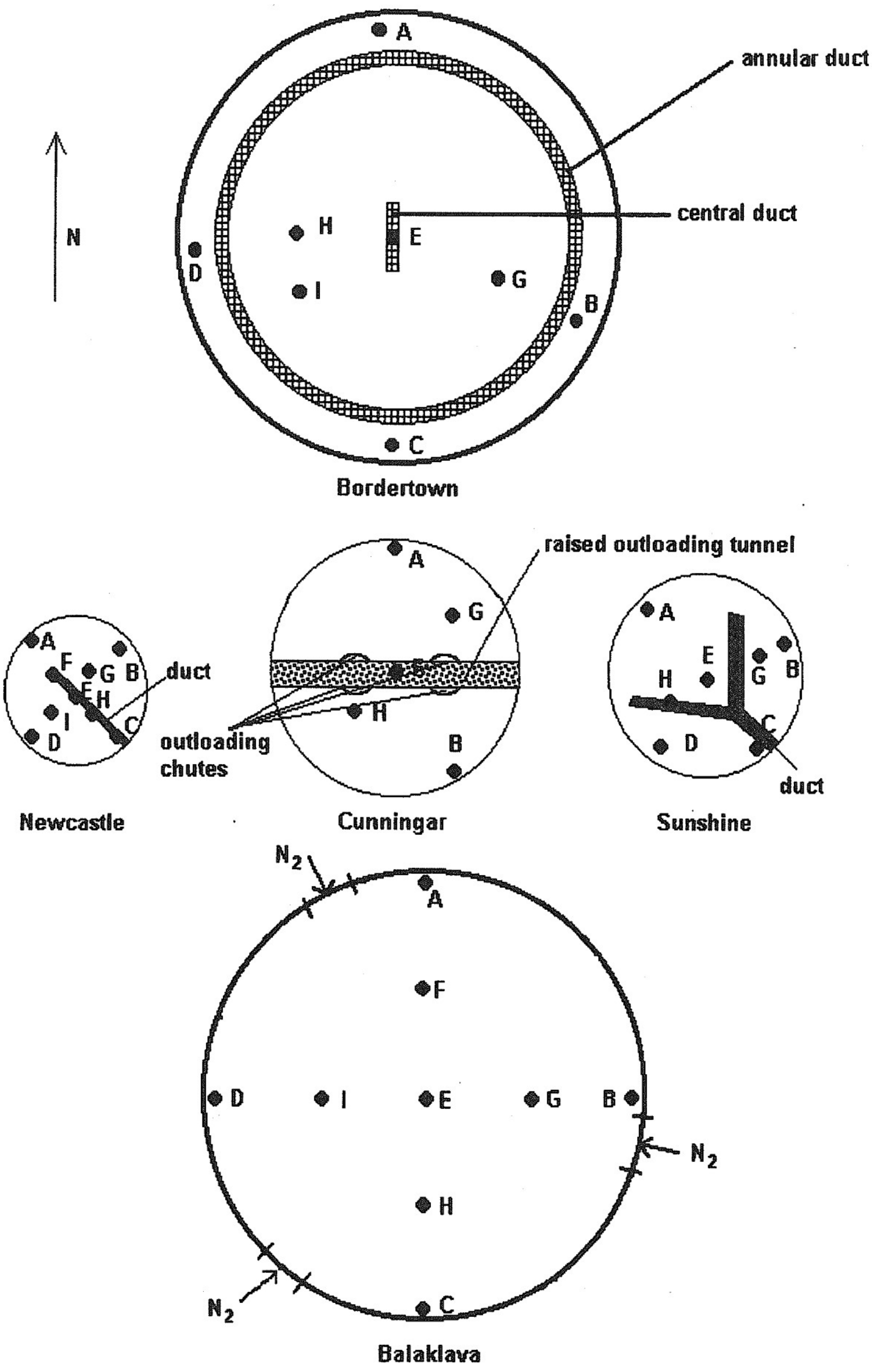

Fig. 1. Distribution of the sampling lines for O_2 analysis.

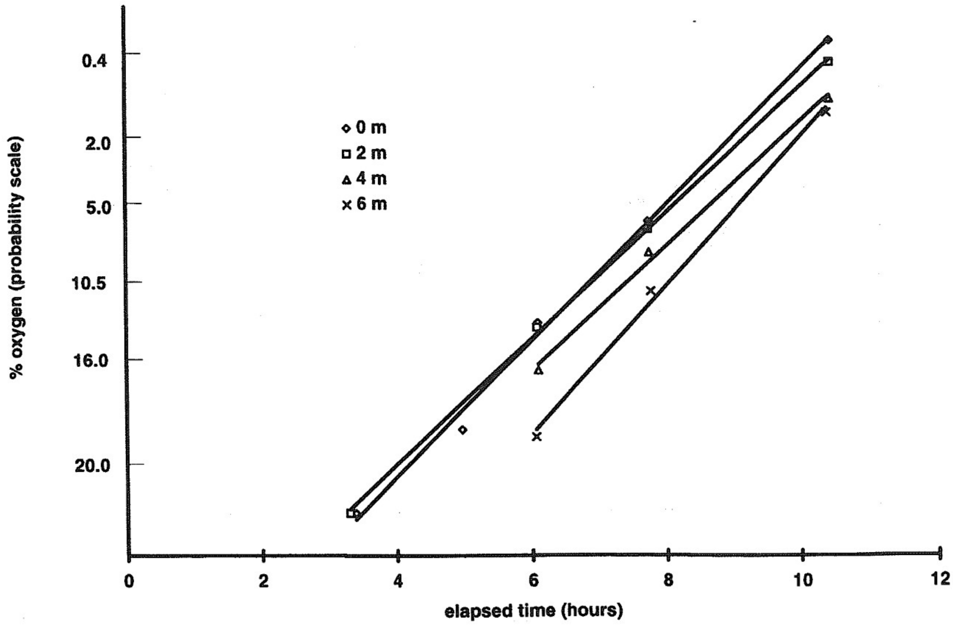

Fig. 2. O_2 concentration fall with time transformed as for Mode 1 in Newcastle trial, line D.

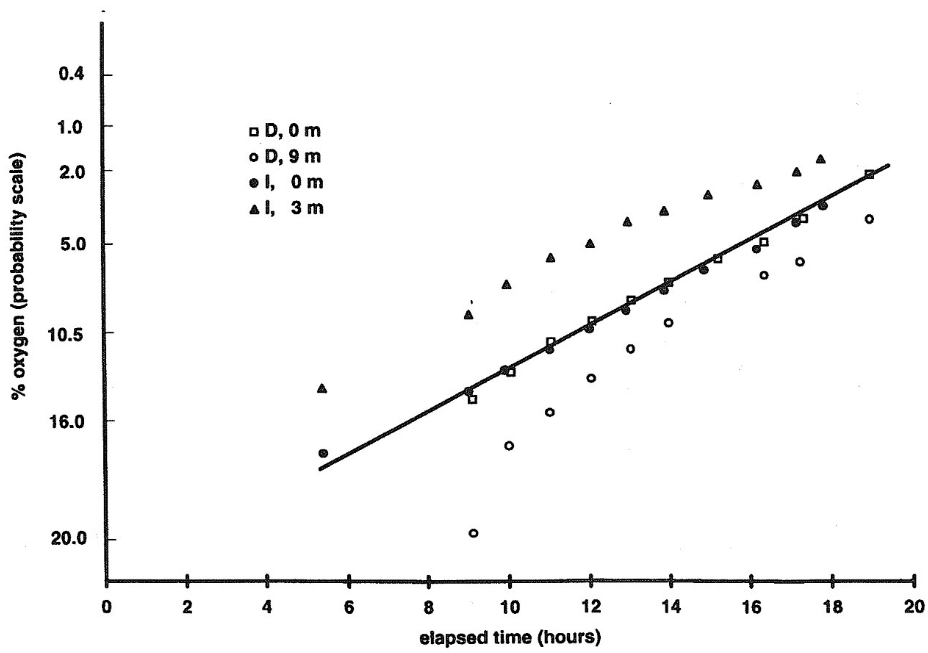

Fig. 3. O_2 concentration fall with time transformed as for Mode 2 for some points in the Balaklava trial.

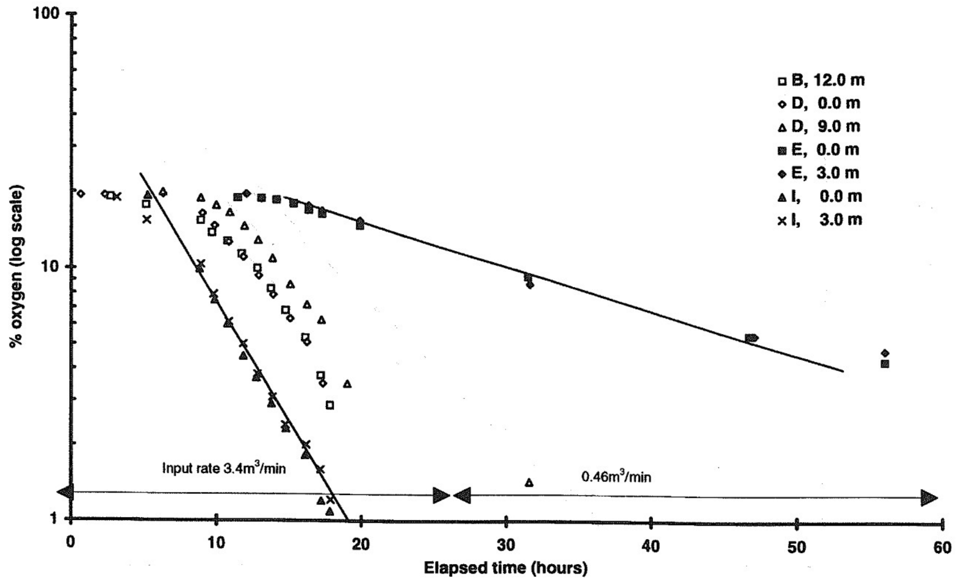

Fig. 4. Variation of O_2 concentration with time for points from the Balaklava trial, transformed as appropriate for Mode 2 and showing variation in N_2 purge rate.

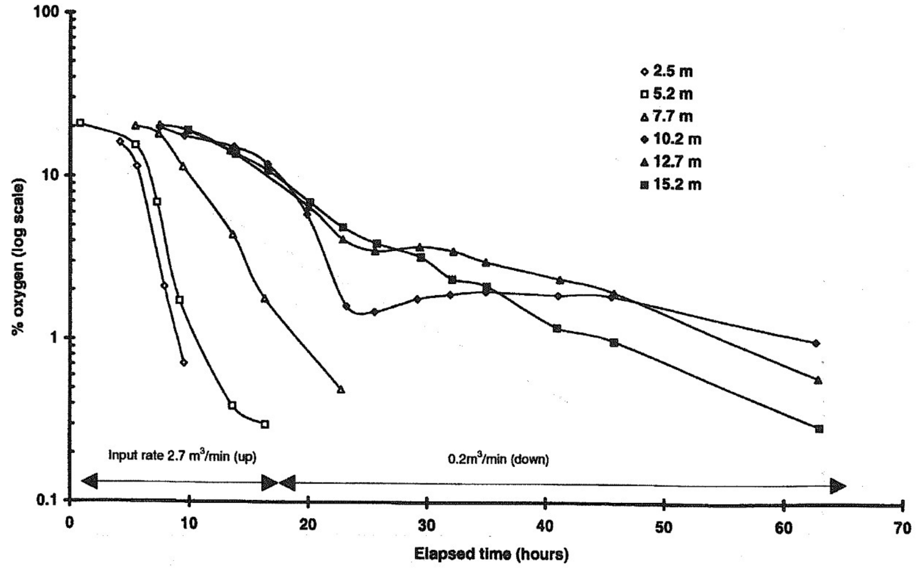

Fig. 5. Variation of O_2 concentration with time for line A, Sunshine trial, transformed as appropriate for Mode 2.

Similar, very slow flow-independent purging, consistent with Mode 2, was observed in one other trial (Fig. 4). Again this appears to be associated with the lack of a horizontal pressure gradient to displace the O_2-containing atmosphere. In this case, the points of concern are in the centre of the bin, on the floor at points equidistant from the N_2 inputs and at three points equally spaced around the bin wall. The modes of O_2 removal for all points sampled in the grain bulks are summarised in Table 2.

Even amongst points that conform to the relatively efficient Mode 1, there is a range of slopes that apparently correspond to different rise rates of the N_2 in different parts of the bin (i.e. the purging front is not level). Table 3 shows face velocities and

TABLE 2
Modes of purging for 193 individual points in seven trials

Number of points	Mode of purging
164	Mode 1 — disperse plug flow
24	Mode 2 — free mixing
5	Mode undefined — neither model holds well

TABLE 3
Nitrogen face velocities and corresponding equivalent gas flow rates for the sampling lines with displacement consistent with Mode 1

Trial	Line	Observed rise rate ($m\ h^{-1}$)	Equivalent input rate ($m^3\ min^{-1}$)	Actual average input rate ($m^3\ min^{-1}$)
Cunningar 1	A	2.14	1.26	2.2
	B	1.66	0.98	
	E	4.44	2.62	
	G	1.85	1.09	
	H	2.27	1.34	
Cunningar 2	A	3.65	2.16	
	B	2.85	1.68	
	E	9.12	5.39	3.2
	G	4.82	2.85	
	H	7.35	4.34	
Sunshine	A	<0.46	<0.58	
	B	2.23	2.82	
	C	2.36	2.98	
	D	2.13	2.69	2.7
	E	10.00	12.65	
	G	6.50	8.22	
	H	10.30	13.02	

Cunningar trials are with wheat, Sunshine trial with barley, hence the different conversion factor for rise rate to equivalent input rate (porosities 38 and 50%, respectively).

equivalent purging rates from several trials. There can be a substantial difference in rise rates in various parts of the bin. It can be seen that within individual trials there was a wide variation in rate for different lines. There is clearly scope for improving efficiency by better application and gas distribution systems. Whether improvement is justified, economically, depends on the balance between the costs of the improvement and the savings in N_2. In all the trials discussed here, or reported in the scientific literature to date, the target level of O_2 has been achievable with the duct-work as used.

Removal of oxygen from the headspace

As expected, in all trials the loss of O_2 from the headspace conformed with Mode 2 (displacement by free mixing). Changes in the rate of gas input were directly reflected in changes in the rate of headspace pull down (Figs. 6 and 7).

Given the volume of the headspace, the flow rate of N_2 entering the headspace can be calculated from the slope of the semilogarithmic decay curve. Table 4 gives a comparison of the calculated rate derived from the rate of loss of O_2 from the headspace with the average input rate. With one exception, the calculated rate was always less than that observed. This can be ascribed either to incomplete mixing and venting of O_2-poor gas or to the entry into the headspace of gas containing significant O_2 amounts rather than pure N_2. The latter possibility is supported by the observed breakthrough times (Table 5). In all but one case, the beginning of the loss of O_2 concentration in the headspace (breakthrough) was observed earlier than was calculated on pure plug-flow bands. In the single exception, Cunningar 2, the calculated and observed breakthrough times were the same. Moreover, the calculated and actual input rates were very similar.

The close correspondence with a free mixing model for O_2 pull down in the headspace makes the extrapolation to target O_2 levels quite reliable. Thus it is possible, given an observed O_2 level of, say, 3% in the headspace, to predict with a high degree of certainty the extra N_2 addition needed to bring the level to 1%. With the few exceptions noted above, the bulk in these trials was already purged to about the target level before the N_2 was added to the headspace. This extrapolation can therefore be used with confidence to estimate the total N_2 usage and purge times needed to create the target atmospheres.

Purging efficiencies

Table 5 gives observed or extrapolated purging data for the systems to reach 1% O_2 throughout in the six trials. Under these conditions, with larger than normal headspaces, N_2 usage was between 1.2 and 2.0 $m^3\ t^{-1}$, with efficiencies E_3 of 73–92%. Other trials, for example Cassells *et al.* (1994) and Banks *et al.* (1980a), have shown similar purge-consumptions and efficiencies.

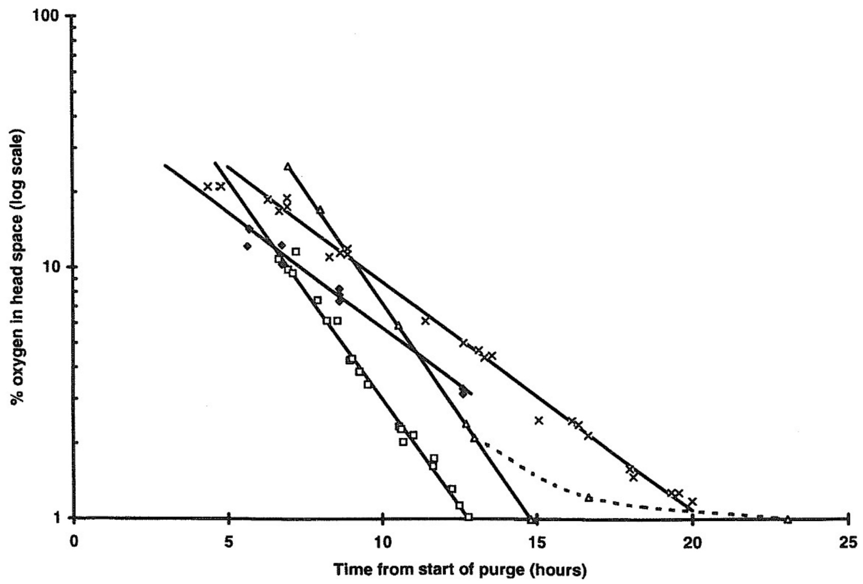

Fig. 6. O_2 concentration decay in the headspace during purging for Cunningar 2 (□); Bordertown 1 (Δ), displaced by + 2 h); Bordertown 2 (♦) and Sunshine (×) trials.

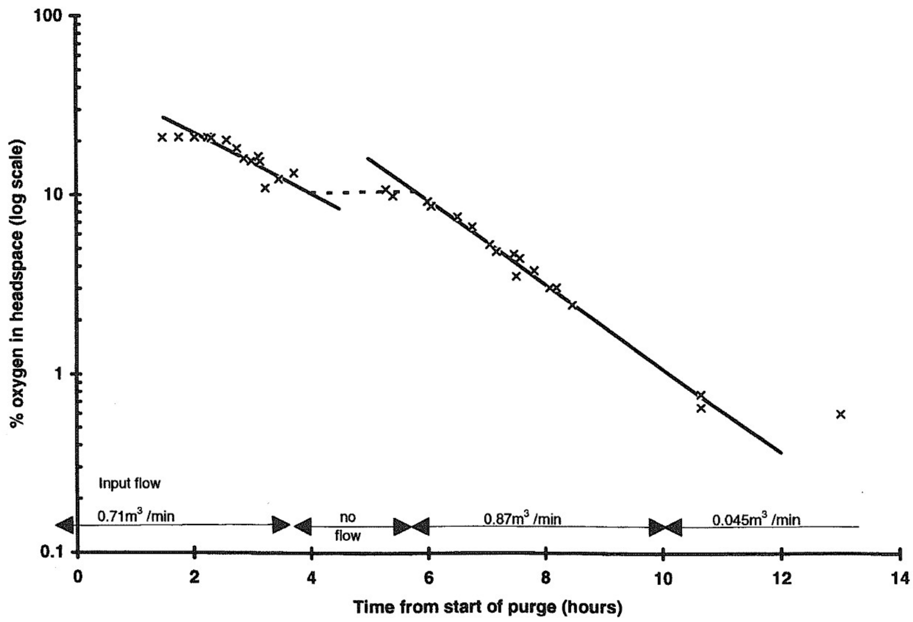

Fig. 7. O_2 concentration decay in the headspace during purging in the Newcastle trial, showing changes in N_2 input rate.

TABLE 4
Efficiency of purging and N_2 usage

Trial	Filling ratio	Headspace O_2% at end of purge	Efficiency of purging (to 1%)	Time required to give 1% O_2 in headspace (h)	N_2 usage calculated for this period (m^3)	N_2 usage ($m^3\ t^{-1}$)
Bordertown 1	0.92	1.1	76	16.2[a]	9170	1.30
Bordertown 2	0.74	3.2	73	18.4[a]	10390	1.99
Cunningar 1	0.85	3.9	84	20.5[a]	3470	1.70
Cunningar 2	0.85	1.0	92	12.8	2430	1.19
Sunshine[b]	0.71	1.2	89	20.4	3340	1.85
Newcastle	0.83	0.7	88	10.2	430	1.46
Balaklava	0.91	2.1	75	141.8[a]	9150	1.41

[a]Extrapolated time on the basis of a semilogarithmic O_2 decay curve. [b]This trial with barley, all others with wheat.

TABLE 5
Comparison of N_2 input flows and usages to breakthrough from regions in the headspace applied flows and intergranular volumes

Trial	Average input rate ($m^3\ min^{-1}$)	Calculated[a] rate ($m^3\ min^{-1}$)	Expected break-through time assuming plug flow (h)	Observed break-through time (h)	Actual N_2 usage to break-through (m^3)	Inter-granular volume[d] (m^3)
Bordertown 1	4.7[b]	5.1	12.5	5.6	1580[b]	3515
Bordertown 2	9.4	8.3	4.6	3.9	2200	2610
Cunningar 1	1.9[c], 4.7	0.57, 2.40	8.3	5.6	803	950
Cunningar 2	3.1	2.9	5.1	5.1	979	950
Sunshine[c]	2.7	2.1	7.6	6.1	988	1227
Newcastle	0.71[c], 0.87	0.47, 0.65	3.1	2.2	92	132
Balaklava	3.4[c], 0.46	1.2, 0.16	16.9	7.4	1541	3459

[a]Calculated from rate of O_2 depletion in headspace. [b]Flow rate measurement approximate only. [c]Flow rate changed during course of purge. [d]Volume without headspace.

CONCLUSIONS

With N_2 added rapidly to the base of a bin, as here, most regions of a grain bulk will purge by disperse plug-flow.

The headspace will lose O_2 by a process giving a semilogarithmic fall with time, though at a slightly lower rate than that predicted by a simple free-mixing model.

With rudimentary gas-introduction duct work, such as that used in these trials, the N_2 purging front is not horizontal. Some areas of the bulk may purge much more rapidly than others.

In a few extreme cases, apparently associated with low horizontal pressure gradients, some regions in some bulks were observed to lose O_2 only very slowly, and that by a process consistent with free mixing rather than disperse plug-flow. This behaviour may be rate-limiting with regard to achieving the target atmosphere throughout a system.

Even with rudimentary gas introduction systems, purging, 70% on the basis of plug-flow in the bulk and free mixing in the headspace, is quite efficient.

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